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Article

Considering a Non-Constant Anisotropy Parameter in the Giesekus Model

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Abstract

The Giesekus model has proven to be one of the most successful constitutive rheological models. Although Giesekus introduced the anisotropy parameter as a constant, recent evidence suggests that it should not be. We herein elaborate on the implications of having a variable anisotropy coefficient which, to our knowledge, has not been considered before. We find that the modification leads to important differences in the predictions of the second normal stress coefficient in simple shear flow, of which the most significant is the shift of the linear viscoelastic envelope of the second normal stress coefficient to higher values in the case of startup simple shear flow, which is more in line with literature experimental data.

Keywords: rheological model; polymer melts; Giesekus model; mobility tensor; friction tensor; Curtiss-Bird model; link tension coefficient

1. Introduction

The use of plastic in almost every aspect of our everyday lives positions the plastic industry as one of the most important industries. Thus, the optimization of processes producing polymeric materials is of the utmost importance to avoid process instabilities [1,2]. Such a task necessitates the execution of numerical simulations that at heart employ an accurate and reliable constitutive rheological model that is based on molecular principles [2]. The Giesekus model is widely regarded as one of the most successful constitutive equations for polymer solutions and melts, as it can predict a nonzero second normal stress difference, stress overshoot at the onset of simple shear flow, and finite extensional viscosity [3–7]. It introduces the notion that in sufficiently concentrated solutions and polymer melts, the frictional properties should be anisotropic [8,9], controlled by the magnitude of the anisotropy or Giesekus parameter, α . By using the phase-space theory for concentrated polymer solutions and polymer melts of Curtiss and Bird [10–12], Giesekus [13] showed that his anisotropy parameter can be related to the so-called link tension coefficient ε via $\alpha = (1 - \varepsilon)/(2 + \varepsilon)$. Stephanou et al. [14,15] provided the theoretical basis indicating that the link tension coefficient should not be considered a constant but should depend on the nematic order parameter. The revised framework predicts a vanishing ε at equilibrium, which remedies the erroneous predictions of the Curtiss-Bird model when considering a constant ε , namely the approach of the transient shear viscosity as $t \rightarrow 0$ to a constant value and the spurious sign changes of the transient second normal stress coefficient in startup simple shear flow.

Given the modification proposed by Stephanou et al. [14,15], in this work, we herein revisit the original Giesekus model by considering a non-constant anisotropy parameter following the relationship between α and ε , and the relation between the former and the nematic order parameter as proposed by Stephanou et al. [14,15]. As such, at equilibrium, the anisotropy parameter is equal to $\frac{1}{2}$. We elaborate on the implications on the modified Giesekus predictions when considering a

variable anisotropy parameter and discuss whether its predictions are more in line with rheological experimental data. In Sec. 2, we provide the original and modified Giesekus constitutive model, and proceed to Sec. 3, where we present a comparison between their predictions. We conclude in Sec. 4 where the most important outcomes of our work are summarized and where we provide our future plans.

2. Constitutive Model

We consider an isothermal and incompressible flow. The Giesekus model in this case reads as follows [16]:

$$\dot{c}_{\alpha\beta,[1]} = -\frac{1}{\tau_R} \left[\frac{1}{2} (c_{\alpha\gamma} \beta_{\beta\gamma} + c_{\beta\gamma} \beta_{\alpha\gamma}) - \beta_{\alpha\beta} \right]. \quad (1a)$$

where $\mathbf{c}$ is the dimensionless conformation tensor and we have defined the upper-convected time derivative:

$$\dot{c}_{\alpha\beta,[1]} \equiv \frac{\partial c_{\alpha\beta}}{\partial t} + u_\gamma \nabla_\gamma c_{\alpha\beta} - c_{\alpha\gamma} \nabla_\gamma u_\beta - c_{\gamma\beta} \nabla_\gamma u_\alpha. \quad (1b)$$

For the dimensionless mobility tensor, we consider the linear expression considered by Giesekus [13,16], namely,

$$\beta_{\alpha\beta} = \delta_{\alpha\beta} + \alpha (c_{\alpha\beta} - \delta_{\alpha\beta}). \quad (1c)$$

where α is the anisotropy or Giesekus parameter. Also, the stress tensor is given as,

$$\sigma_{\alpha\beta} = G (c_{\alpha\beta} - \delta_{\alpha\beta}). \quad (2)$$

In this work, which we believe to be the first such work, we consider the Giesekus parameter to be given explicitly by the expression relating it to the link tension coefficient,

$$\begin{aligned} \alpha &= \frac{1-\varepsilon}{2+\varepsilon}, \\ \varepsilon &= \varepsilon_0 S_2^2 \end{aligned} \quad (3a)$$

where ε_0 is a constant coefficient and

$$S_2^2 = \frac{3}{2} \left[\text{tr} \left(\langle \mathbf{u}\mathbf{u} \rangle_{\text{an}} \cdot \langle \mathbf{u}\mathbf{u} \rangle_{\text{an}} \right) \right], \quad (3b)$$

is the nematic order parameter [17] which depends on the anisotropic orientation tensor $\langle \mathbf{u}\mathbf{u} \rangle_{\text{an}}$ defined as [14,15]

$$\langle \mathbf{u}\mathbf{u} \rangle_{\text{an}} \equiv \langle \mathbf{u}\mathbf{u} \rangle - \frac{1}{3} \mathbf{I}. \quad (3c)$$

where $\mathbf{I}$ is the unit tensor and $\mathbf{u}$ the unit end-to-end vector of polymer chains. Note that the link tension coefficient is not constant: it vanishes at equilibrium, it increases with time in startup flows, and its steady-state value increases with the imposed flow rate [14]. By considering $\langle \mathbf{u}\mathbf{u} \rangle = \mathbf{c}/\text{tr} \mathbf{c}$, then

$$S_2^2 = \frac{3}{2} \left[\frac{\text{tr} \mathbf{c}^2}{(\text{tr} \mathbf{c})^2} - \frac{1}{3} \right]. \quad (3d)$$

As a final note, since the order parameter asymptotes at large shear rate to unity, the Giesekus parameter approaches the value $\alpha = (1 - \varepsilon_0)/(2 + \varepsilon_0)$; since thermodynamic admissibility dictates that the Giesekus parameter should be $0 \leq \alpha \leq 1$ [16,18–20] we need to bound $\varepsilon_0 \leq 1$. Finally, note that when $\alpha=0$, the Giesekus model simplifies to the Upper-Convected Maxwell (UCM) one.

Asymptotic Behavior of the Model for Steady-State and Transient Shear Flow

We then analyze the asymptotic behavior of the two versions of the Giesekus model in the case of a steady and startup simple shear flow (SSF), described by the kinematics $\mathbf{u} = (\dot{\gamma}y, 0, 0)$ where y denotes the Cartesian coordinate and $\dot{\gamma}$ the shear rate. We present the following material functions: the shear viscosity $\eta = \sigma_{yx}/\dot{\gamma}$ and the first and second normal stress coefficients, $\Psi_1 = (\sigma_{xx} - \sigma_{yy})/\dot{\gamma}^2$ and $\Psi_2 = (\sigma_{yy} - \sigma_{zz})/\dot{\gamma}^2$, respectively. The asymptotic behaviour is the same as the original Giesekus model, minus the fact that in the modified model that at equilibrium $\alpha=1/2$ irrespective of the choice of ε_0 . Thus, for the original model

$$\begin{aligned} c_{xx} &= 1 + (2 - \alpha) \text{Wi}^2 \\ c_{xy} &= \text{Wi} \\ c_{yy} &= 1 - \alpha \text{Wi}^2 \\ c_{zz} &= 1 \end{aligned} \quad , \quad (4a)$$

whereas for the modified model,

$$\begin{aligned} c_{xx} &= 1 + \frac{3}{2} \text{Wi}^2 \\ c_{xy} &= \text{Wi} \\ c_{yy} &= 1 - \frac{1}{2} \text{Wi}^2 \\ c_{zz} &= 1 \end{aligned} \quad . \quad (4b)$$

We thus obtain the zero-shear-rate material functions: for the original model

$$\begin{aligned} \eta_0 &= G\tau_R \\ \Psi_{1,0} &= 2\eta_0\tau_R \\ -\Psi_{2,0} &= \alpha\eta_0\tau_R \end{aligned} \quad , \quad (5a)$$

and for the modified model,

$$\begin{aligned} \eta_0 &= G\tau_R \\ \Psi_{1,0} &= 2\eta_0\tau_R \\ -\Psi_{2,0} &= \frac{1}{2}\eta_0\tau_R \end{aligned} \quad . \quad (5b)$$

Upon inception of the simple shear flow, we follow the methodology of Stephanou et al. [20] to obtain the explicit solutions for the time-dependent viscometric functions in the linear viscoelastic (LVE) limit. The results are the same as those of the original Giesekus model,

$$\begin{aligned}
\eta^+(t) &= \eta_0 \left[1 - \exp\left(-\frac{t}{\tau_R}\right) \right] \\
\Psi_1^+(t) &= \Psi_{1,0} \left[1 - \left(1 + \frac{t}{\tau_R}\right) \exp\left(-\frac{t}{\tau_R}\right) \right] \\
\Psi_2^+(t) &= \alpha \eta_0 \tau_R \left[1 - \frac{2t}{\tau_R} \exp\left(-\frac{t}{\tau_R}\right) - \exp\left(-\frac{2t}{\tau_R}\right) \right]
\end{aligned} \tag{6a}$$

except for the fact that the LVE envelope of the second normal stress coefficient for the modified version differs,

$$\Psi_2^+(t) = \frac{1}{2} \eta_0 \tau_R \left[1 - \frac{2t}{\tau_R} \exp\left(-\frac{t}{\tau_R}\right) - \exp\left(-\frac{2t}{\tau_R}\right) \right]. \tag{6b}$$

3. Results and Discussion

3.1. Model Predictions in Steady-State Shear Flow

Figure 1 illustrates the variations in the conformation tensor components as functions of the dimensionless shear rate (Wi) in the case of steady-state SSF when the original Giesekus model is compared with the modified model at two different values of $\mathcal{E}$ or $\mathcal{E}_0$ (0.1 and 0.7). The behavior of c_{xx} as a function of Wi is the same for both models when small values of $\mathcal{E}$ or $\mathcal{E}_0$ ($=0.1$) are used. Similarly, as expected for small Wi ($Wi < 0.1$), c_{xx} remains close to its equilibrium value of unity in both models and then increases as Wi increases. This is expected since in the linear regime the mobility tensor is equal to the unit tensor. However, when $\mathcal{E}$ or $\mathcal{E}_0$ is raised to 0.7, we note that the two models provide different predictions in the intermediate range ($0.1 < Wi < 100$) with the modified model consistently predicting lower c_{xx} values than the original model. However, at large shear rates, the two models agree again, since in this case $S_2^2 \approx 1$ and due to Eq. (3a) $\mathcal{E} \approx \mathcal{E}_0$. A similar behavior is noted also for c_{xy} [Figure 1(b)] and c_{yy} [Figure 1(c)]: in both cases, the predictions from the two models are the same at small and large shear rates, whereas the prediction of the modified model is below the original model's one in the intermediate range. The scaling at large shear rates is the same for both models: c_{xx} increases as $Wi^{\frac{1}{2}}$, c_{xx} reaches an asymptotic value, and c_{yy} decreases as $Wi^{-\frac{1}{2}}$. Then, Figure 2 shows the variations of the material viscometric functions as a function of Wi for both the modified and original models. For the shear viscosity and the first normal stress coefficient, we note a similar behavior as for the conformation tensor: the two models provide similar predictions for η and Ψ_1 for small and large shear rates, but the prediction of the modified model is below that of the original model predictions in the intermediate range. On the contrary, the behavior in the scaled second normal stress coefficient, $-\Psi_2 / (\eta_0 \tau_R)$, is quite different [Figure 2(c)]: the two models provide a similar prediction for a small value of $\mathcal{E}$ or $\mathcal{E}_0$ ($=0.1$), with a similar zero-shear rate value (equal to 9/21 for the original model and $\frac{1}{2}$ for the modified model) as dictated by Eqs. (5), and the scaling at large shear rates is $\eta \propto Wi^{-1}$, $\Psi_1 \propto Wi^{-\frac{3}{2}}$, and $\Psi_2 \propto Wi^{-2}$ (Table 7.3-5 of Bird et al. [6]); however, they differ significantly at low and intermediate shear rates at larger values of $\mathcal{E}$ or $\mathcal{E}_0$ ($=0.7$): the original Giesekus model predicts $-\Psi_2 / (\eta_0 \tau_R)$ to be equal to 1/9, whereas the modified version always predicts it to be equal to $\frac{1}{2}$. This also has implications to the startup behavior as will be described below.

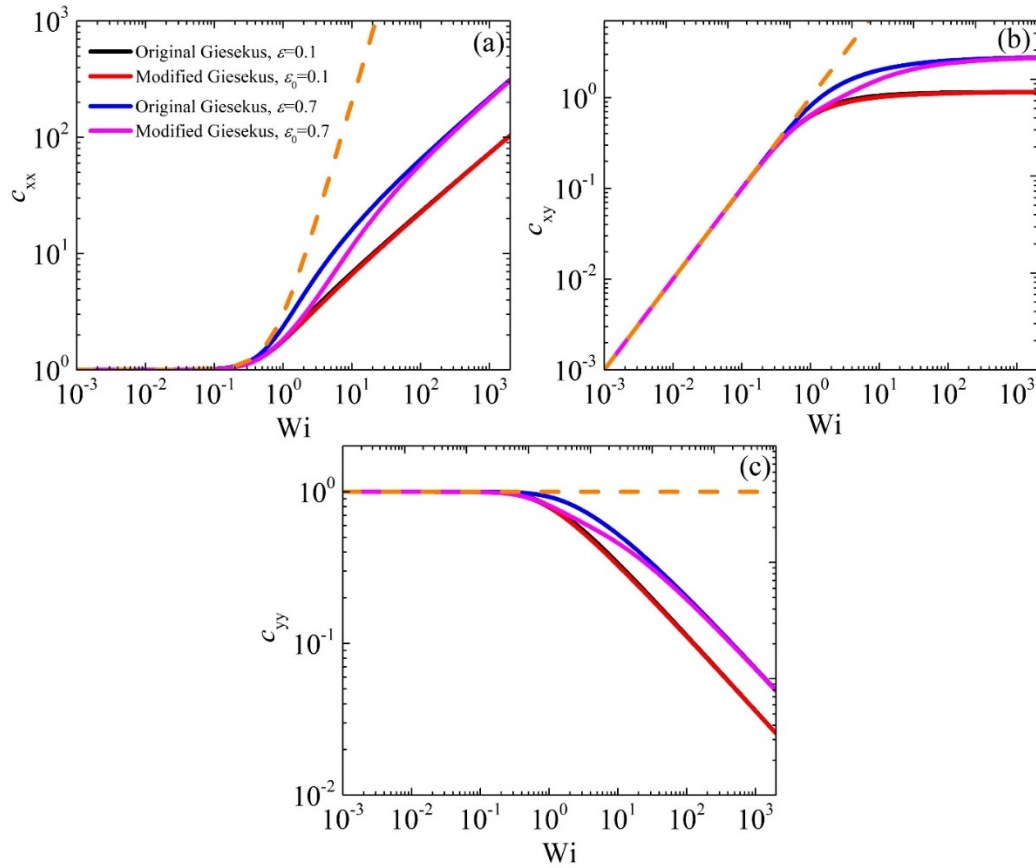


Figure 1. Model predictions for the conformation tensor components (a) c_{xx} , (b) c_{xy} , (c) c_{yy} , in steady SSF as a function of the dimensionless shear rate, and dependency on the parameter α for the original model, and ε_0 for the modified model. The dotted lines depict the predictions of the UCM model (with $\varepsilon=0$).

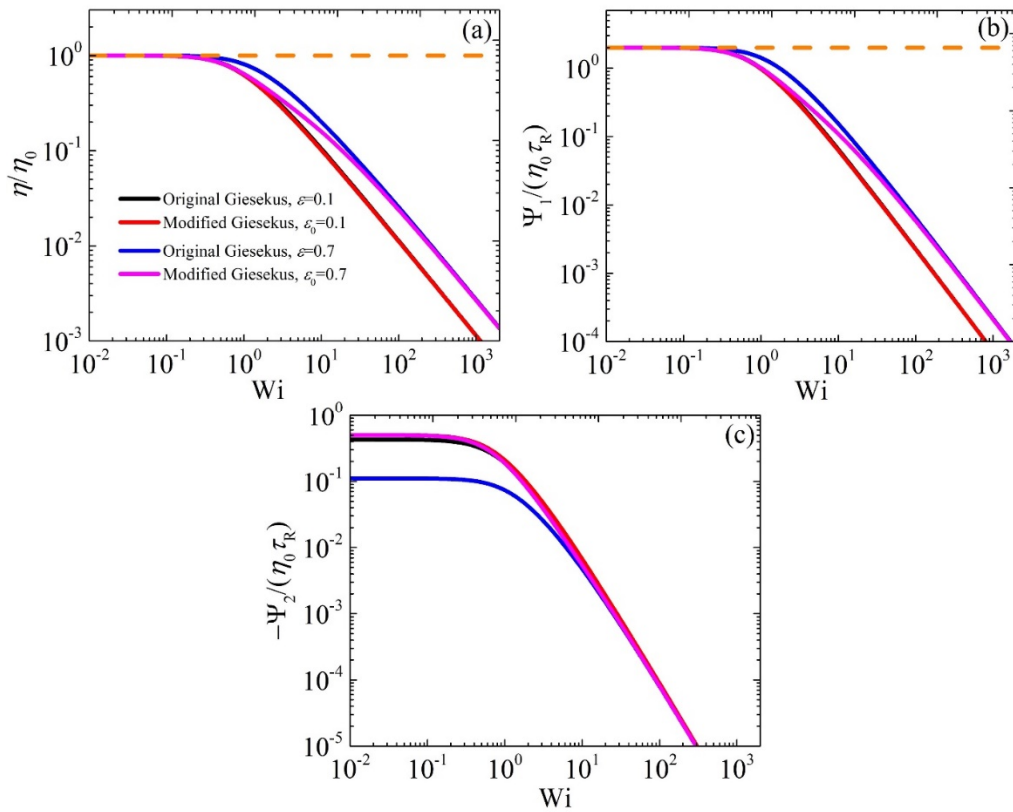


Figure 2. Same as Figure 1 but for the viscometric functions.

3.2. Model Predictions in Steady-State Uniaxial Elongational Flow

In the case of uniaxial elongational flow (UEF), described by the kinematics $\mathbf{u} = (\dot{\mathcal{E}}x, -\frac{1}{2}\dot{\mathcal{E}}y, -\frac{1}{2}\dot{\mathcal{E}}z)$, where $\dot{\mathcal{E}}$ is the elongation rate (x is the flow direction, y is the velocity gradient direction, and z is the neutral direction), the material function to be analyzed is the elongational viscosity, $\eta_E = (\sigma_{xx} - \sigma_{yy})/\dot{\mathcal{E}}$. A simple analytical solution can easily be obtained:

$$\begin{aligned} c_{xx} &= \frac{1}{2\alpha} \left\{ \sqrt{1 + 4Wi^2 - 4\alpha(1-2\alpha)Wi} - (1 - 2\alpha - 2Wi) \right\} \\ c_{yy} &= \frac{1}{2\alpha} \left\{ \sqrt{1 + Wi^2 + 4\alpha(1-2\alpha)Wi} - (1 - 2\alpha + Wi) \right\} \end{aligned} \quad (7a)$$

so that

$$\frac{\eta_E}{\eta_0} = \frac{1}{2\alpha} \left\{ 3 + \frac{1}{Wi} \sqrt{1 + 4Wi^2 - 4\alpha(1-2\alpha)Wi} - \sqrt{1 + Wi^2 + 4\alpha(1-2\alpha)Wi} \right\}. \quad (7b)$$

The result Eq. (7b) is well-known (see Table 7.3-5 with $\lambda_2=0$ in Bird et al. [6]). In the case of the original Giesekus model, the Giesekus parameter is a constant, whereas in the modified version it is given from Eq. (3a). In both models, the elongation viscosity reaches the asymptote $\eta_E/\eta_0 = 2/\alpha$ as we may easily note from Eq. (7b) (also see Eq. (7B.8-4) of Bird et al. [6]).

Figure 3 shows the conformation tensor elements c_{xx} , c_{yy} , and the dimensionless elongation viscosity (η_E/η_0) versus Wi . In Figure 3(a), c_{xx} starts from its equilibrium value of unity at low Wi for both models, and then increases with Wi , with higher values as $\mathcal{E}$ or $\mathcal{E}_0$ increases to 0.7. Similarly, in Figure 3(b), c_{yy} also starts from its equilibrium value of unity at low Wi for both models, but then decreases as the shear rate increases, exhibiting higher values as $\mathcal{E}$ or $\mathcal{E}_0$ increases. The elongation viscosity starts from the value of 3, as per Trouton's law, and increases as the elongation rate increases, exhibiting smaller values as $\mathcal{E}$ or $\mathcal{E}_0$ increases [Figure 3(c)]. When $\mathcal{E}_0 = 0.1$, both models provide similar predictions for both c_{xx} and c_{yy} , whereas the prediction of the modified model is below the prediction of the original model for c_{xx} ; on the contrary, it seems that the prediction for c_{yy} is the same for both models, even with a larger value of $\mathcal{E}$ or $\mathcal{E}_0$. The predictions for η_E/η_0 are noted to be the same for both models at large values of $\mathcal{E}$ or $\mathcal{E}_0$ whereas at smaller values, the modified version is below the original Giesekus model's prediction in the intermediate range of elongation rates. The scaling at large shear rates is the same for both models, namely $\tilde{c}_{xx} \propto Wi$ and $\tilde{c}_{yy} \propto Wi^{-1}$, whereas, as mentioned above, the elongation viscosity always reaches the asymptote $\eta_E/\eta_0 = 2/\alpha$.

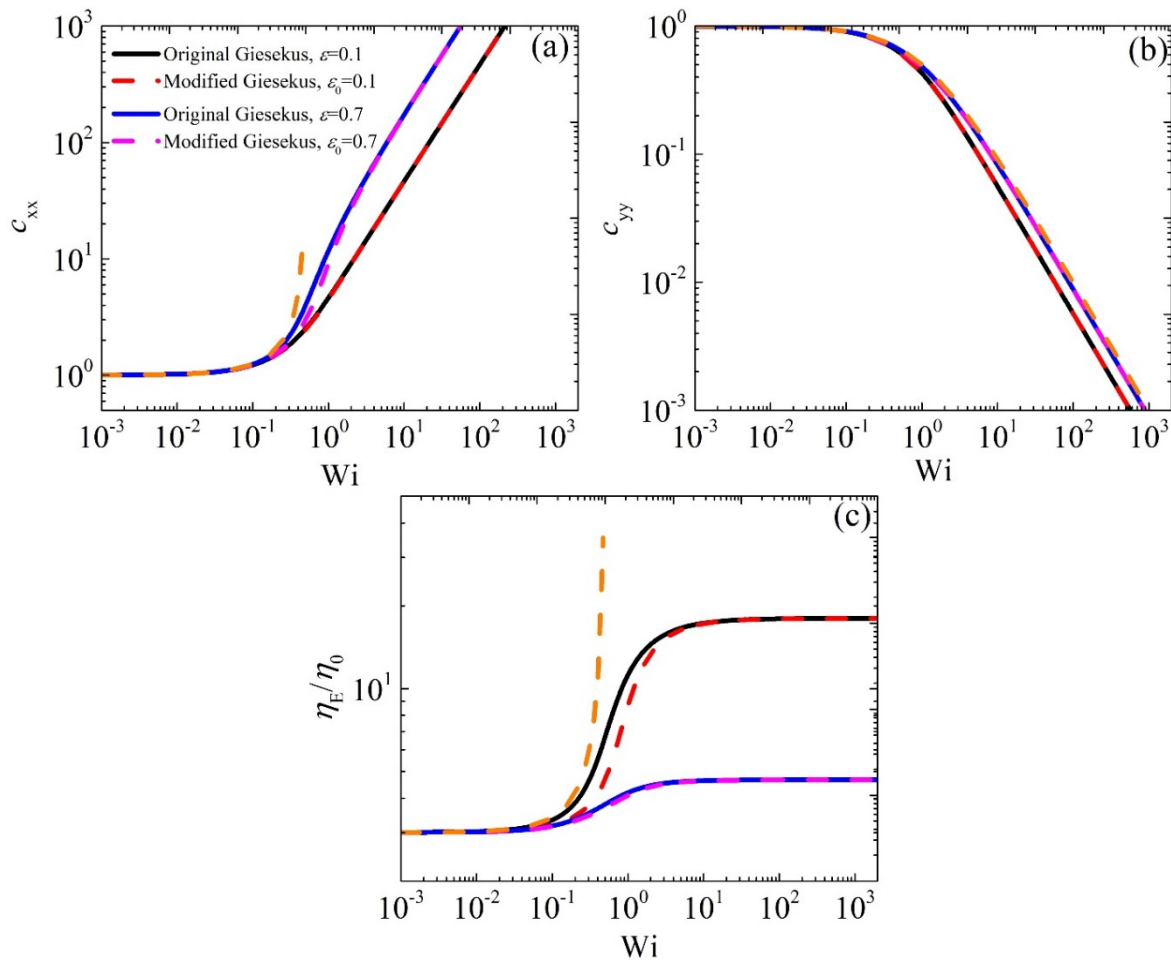


Figure 3. Model predictions for the conformation tensor components (a) c_{xx} , and (b) c_{yy} , and (c) the scaled elongational viscosity η_E in steady UEF as a function of the dimensionless elongation rate, and dependency on the parameter α for the original model and ϵ_0 for the modified model. The dotted lines depict the predictions of the UCM model (with $\epsilon=0$).

3.3. Model Predictions in Start-Up Shear Flow

Figures 4 and 5 show the dimensionless material functions η^+/η_0 , $\Psi_1^+/(\eta_0\tau_R)$, and $-\Psi_2^+/(\eta_0\tau_R)$ for startup SSF as a function of dimensionless time for $\epsilon_0 = 0.1$ and $\epsilon_0 = 0.7$, respectively, at $Wi = 1, 10$, and 100 . In Figures 4(a) and 4(b), the predictions of η^+/η_0 and $\Psi_1^+/(\eta_0\tau_R)$ for both models exhibit similar overall trends for all Wi : they increase at early times following the LVE envelope, Eqs. (6), reach an overshoot and then decrease, before reaching their steady state value. Also, both models' predictions the shear viscosity present an undershoot after the overshoot, which is more easily noticeable at $Wi=100$. This oscillatory behavior for the original Giesekus model was first noted by Giesekus [8]. Such undershoots, first provided by the experimental work of Costanzo *et al.* [21], has been associated with the tumbling of polymer chains in simple shear [22,23]. When ϵ or ϵ_0 is small ($=0.1$), the predictions of the original and modified Giesekus models are identical, as seen in Figure 4, whereas at a larger value ($=0.7$), there are clear differences between the original and modified models (Figure 5). In particular, the predictions of the modified Giesekus model for η^+/η_0 and $\Psi_1^+/(\eta_0\tau_R)$ are noted to be below the original Giesekus model predictions at large times for intermediate shear rates ($Wi=1$ and 10), whereas the predictions are similar at larger shear rates irrespective of time. On the other hand, the predictions of the two

models for $-\Psi_2^+ / (\eta_0 \tau_R)$ are noted to be completely different even in the LVE regime, as indicated by the different expressions for the LVE envelope [see Eqs. (6)]. The modified model is seen to shift upwards following Eq. (6b) and is noted to predict a larger value than the original model at small shear rates ($Wi=1$). However, at larger shear rates ($Wi=10$ and 100), both models exhibit the same steady-state value. This behavior is more in line with experimental data [14]; as seen in Figure 8(b) of Stephanou et al. [20], the LVE envelope predicted by the original Giesekus model (note that the LVE envelope provided there is not exactly the same as the original Giesekus model provided here; however, due to the very small value of the slip parameter considered in their work, $\xi=0.03$, their LVE envelope should not differ substantially from the original Giesekus model one) is below the LVE envelope suggested by the experimental data. Thus, the use of a variable Giesekus parameter, as done in the modified model, would certainly improve the comparison against said experimental data.

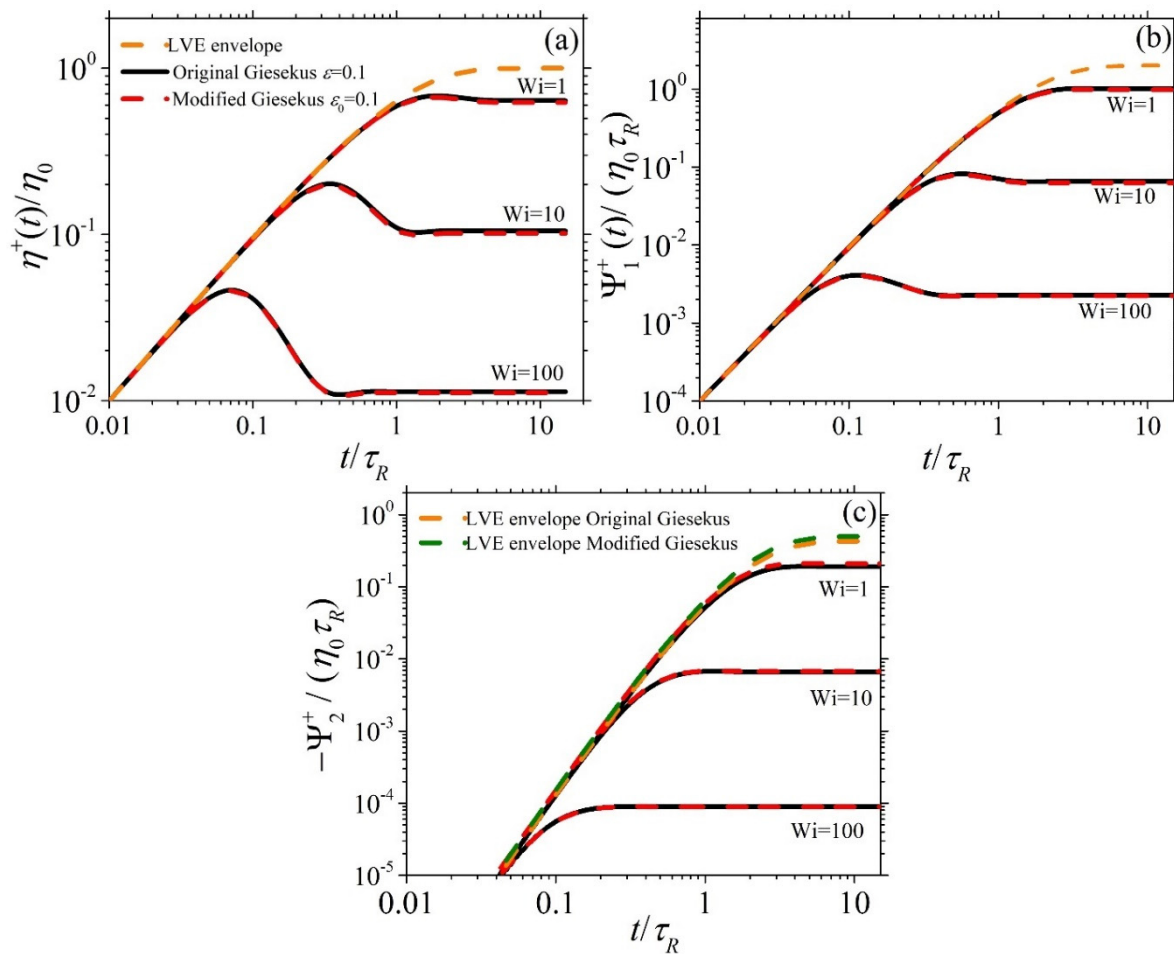


Figure 4. Model predictions when $\varepsilon_0 = 0.1$ for the growth of the (a) shear viscosity, (b) first normal stress coefficient, and (c) second normal stress coefficient, upon the inception of shear flow at different dimensionless shear rates as a function of dimensionless time. The dotted dark yellow lines in each panel and the dotted olive line in panel (c) depict the LVE envelope given by Eq. (6).

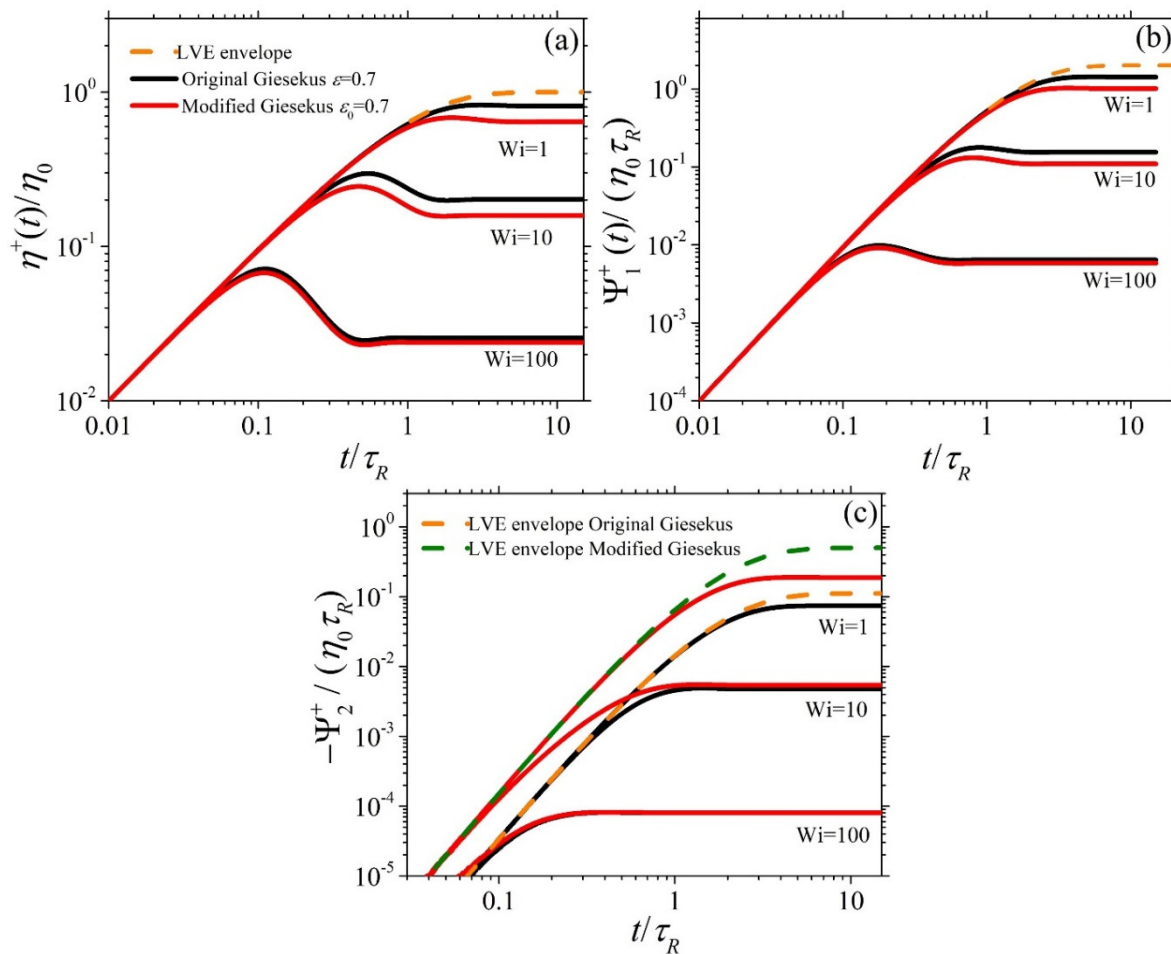


Figure 5. Same as Figure 4 but for $\varepsilon_0 = 0.7$.

4. Conclusions

The Giesekus model has accumulated since its introduction in the early 1980s increased attention due to its simplicity and its capacity to conform to various experimental attributes [3]. Giesekus [13], to validate his postulate, showed that his expression for the mobility tensor can be obtained through a linearization of the mobility tensor proposed in phase-space theory for concentrated polymer solutions and polymer melts of Curtiss and Bird [10–12], which allowed him to relate his anisotropy parameter to the link tension coefficient of Curtiss-Bird. However, recent evidence by Stephanou et al. [14,15] unambiguously shows that the link tension coefficient should not be considered a constant but should depend on strain rate and time via the nematic order parameter. Since the implications of using a variable anisotropy parameter have not been presented before, in this work, we provide the predictions of a modified Giesekus parameter where the Giesekus parameter is no longer constant. Under steady-state simple shear flow, we note that the two models exhibit the same behaviour when ε or ε_0 is small, and at any value of ε or ε_0 at small and large shear rates. A similar behavior is noted for the transient shear viscosity and the transient first normal stress coefficient in start-up simple shear flow; however the predictions are noted to be very different in the case of the transient second normal stress coefficient in start-up simple shear flow where the LVE envelope is noted to shift to larger values, which is more in line with experimental data [14,20]. We thus expect that the use of a variable Giesekus parameter will improve the predictive capacity of constitutive models that employ the Giesekus parameter to more accurately predict the rheological behaviour of polymeric systems. We aim to undertake such an improvement of the Stephanou et al. [20] in the future.

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