



# Polymeric suspensions in shear flow: Relaxation and normal stress differences



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## ARTICLE INFO

### Article history:

Received 5 July 2016

Revised 12 December 2016

Accepted 17 December 2016

Available online 19 December 2016

### Keywords:

Polymeric suspension

Relaxation

Normal stress difference

## ABSTRACT

Viscometric properties of non-colloidal polymeric suspensions are investigated, showing a longer relaxation time with increasing volume fraction of the particulate phase, due to the influence of particles that form a pairwise close-contact microstructure. This results in a shift of the onset of the shear thinning to a lower shear rate, with a corresponding shift in the first normal stress difference. The effective relaxation time derived from the first normal stress difference and the shear stress is investigated as a function of the particle volume fraction. The relative relaxation time (normalized by the relaxation time of pure polymeric fluid) is independent of the polymeric suspending mediums used in our experiment (various silicone oils of different molecular weights).

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## 1. Introduction

Viscoelastic properties of polymeric suspensions can be greatly modified with the addition of particles, and its understanding is therefore very important in industrial processes in crude oil transport, injection molding of filled polymeric materials, etc. The interaction between polymer molecules and nano-particles in polymeric nanocomposites has been studied both experimentally and numerically [1–10], at low Peclet number,  $Pe \approx 0$  ( $Pe = \dot{\gamma}a^2/D$ , where  $\dot{\gamma}$  is shear rate,  $D = kT/6\pi\eta_0a$  is the self-diffusivity of a particle of radius  $a$  and thermal energy  $kT$  in a fluid of viscosity of  $\eta_0$ ). The conformations of a polymer chain can be significantly altered, compared to the polymers in bulk, if a typical length scale of the confined space is less than a diameter of gyration,  $2R_g$ , of the polymer molecules. This behavior is subject to many factors, such as the end effect of polymers at the interface, interactions between walls and polymers, decrease of entanglement and segregation of polymer chains [4]. In nanocomposites, at medium and high volume fractions of the particulate phase, since the gap between particles is smaller than a diameter of gyration of the molecular chains ( $R_g \sim 10$  nm), the motion of the macromolecules is significantly affected by the particles. The entangled molecular chains are found to be significantly disentangled, which results in a transition from the confinement between molecular chains to the confinement by particles [9]. With increasing particles volume fraction,  $\phi$ ,

the relaxation behavior of polymeric nanocomposites is considerably slowed down due to the confinement effect of the particles, and results in the considerable change of the rheological behavior [8].

For polymeric suspensions with non-colloidal (micron-sized) particles, by contrast, the behavior of polymer phase is unlikely to be affected by particles at static state ( $Pe \approx 0$ ), due to the large gap between particles at equilibrium [11] (Fig. 1(a)). Nevertheless, in shear flow ( $Pe \gg 1$ ), the pairwise particle configuration becomes completely different. Some works have been done on the motion of particles in the viscoelastic matrix and the interaction between the particle phase and the polymeric phase [12–18], Hwang et al. [12] simulate single-, two- and many-particle motions in simple shear flow in an Oldroyd-B fluid, and find that two neighboring particles exhibit a tumbling behavior. Yoon et al. [18] study the two-particle interactions, and find three types of interactions: pass (particles approach and then rotate around each another and continue on moving in their original direction after interaction), return (particles approach one another and return), and tumble (particles approach and continually rotate around one another). They also find that particles are pushed closer with increasing elasticity of the fluid. Experimentally, Snijkers et al. [15] find that the fore-aft symmetry (an equal probability of a second particle lying on a trajectory where it is approaching (fore) or receding (aft) relative to a reference particle) of the open trajectory of an isolated pair of particles in a Newtonian fluid is distorted in a shear-thinning viscoelastic suspending medium. The particles is less likely to tumble compared to simulation. The particle microstructure (configura-

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## Nomenclature

$a$	radius of particle
$D$	self-diffusivity
$\mathbf{D}$	deformation-rate tensor
$e$	gap between two nearest particles
$F_{pp}$	normal force measured by parallel-plate geometry
$g_{bl}(\mathbf{r})$	pair-distribution function at boundary layer of particles
$g^0(\mathbf{r})$	equilibrium pair distribution function for hard spheres
$g^\infty(\mathbf{r})$	pair distribution in flow with high Peclet number
$G$	rigidity modulus
$k$	Boltzmann's constant
$\mathbf{L}$	effective velocity gradient
$M_n$	number average molecular weight
$M_w$	weight average molecular weight
$N_1$	first normal stress difference
$N_2$	second normal stress difference
$Pe$	Peclet number
$R$	radius of plate in parallel-plate and cone-plate geometries
$R_g$	radius of gyration
$\mathbf{S}_{bl}^H$	hydrodynamic stress tensor at boundary layer of particles
$\mathbf{S}_{bl(xy)}^H$	hydrodynamic shear stress at boundary layer of particles
$T$	absolute temperature
$\mathbf{u}^T$	velocity gradient in shear flow
$We$	Weissenberg number
$\dot{\gamma}$	shear rate
$\dot{\gamma}_c$	critical shear rate accounting for shear thinning
$\dot{\gamma}_R$	rim shear rate
$\varepsilon$	parameter related to elongation behavior of polymeric fluid
$\eta$	constant viscosity coefficient
$\eta_\infty^0$	dynamic viscosity at high frequency
$\lambda$	characteristic relaxation time of suspension
$\lambda_0$	characteristic relaxation time of pure fluid from experiment
$\mu$	viscosity of polymeric suspension
$\mu_0$	viscosity of pure fluid
$\mu_p$	viscosity of polymer matrix from PTT model
$\xi$	parameter accounts for slip between molecular network and continuum medium
$\boldsymbol{\tau}$	stress tensor
$\tau_{xy}$	shear stress
$\phi$	volume fraction

tion of a pair of interacting particles) in a polymeric suspension in shear is found to be similar to that in a Newtonian suspension of rough particle, as illustrated in Fig. 1(b) [19,20]. There is an excess of particles along the compressional axes (approaching the reference particle) and relatively a smaller number of particles in the extensional quadrants (leaving the reference particle).

Therefore, with approaching of particles in shear flow (Fig. 1(b)), a lubrication force arises tending to separate particles by a gap that could be smaller than  $2R_g$ . This could then affect the dynamics of the polymer matrix by confining a fraction of polymer chains in the microstructure formed by the approaching particles. As a result, the relaxation behavior of a polymeric suspension is expected to be different from that at static state [11]; the approach of particles can also induce local shearing in the microstructure [21], both of which result in the complicated

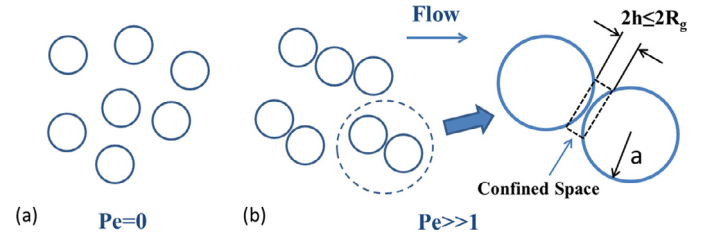


Fig. 1. A sketch of the particle microstructure in suspension (a) in static state and (b) in shear flow with  $Pe \gg 1$ .

rheological behavior. Zarraga et al. [22] find that the magnitude of the first normal stress difference,  $N_1$ , of the non-colloidal polymeric suspension can be attributed to the viscoelasticity of the suspending fluid, while the second normal stress difference,  $N_2$ , at high concentration approaches that measured for a similar Newtonian suspension. Tanner and Qi [23] point out that the stress tensor of the polymeric suspension composes of the stress from a Newtonian part and a viscoelastic part, and the relaxation time of the viscoelastic part increases with increasing volume fraction. It is found in a previous study [11] that, the shift of  $N_1$  with increasing  $\phi$  in a non-colloidal polymeric suspension, which has been reported and much studied [22,24–27], is caused by a change of the relaxation behavior of polymer molecules in the confined space between particles. Furthermore, the local shear-rate enhancement in the confined space may also contribute to the shift of the onset of shear thinning behavior. At high volume fractions, the second normal stress difference,  $N_2$ , mainly arises from the hydrodynamic interactions of the particle phase, and can be described by the theory of Brady and Morris [19] for the Newtonian suspensions.

In this paper, we study the rheological behavior of non-colloidal polymeric suspensions using three types of silicone oils with different molecule weights as suspending mediums. Based on the particle microstructure that is similar to that in Newtonian suspensions of rough particles, the relation between the effective relaxation time and the particle volume fraction is identified. Normal stress differences are also investigated.

## 2. Theoretical background

There are many representative constitutive equations for polymer solutions. We choose the Phan–Thien–Tanner (PTT) model as a convenient means to discuss our experimental data. This choice is dictated by convenient for discussing our experimental data, and by no means, a necessary choice. In the PTT model, the extra stress is written as

$$f(\text{tr}(\boldsymbol{\tau}))\boldsymbol{\tau} + \lambda \overset{\nabla}{\boldsymbol{\tau}} = 2\eta\mathbf{D}, \quad (1)$$

where  $\boldsymbol{\tau}$  and  $\mathbf{D}$  are the extra-stress and deformation-rate tensors,  $\text{tr}$  is the trace operation,  $\lambda$  is a relaxation time,  $\eta = G\lambda$  is the constant viscosity coefficient, where  $G$  the rigidity modulus. Here,  $\overset{\nabla}{\boldsymbol{\tau}}$  is upper convected derivative defined as

$$\overset{\nabla}{\boldsymbol{\tau}} = \frac{d}{dt}\boldsymbol{\tau} - \mathbf{L} \cdot \boldsymbol{\tau} - \boldsymbol{\tau} \cdot \mathbf{L}^T,$$

where the effective velocity gradient,  $\mathbf{L}$ , is defined by

$$\mathbf{L} = \nabla\mathbf{u}^T - \xi\mathbf{D}.$$

In these equations,  $\nabla\mathbf{u}^T$  is the velocity gradient and the parameter  $\xi$  accounts for the slip between the molecular network and the continuum medium. The linearized form of  $f(\text{tr}(\boldsymbol{\tau}))$  is used in this study

$$f(\text{tr}(\boldsymbol{\tau})) = 1 + \frac{\varepsilon\lambda}{\eta}\text{tr}(\boldsymbol{\tau}),$$

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