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Non-Linear Viscoelasticity of Filled Polymer Melts: Stress and Strain Amplification Approach

J. Domurath,[1] M. Saphiannikova,*[1] G. Heinrich[1,2]

Summary: The aim of the present study is to develop an analytical stress and strain amplification approach (SSAA) for description of non-linear mechanical response in polymer melts containing spherical hard inclusions in a non-dilute regime. The main presumption of this approach is that non-aggregating filler particles reinforce a polymer matrix purely hydrodynamically. The SSAA is verified by comparison with two other approaches existing in literature, when only stress or strain is amplified, and selected experimental results.

Keywords: amplification approaches; filled polymer melts; rheology; strain; stress

Introduction

Filled polymers have been in the focus of industrial interest since decades due to a plenty of applications as functional materials in high-tech branches, in production of goods and in everyday life. Depending on the application purpose, polymers can be filled with different particles which dramatically change their physical and mechanical properties. For example, addition of particles increase electrical conductivity, increase photostability, decrease flammability or improve mechanical characteristics. In particular, mechanical reinforcement is of great importance for soft materials, that is polymer melts and elastomers, because it can open a new field of application, extend the usable temperature range or make the final composite materials lighter and tougher.

Theoretical description of reinforced viscoelastic properties of the filled polymer materials is an exiting topic, albeit extremely challenging. The main reason is that the soft polymer matrices such as polymer melts and networks are usually filled with the particles, which actively interact not only with each other building fractal agglomerates,[1–3] but also with the surrounding polymer chains changing their equilibrium and dynamic properties in a nanosize layer in the vicinity of a particle surface.[3–5] Therefore, prediction of final mechanical properties of a reinforced polymer material stays in many cases an unsolved problem due to the interplay of too many parameters. The main challenge nowadays is the prediction of these properties from first principles, that is, by knowing the mechanical properties of a polymer matrix (relaxation spectrum, constitutive equation), the size, distribution and volume fraction of particles and, when known and well-characterized, mutual interactions between the particles as well as particle-matrix interactions. However, exactly the presence of different interactions makes this prediction from first principles an extremely difficult and nearly impossible task. Even when the particle-particle and particle-matrix interactions can be neglected and there exist only so-called hydrodynamic interactions, arising as a result of perturbations of the deformation field due to the presence of particles,[6] the task of prediction of the final properties from that of the matrix becomes immediately complicated when one enters into the non-linear regime of deformation.

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[1] Leibniz-Institut für Polymerforschung Dresden e. V., Hohe Straße 6, D-01069 Dresden, Germany
E-mail: grenzer@ipfdd.de

[2] Technische Universität Dresden, Institut für Werkstoffwissenschaft, D-01069 Dresden, Germany

We start the present paper from discussing the main effect upon addition of hard filler particles to soft polymer matrices, namely, an increase of average strain in the matrix, since the particles do not deform. The magnitude of strain amplification is known to be the same for polymer melts and elastomers. In both cases to perform the transition from a micro to a macro level one should satisfy the condition of energy balance,[7,8] which requires an amplification of the stress tensor. Basing on this condition we propose an analytical stress and strain amplification approach for description of non-linear mechanical properties of filled polymer melts.

Amplification Approaches Existing in Literature

It is well-known that the shear viscosity η of a suspension of non-interacting hard spheres can be predicted from the matrix viscosity η_0 in the dilute regime, at low volume fractions $\varphi \leq 0.03$, by Einstein's formula[9,10]

$$\eta = \eta_0(1 + 2.5\varphi) \tag{1}$$

and in the non-dilute regime, at higher volume fractions $\varphi \leq 0.10$, by Batchelor's formula[11,12]

$$\eta = \eta_0(1 + 2.5\varphi + 6.2\varphi^2). \tag{2}$$

Both expressions were derived for the case of an incompressible Newtonian viscous fluid. Interestingly, very similar expressions exist for the case of an incompressible linear elastic solid. So, the shear modulus G of an elastomer filled with the same hard spheres can be predicted from the shear modulus G_0 of the matrix in the dilute regime by Smallwood's formula[13]

$$G = G_0(1 + 2.5\varphi) \tag{3}$$

and in the non-dilute regime by the Chen-Acrivos formula[14]

$$G = G_0(1 + 2.5\varphi + 5.0\varphi^2). \tag{4}$$

The difference between a viscous fluid and an elastic solid can only be seen in the pre-factor of the second-order term in Eqs. (2) and (4), while the first-order term is exactly the same due to a mathematical analogy between the linear elasticity and slow viscous flow (also known as creeping flow).[7] Therefore, it is commonly assumed that both viscosity and shear modulus of a linear viscoelastic fluid (solid) should increase in the same manner upon addition of hard filler particles. This approach, originally proposed by Leonov,[15] is equivalent to multiplying the expression for a matrix stress by the hydrodynamic amplification factor

$$X = \eta/\eta_0, \tag{5}$$

which can be calculated using for example Batchelor's formula (Equation (2)) or further expansions to higher terms like the Krieger-Dougherty equation[16]

$$X = \left(1 - \frac{\varphi}{\varphi_m}\right)^{-[\eta]\varphi_m}, \tag{6}$$

where φ_m is the maximum-packing volume fraction and $[\eta]$ is the intrinsic viscosity.

While in the linear regime of a filled viscoelastic polymer melt the validity of Einstein's equation or further expansions to higher terms is justified, there is little to no knowledge about how to modify constitutive equations in the non-linear regime. For example, Macosko[17] writes that in a shear thinning fluid the non-linear viscosity is expected to be smaller than the equations for the linear regime predict and that shear thinning effects are expected to be seen at lower shear rates. The reason is the higher deformation of the matrix fluid in the vicinity of the rigid particles, which can be characterized by a strain amplification factor a_n. This effect was already predicted by Einstein in his seminal work.[9] The exact expression for the strain amplification factor a_n was derived in the case of an elastic solid by Smallwood[13] and in the case of Newtonian fluid in a non-dilute regime by Batchelor and

Green[11]

$$a_n = \frac{1}{1-\varphi}. \tag{7}$$

Unfortunately, this very important result stays unnoticed when modifying non-linear rheological constitutive equations. Usually, the stress amplification approach is used, which describes the hydrodynamic reinforcement in filled polymer melts by multiplying the expression for matrix stress by the hydrodynamic amplification factor X, even in the non-linear regime.[15,18,19] On the contrary the elastomer community usually multiplies the strain in non-linear hyperelastic models with the same hydrodynamic amplification factor $X = G/G_0$,[2,20–22] which is obviously much higher than the strain amplification factor a_n (Equation 7) predicted by Einstein,[9] Batchelor and Green[11] and Smallwood[13] (for a recent study see Castaneda and Tiberio[23]).

The strain amplification approach used by the elastomer community[2,20–22] is based on the old recommendation of Mullins and Tobin,[24] who wrote in 1965: "For larger deformations where the stress and strain are no longer directly proportional it is necessary to consider whether it is the stress or the strain or a function of both the stress and the strain which is increased by the presence of field…*a priory* it appears more reasonable to consider that the local strains are on average X times greater than the overall strain." More precisely, the assumption of Mullins and Tobin[24] requires that instead of the macroscopic extension ratio λ one should use in the constitutive equation the microscopic extension ratio Λ, calculated from the engineering strain ε as

$$\Lambda = 1 + X\varepsilon, \tag{8}$$

where ε is defined as $\Delta L/L$, the ratio of total deformation to the initial dimension of the material. It is easy to show that the concept of Mullins and Tobin violates the energy balance condition[8,25] (this condition should be satisfied when performing the micro-to-macro transition) by applying this concept to the Hookean solid and comparing the stored macroscopic and microscopic elastic energies:

$$G\varepsilon^2 = G_0(X\varepsilon)^2. \tag{9}$$

This predicts a wrong value of the shear modulus $G = X^2G_0$ instead of the correct one $G = XG_0$. In spite of this obvious result, a criticism of the concept of Mullins and Tobin,[24] to the best of our knowledge, has been never published and can be only found in the PhD thesis of Schikowsky[26] from 1988, who writes (translated from German): "The concept of strain amplification has two essential drawbacks. On the one hand, it is not taken into account that due to the presence of filler the number of network chains per total volume reduces. On the other hand, according to molecular statistical theory one should put the effective (microscopic) extension ratio not into the stress-strain equation but into the elastic energy". Therefore, Schikowsky[26] proposes a so-called consequent model of strain amplification with

$$a_n = \sqrt{\frac{X}{1-\varphi}}. \tag{10}$$

It is important to point to another drawback of the strain amplification approach proposed by Mullins and Tobin.[24] Equation (8) is only valid at small deformations and thus, strictly speaking, cannot be applied in the case of finite deformations, where instead of amplified engineering strain ε one should use appropriately amplified finite deformation tensors (true strain measures). Nowadays, in spite of all these drawbacks, the use of the same reinforcing factor of the modulus, X, for estimation of the effective strain in the filled polymer matrix according to Equation (8) is a common thing in the industry and science of filled rubbers.[2,3,27] The price for violating the energy balance condition due to overestimation of the strain amplification factor is however high. One unavoidably confronts a number of mathematical problems, among them difficulties with keeping the condition of constant volume in the case of incompressible rubbers and a need to exchange original matrix parameters for example cross-linking and entanglement densities on some effective fitting parameters.[27,28] The latter modification contradicts the very principle of hydrodynamic amplification concept, namely prediction of composite properties from the matrix values, as well as recent findings showing that the density of crosslinks and entanglements is not changed by the presence of filler particles.[29]

Following the common praxis in the elastomer community, the concept of Mullins and Tobin has been overtaken by some of rheologists for description of shear stresses in filled polymer melts (see for example the studies of the Sarvestani group [30–32]). Such an approach, when applied to a shear-thinning viscoelastic melt filled with hard non-interacting spheres, inevitably results in much lower viscosities of the filled system than the matrix viscosity as will be shown in the next section.

Stress and Strain Amplification Approach

Previously, to describe the hydrodynamic reinforcement effects in filled polymer melts we followed the stress amplification approach[19,33] as well as the strain amplification approach.[34] However, the studies of low density polyethylene filled with layered double hydroxide particles[34] revealed that both approaches are enable to reproduce essential observations in the non-linear regime of shearing, specially a shift of the stress overshoot to lower strain rates upon increase of the particle loading.

To overcome this drawback, we have been developing a new approach, which we call the stress and strain amplification approach (SSAA), that accounts properly for the higher deformations near the non-aggregating hard particles in a suspension with a polymer melt as a matrix. Similar to the original derivation of Einstein,[9] we propose to split the hydrodynamic amplification factor predicted by Batchelor and Green[11] into two contributions. One, called the strain amplification factor a_n (see Equation 7), should hold for any kind of flow (shear, elongation) and any type of matrix (viscous, elastic, viscoelastic). The other, called the stress amplification factor, depends on the flow and matrix type, and is given for a simple shear flow as

$$a_s \equiv X/a_n^2 = 1 + 0.5\varphi + 2.2\varphi^2, \tag{11}$$

if one uses the Batchelor's expression for hydrodynamic amplification factor (see Equation (2) and (5)). The viscosity of the suspension can be calculated from the energy balance condition, which requires that the microscopic and macroscopic dissipated energies per unit time should be equal.[9,35] This gives for the viscosity

$$\eta^*(\dot{\gamma}) = a_s a_n^2 \eta(a_n \dot{\gamma}) \tag{12}$$

with the zero-shear value of $\eta^* = a_s a_n^2 \eta_0$ (compare with Equation (11)) and for the stress tensor

$$\boldsymbol{\sigma}^* = a_s a_n \boldsymbol{\sigma}(a_n \dot{\boldsymbol{\gamma}}), \tag{13}$$

where the asterisk denotes the bulk (macroscopic) property of the suspension, whereas η and $\boldsymbol{\sigma}$ describe the matrix values.

It is instructive to compare predictions of the stress and strain amplification approach with the results of two other approaches found in literature. The first one was proposed by Leonov,[15] who amplifies the stress only with the hydrodynamic amplification factor X ($a_n = 1$ and $a_s = X$ in Equation (12-13)). This stress amplification approach used by us in a number of studies[19,33] automatically satisfies the energy balance condition, i.e. the microscopic and macroscopic dissipated energies are equal to one another. The second approach is found in the recent studies of the Sarvestani group,[30–32] where only the strain is amplified with the hydrodynamic amplification factor X. This leads, if one compares the energies, to Equation (12-13) with $a_n = X$ and $a_s = 1$ and a zero-shear viscosity of $X^2\eta_0$, which is obviously too high. Thus, the strain amplification approach proposed by the Sarvestani group[30–32] violates the energy balance condition, similar to the strain amplification approaches used in the elastomer community.[20,22] On the other hand, if one does not compare energies ($a_s = 1/X$ and $a_n = X$ in Equation (11-12)), the strain amplification approach predicts correctly the expected result for the zero shear viscosity $\eta = X\eta_0$.

We tested the three approaches for the case of a simple shear flow using two relatively simple constitutive models: the first engineering model of Wagner[36] and the original Doi-Edwards model with the independent alignment approximation.[37] In all cases the hydrodynamic amplification factor X has been given by the Equation (2) and (5), i.e. by Batchelor's formula for a suspension of non-aggregating hard spheres in the non-dilute regime. Basically we obtained the same qualitative results for both models, therefore here only the predictions based on the modified Doi-Edwards model will be presented. For simplicity we consider only one relaxation mode characterized by the relaxation time λ and the viscosity parameter η_0, since the relaxation time (spectrum respectively) has no influence on the position of the overshoot peak.

Figure 1 shows the shear stresses σ_{xy} in the stress growth experiment for all three approaches as well as the data for a pure matrix material ($\varphi = 0$). It can be seen, that the overshoot peaks shift differently for the three approaches. In the case of stress amplification the overshoot peak shifts only vertically, while the other two approaches result in a vertical and horizontal shift of the overshoots. The position of the shear overshoot peak is given for the stress amplification approach and no amplification (pure matrix) by

$$\gamma_{\max} \approx 2. \tag{14}$$

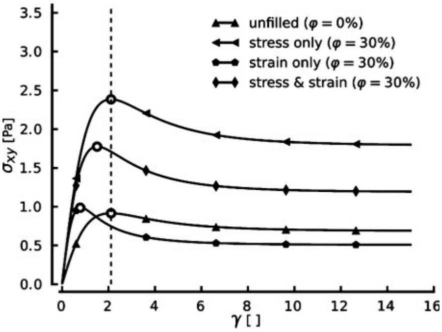

Figure 1.
Transient shear stress at the startup of a shear flow with $\dot{\gamma} = 5\ \mathrm{s}^{-1}$, $\lambda = 1$ s, $\eta_0 = 1$ Pa s, $\varphi = 30\%$, circles indicate maxima.

For the stress and strain amplification approach the shear overshoot peak shifts to

$$\gamma_{\max} \approx \frac{2}{a_n} \tag{15}$$

and for the case of the strain amplification approach to

$$\gamma_{\max} \approx \frac{2}{X}. \tag{16}$$

Figure 2 presents the comparison of the shear dependences of the steady-state viscosities predicted by the three approaches. In the case of stress amplification one observes only a vertical shift of the viscosity compared to the viscosity of the pure matrix. Hence, this approach is not

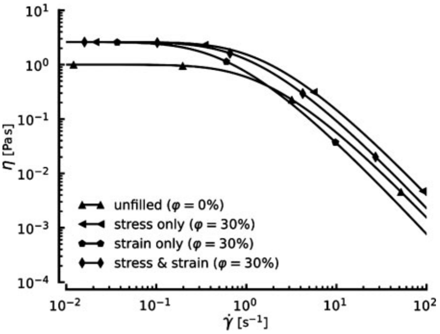

Figure 2.
Shear rate dependences of steady-state viscosity for the three approaches, parameters as in Figure 1.

able to describe an important experimental observation that the shear thinning for the filled system appears at lower shear rates than for the pure matrix. This effect is described by the other two approaches. The new approach with simultaneous strain and stress amplification predicts that the viscosity lies between that for the pure matrix and the viscosity predicted by the stress amplification approach. Contrary, the strain amplification approach predicts that at high shear rates the viscosity drops below the value of the pure matrix. Thus, the strain amplification approach predicts unphysical results for the shearing of filled polymer melts in the non-linear regime. The reason is that this approach violates the energy balance condition, while the macroscopic and microscopic dissipated energies are not equal anymore.

The stress and strain amplification approach predicts the shift of stress overshoot to lower strains with the increase of particle concentration (see Figure 3). Such peculiar behaviour of the matrix overshoot peak was indeed observed recently for the LDPE melts filled with layered double hydroxides[34] and cannot be explained in the frame of two other approaches existing in literature, when only stress or strain is amplified. Figure 4 shows predictions of the stress and strain amplification approach for the steady-state viscosities at different concentrations of particles. It is clearly seen that the shear thinning starts at lower shear rates with increase of the particle concentration.

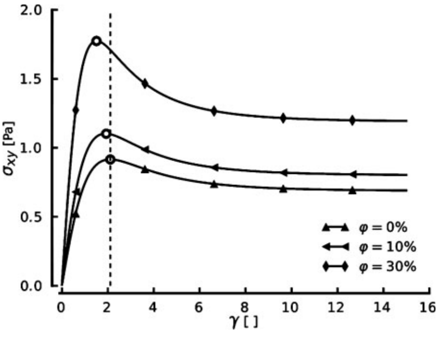

Figure 3.
Transient shear stress at the start-up of a shear flow with $\dot{\gamma} = 5\,s^{-1}$, $\lambda = 1$ s, $\eta_0 = 1$ Pas for different volume fractions of particles, circles indicate maxima. The vertical dashed line indicates the position of shear maximum for the pure matrix.

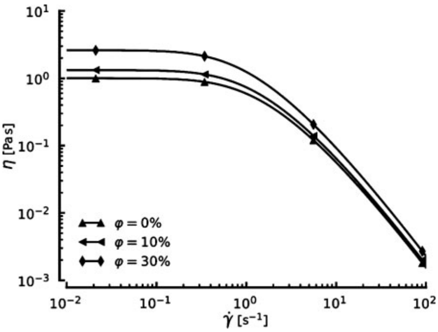

Figure 4.
Shear rate dependences of steady-state viscosity for different volume fractions of particles, parameters as in Figure 3.

Experimental Validation of Stress and Strain Amplification Approach

Additionally, we tried to verify the SSAA by comparison with selected experimental results found in literature. For this purpose one can use for example the shear rate dependences of steady-state viscosity for polymer melts filled with non-interacting particles. So, we compared the SSAA predictions with the data obtained by Poslinski et al.[38] for a thermoplastic polymer filled with non-interacting glass spheres. The matrix viscosity is found to obey the Bird-Carreau formula[39] (see Figure 5)

$$\eta_M = \eta_M^0\left[1 + (\lambda_M \dot{\gamma}_0)^2\right]^{\frac{n-1}{2}} \tag{17}$$

with the zero shear rate viscosity $\eta_M^0 = 117\,Pa$, the time constant $\lambda_M = 0.14\ s$ and the shear thinning exponent $n = 0.49$. The SSAA implies (see Equation 12) that the viscosity of filled thermoplastics in the non-dilute regime (volume fractions $\varphi \leq 0.10$) can be predicted from the matrix viscosity,

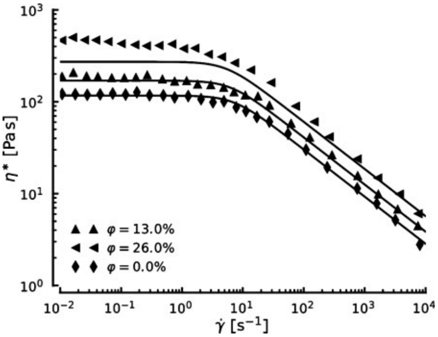

Figure 5.
Comparison of SSAA predictions with the data of Poslinski et al.[38] The matrix is a thermoplastic polymer at 150 °C. It is filled with non-interacting glass spheres with the average diameter of 15 μm and a narrow-size distribution.

if the Bird-Carreau formula[39] will be modified as follows:

$$\eta(\varphi) = X\eta_M^0\left[1 + (\lambda_M a_n \dot{\gamma}_0)^2\right]^{\frac{n-1}{2}}, \tag{18}$$

where X and a_n are given by Equation (5) and (7), correspondingly. Figure 8 shows a very good agreement between the SSAA predictions and experimental data for the system with low volume fraction ($\varphi = 0.13$), where Batchelor's formula is still valid. The steady-state viscosity data for the system with $\varphi = 0.26$ are however underpredicted by the SSAA, as in this intermediate regime one should use an appropriate dependence $X(\varphi)$ given for example by Krieger-Dougherty equation[16] (see Equation (7)) and presumably a stronger dependence of the strain amplification factor a_n on volume fraction φ than that given by Equation (7), for example diverging when approaching the maximum-packing volume fraction φ_m:

$$a_n = \left(1 - \frac{\varphi}{\varphi_m}\right)^{-1}. \tag{19}$$

Separation of the stress and strain amplification factors at higher filler loadings will be a topic of further generalizations of the SSAA.

Conclusion

In this contribution we propose a new approach, which we call the stress and strain amplification approach (SSAA), to account properly for the higher deformations near the non-aggregating hard spherical particles in a suspension with a polymer melt as a matrix. The SSAA as well as two other amplification approaches existing in literature have been tested using the original Doi-Edwards model with the independent alignment approximation.[37] The stress and strain amplification approach predicts the shift of the stress overshoot to lower shear rates with the increase of particle concentration. This effect was observed recently for the LDPE melts filled with layered double hydroxides[34] and cannot be explained in the frame of two other amplification approaches, when only stress or strain is amplified. Additionally, we show that the strain amplification approach predicts unphysical results (stresses lower than the matrix stress) for the shearing of filled polymer melts in the non-linear regime. The reason is that this approach violates the energy balance condition, while the macroscopic and microscopic dissipated energies are not equal anymore.

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