MODELLING STRESS RESPONSE OF GLASS-FIBRE COMPOSITES DURING SHEAR FLOW USING 3-DIMENSIONAL FIBRE ORIENTATION EVOLUTION AND FIBRE MIGRATION DATA

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Abstract
Constitutive models for stress predictions in fibre filled composites have considered the effects of evolving fibre orientation distribution. However, studies incorporating effects of fibre concentration migration have been limited. This work is aimed at evaluating the combined effects of fibre orientation evolution and fibre concentration migration on shear stress predictions. Steady shear rheology was performed on Nylon-6 containing 33% weight short glass fibres (PA6-33GF) which displayed a stress overshoot before reaching steady state values. This has been attributed to fibre orientation evolution and fibre migration taking place during shear. Samples subjected to different strain units were obtained from the rheometer and X-ray Computed Tomography (X-CT) was employed to obtain 3-dimensional fibre orientation tensors and fibre concentration distribution. With higher applied strains, the orientation of fibres in the shearing direction was observed to increase. The occurrence of fibre migration was also observed. The Reduced Strain Closure (RSC) model and Suspension Balance Model (SBM) were used for fibre orientation and fibre concentration modelling respectively. Empirical parameters for these models were fitted using experimental data. The fibre orientation and concentration values obtained from these models were incorporated into the Lipscomb stress equation to obtain stress predictions for simple shear flow. These predictions were compared with results obtained from a parallel plate rheometer. This constitutive model can be used for predicting stress responses more accurately in complex flow processes such as injection moulding.

1. Introduction

In recent times, fibre filled polymer composites are increasingly replacing metal products as they require lower processing energy requirements and are potentially lighter while offering comparable mechanical and thermal properties. Much of the recent research on composites has focused on the mechanical properties of the finished product and rheological properties in the molten phase. The complexity in predicting these physical properties arises from the anisotropy introduced by the fibres. The orientation and concentration distribution can have a significant influence on both mechanical and rheological properties and hence understanding of orientation and concentration effects is essential for constitutive modelling. The focus of this work will look at the effects of fibre orientation and concentration on the melt rheological properties of composites containing short glass fibres.

Techniques for measuring fibre orientation distribution have mostly been 2-dimensional techniques in the past which presented a directional ambiguity in determining 3-dimensional fibre orientation in composites [1]. X—ray Computed Tomography (X-CT) is a recently introduced technique for fibre orientation measurement that mitigates the directional ambiguity [2]. Furthermore, X-CT provides an added benefit of measuring fibre volume fraction distribution [3]. This had previously been a difficult task as fibre volume fraction was determined by matrix burn-off which meant only bulk concentration values could be obtained.

Constitutive models for predicting viscosity and shear stress response of composites in shear flow have been developed with the intention of improving predictive simulations used for industrial processes such as injection and compression moulding. These models have at present considered the effects of evolving fibre orientation on shear stress [4, 5]. However, experimental validations on the combined effects of fibre orientation and concentration have been limited. The aim of this work is to present experimental characterisation of 3D fibre orientation and fibre concentration evolution in glass fibre composites that are subjected to simple shear flow. Predictive models for fibre orientation and concentration are used to match the experimental data. Following this, a rheological model is developed that incorporates these effects and the stress predictions are compared with experimental rheological data.

2. Theory

2.1 Fibre orientation modelling

Based on the initial work done by Jeffery [6] in modelling the periodic motion of ellipsoidal particles, fibre orientation theory has been modified to better explain the observed behaviour in semi-dilute and concentrated suspensions. The modifications are required primarily due to the lower alignment observed in suspensions containing higher concentrations of fibres arising from fibre-fibre interactions. For semi-dilute suspensions, the Folgar-Tucker model [7] estimates the rate of change in the second-order orientation distribution based on the deformation and vorticity of the flow. The function incorporates the second-order and fourth-order orientation distribution along with an additional term (Ci) for fibre interactions. Ci values can be determined experimentally or using empirical equations that typically depend on fibre volume fraction and aspect ratio.

In the case of concentrated suspensions, researchers determined that the Folgar-Tucker model was predicting quicker evolution of fibre orientation compared to experimentally observed values. This required slowing down the evolution rate in the Folgar-Tucker Model without violating the objectivity of the equation. Wang et al. [8] added a scalar parameter to the term that encompasses the rate of change of the eigenvalues. As shown in Equation (1), the RSC model incorporates a scalar term K which ranges between 0 and 1. L_4 and M_4 are forth-order tensor functions related to the eigenvalues (λi) and eigenvectors (ei) of the second-order orientation tensor. The RSC model has shown greater accuracy for predicting fibre orientation in concentrated suspensions and hence will be used for obtaining fibre orientation predictions.

$$dA/dt = (W\cdot A - A\cdot W) + \xi\{D\cdot A + A\cdot D - 2[A_4 + (1-\kappa)(L_4 - M_4{:}A_4)]{:}D\} + 2\kappa Ci\dot{\gamma}(I - 3A) \quad (1)$$

Legend:

dA/dt = Time evolution of second-order orientation tensor
W = Vorticity tensor
A = Second-order orientation tensor
ξ = Particle shape factor
D = Shear deformation tensor
Ci = Interaction coefficient
$\dot{\gamma}$ = Scalar strain rate
L_4 = Fourth-order orientation tensor function
M_4 = Fourth-order orientation tensor function
κ = Scalar factor

2.2 Fibre concentration Modelling

Models for fibre migration have been employed to predict changes in particle concentration during flow. The basic principles of migration models are governed by the mass and momentum conservation equations. Fibre migration in non-colloidal suspensions has been observed experimentally during shear flow caused by differences in viscosity and particle concentration. The diffusive flux model developed by Phillips et al. [9] determines the migration flux based on the gradients of particle concentration and viscosity. The diffusive flux model has however shown discrepancies when predicting fibre migration in shear slow taking place in a rotational rheometer. The experimental results showed limited migration in the radial direction while the model predicted fibre migration due to shear rate variance in a parallel plate flow.

The Suspension Balance Model (SBM) as shown in Equation (2a) was then developed by Nott and Brady [10] to determine migration flux based on the total particle stress in addition to the gradient in particle concentration and viscosity. The particle stress component encompasses shear stress and non-Newtonian normal stress components. This method has shown to be more accurate in predicting fibre migration is different types of flow regimes. The calculation of the particle stress components is performed as per Equation (2c) which was developed by Morris and Boulay [11] for shear flow of composites containing spherical particles. It must be noted that these relationships were developed for spherical particles and fibre filled systems would potentially have different contributions due to the additional orientation effects. For the purposes of this work, the SBM model for spherical particles will be used for obtaining fibre migration data.

$$d\emptyset/dt = -\nabla\cdot\{(2r_{eff}^2/9\eta_m)[f(\emptyset)](\nabla\cdot\Sigma_{Particles})\} \quad (2a)$$

Legend:
$d\emptyset/dt$ = Time evolution of particle concentration
$f(\emptyset)$ = Sedimentation hindrance function
$\Sigma_{Particles}$ = Particle contribution to bulk stress
r_{eff} = Effective particle radius
η_m= Matrix viscosity

$$f(\emptyset) = (1-\emptyset/\emptyset_m)[(1\text{-}\emptyset)^{(n-1)}] \quad (2b)$$

Legend:
$f(\emptyset)$ = Sedimentation hindrance function
$\emptyset_m$ = Maximum packing volume fraction
n = Power coefficient = [2:5]

$$\Sigma_{Particles} = 2\eta_m\eta_p(\emptyset)D - \eta_m\eta_n(\emptyset)\dot{\gamma}Q \quad (2c)$$

Legend:
$\Sigma_{Particles}$ = Particle contribution to bulk stress
η_m = Matrix viscosity
$\eta_p(\emptyset)$ = Dimensionless shear stress contribution $= [(1-\emptyset/\emptyset_m)^{-2} - 1]$
D = Shear deformation tensor
$\eta_n(\emptyset)$ = Dimensionless normal stress contribution $= K_n(\emptyset/\emptyset_m)^2(1-\emptyset/\emptyset_m)^{-2}$
$\dot{\gamma}$ = Scalar strain rate
Q = Anisotropic tensor for the normal stress contributions

3. Materials and Methods

Nylon-6 containing no glass fibres (PA-6) and containing 33% by weight short glass fibres (PA6-33GF) supplied by Martogg Group in the form of pellets were used in this study. The materials had a melting point of 215°C with a recommended processing temperature of 250°C. Thermal decomposition was performed using Thermo gravimetric Analysis (TGA) to verify the mass fraction of fibres. The pellets were compression moulded into 25mm diameter discs for the fibre orientation evolution studies. Centre-gated injection moulded samples were prepared for fibre concentration migration studies as a highly reproducible initial concentration profile was obtained. Samples of 25mm diameter were cut from the moulded part for rheological testing. All rheological tests were carried out in an

ARES G2 parallel plate rheometer at a temperature of 250°C. Fibre orientation and fibre concentration measurements were performed by scanning the samples using X-ray Computed Tomography (X-CT). The scans were performed at a voxel size of 2.5µm on a 6mm wide rectangular strip cut from the 25mm diameter disc. Images obtained from X-CT were rendered into a 3-dimensional representation of the scanned sample using post-processing software VGStudio Max 3.0. After applying a grayscale threshold that separates the polymer and fibres, fibre orientation tensor and fibre concentration distribution data were obtained from the analysis.

4. Results and Discussion

4.1 Fibre orientation experimental and modelling results

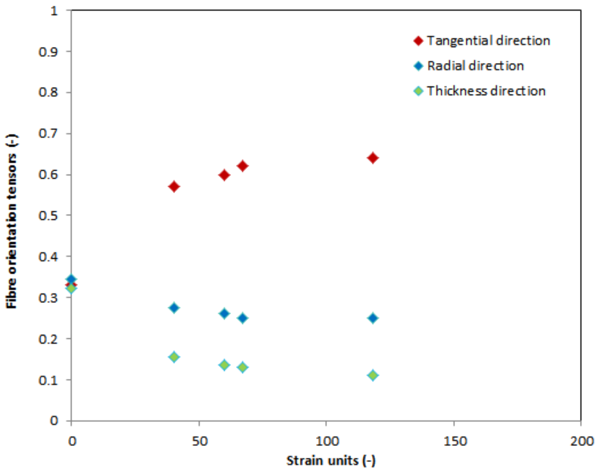
The compression moulded composite samples were subjected to simple shear flow and imaged using X-CT. The initial orientation for the compression moulded samples was 3-dimensional random in the principle directions. The fibre orientation tensor components for different applied strain units were obtained and are shown in Figure 1a. As observed in the figure, when the applied strain is increased, an increase in the orientation tensor component in the shearing (tangential) direction was observed. At the same time, a reduction in the orientation tensor components in the radial and thickness directions were observed. This is typical of fibre orientation evolution in simple shear flow.

Parameters for the RSC fibre orientation evolution model have been fitted for the composite system tested in this study. An interaction coefficient (Ci) value of 0.025 and RSC factor (K) value of 0.20 have been chosen as the best fit. A comparison between the model data and experimental data is shown in Figure 1b and a good agreement is obtained, which allows these fitted parameters to be used for further investigation. Furthermore, the fitted parameters were compared with empirical models currently available for determining Ci values. Models developed by Phan-Thien et al. [12] determined Ci based on bulk fibre volume fraction and fibre aspect ratio as show in Equation (3).

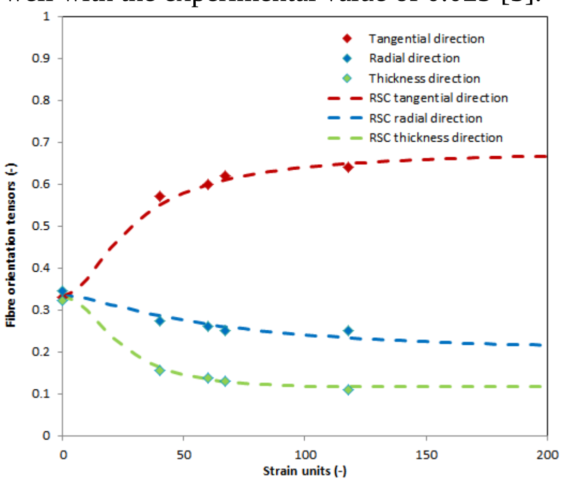
$$Ci=0.03[1-\exp(-0.224\emptyset_t a_r) \quad (3)$$

Legend:
Ci = Interaction coefficient
$\emptyset_t$ = Bulk fibre volume fraction
a_r = Fibre aspect ratio

For the system that was tested in this work, the bulk fibre volume fraction was 0.18 and the average aspect ratio was determined to be 30 based on the fibres imaged using X-CT. The Ci calculated from Equation (3) yield a value of 0.0215 which compares well with the experimental value of 0.025 [3].



1a.

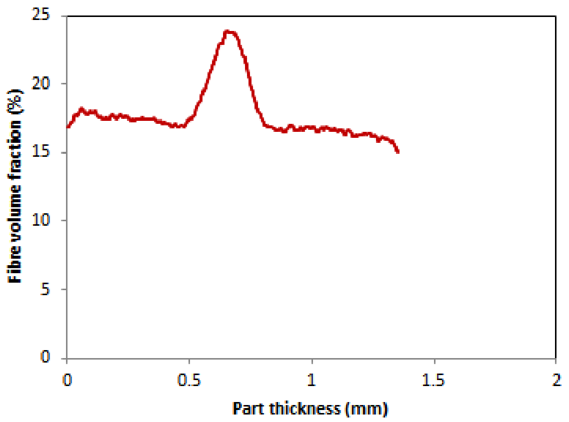


1b.

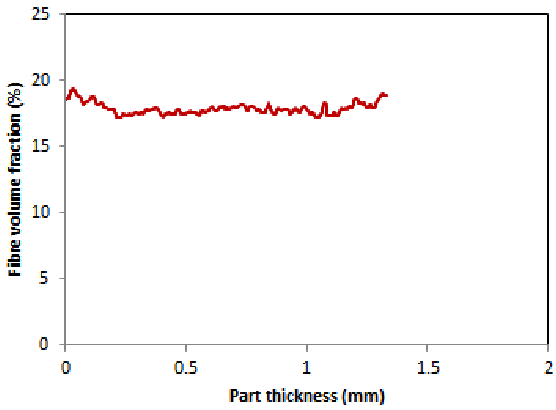
Figure 1a. Experimental fibre orientation evolution data as a function of shear strain
Figure 1b. Experimental and RSC model predictions for fibre orientation evolution data

4.2 Fibre concentration experimental and modelling results

While X-CT has provided an accurate method for determining fibre orientation distribution, it has the added incentive of providing fibre concentration distribution for further investigation. Injection moulded samples were used for this study and a skin-core fibre concentration distribution was observed as shown in Figure 2a before any shearing was performed. This is due to the different magnitudes of shear forces exerted on the sample during preparation. The lower shear rates in the core region of the part cause a migration of fibres from the skin to the core. As a result, the fibre volume fraction at the core region is almost double (27%) compared to that of the skin region (14%). Measuring fibre concentration of samples upon shearing in a rotational rheometer, it is evident that the fibre concentration distribution no longer follows a skin-core structure as observed in Figure 2b. A fairly uniform fibre concentration across the sample thickness direction. This indicates that when the injection moulded sample is subjected to shear forces in the molten phase, fibre migration takes place from regions of higher concentration to lower concentration which eventually leads to a uniform concentration profile. Modelling of this behaviour is carried out using the SBM discussed earlier.



2a.



2b.

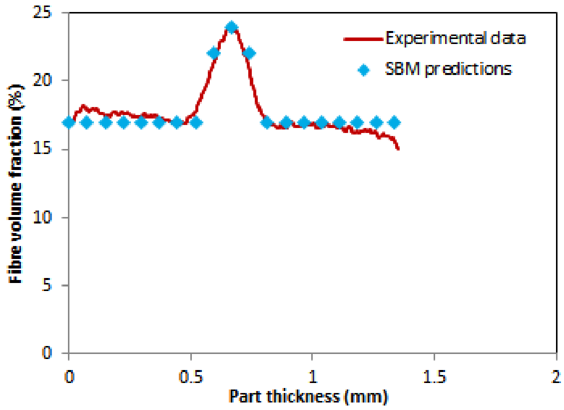
Figure 2a. Fibre concentration distribution before shearing
Figure 2b. Fibre concentration distribution at steady state conditions

In order to obtain predictions of fibre migration from the SBM, several parameters as discussed in Equations (2a), (2b) and (2c) are required. Table 1 lists the values obtained from experimental measurements and other researchers [13] that are needed to calculate fibre migration using the SBM. As shown in Figure 3b, the predicted fibre concentration data at the end of shearing is reasonably accurate representation of the fibre concentration data obtained from experiments. Based on these results, using the SBM for subsequent rheological modelling has been validated.

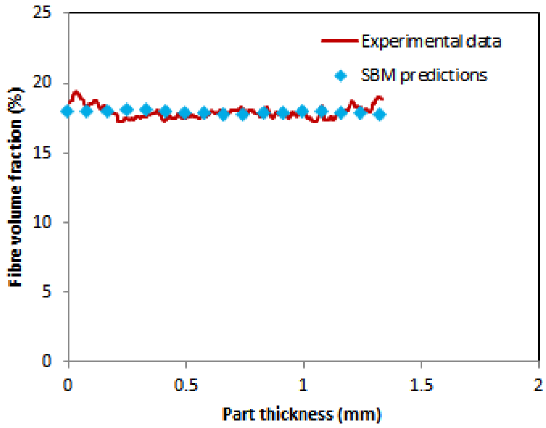
Table 1. Parameters required for SBM

Parameter	Units	Value
Fibre length	µm	450
Fibre diameter	µm	15

r_{eff}	µm	26.67
K_n	-	0.75
η_m	Pa.s	400
$\emptyset_m$	-	0.72



3a.



3b.

Figure 3a. Fibre concentration distribution before shearing with model predictions
Figure 3b. Fibre concentration distribution at steady state conditions with model predictions

4.3 Rheological characterisation and modelling

The transient rheological response of PA6-33GF during steady shear shows a distinct stress overshoot compared to the behaviour of unfilled PA6 as shown in Figure 4. The stress overshoot for the composite material gradually reduces to reach a steady value. This has been attributed to the reorientation and migration of fibres from the initial conditions towards the steady state. In order to develop a rheological model to predict this overshoot behaviour, the fibre orientation and concentration distributions would be a crucial input. Provided that the rheological model can capture the intricacies in simple shear flow, it can then be implemented to complex flow patterns experienced during processing such as injection moulding.

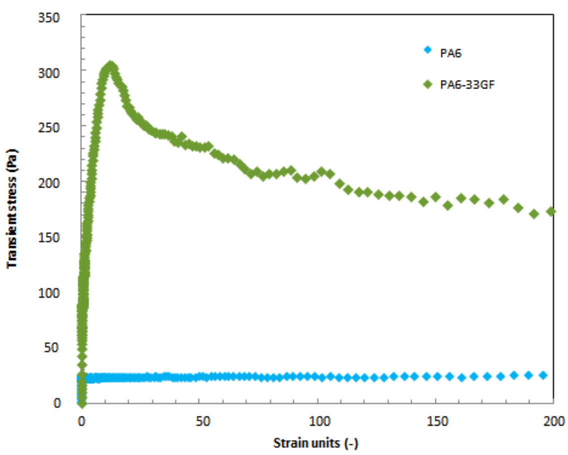


Figure 4. Comparison of transient stress between PA6-33GF and unfilled PA6

The Lipscomb stress equation [14] as shown in Equation (4) has been used in modelling transient rheology of fibre filled composites in shear flow. The Lipscomb model contains terms that incorporate stress contribution from the matrix, from the addition of particles and from the orientation of the particle.

$$\sigma = \eta_m D + 2\eta_m \phi D + 2\eta_m \phi N D{:}A_4 \quad (4)$$

Legend:
η_m= Matrix viscosity
D = Shear deformation tensor
ϕ = Fibre volume fraction
N = Fibre coupling coefficient
A_4 = Fourth-order orientation tensor

However, studies incorporating the combined effects of fibre orientation and concentration have been limited. It must be noted that the Lipscomb model was developed for dilute suspensions containing fibres and hence the equation might require empirical modifications to fit the behaviour seen in concentrated suspensions. Using the Lipscomb model, predictions for shear stress were obtained with values for fibre orientation and concentration taken from the RSC model and SBM respectively. The comparison between the experimental and model results is shown in Figure 5. It can be seen that the model predicts a stress overshoot which decays to a steady value. However, further refinements may need to be performed to increase the accuracy of these predicted values.

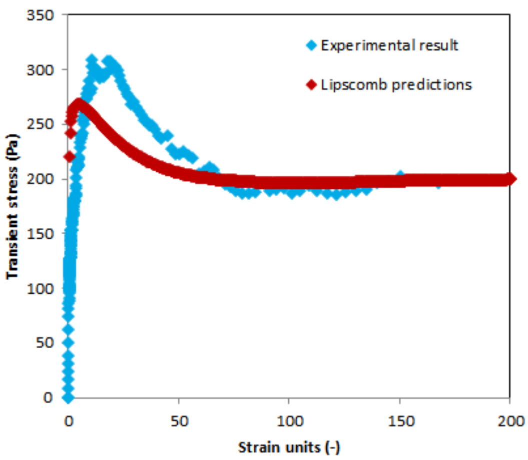


Figure 5. Comparison of experimental transient stress with predictions from Lipscomb model

5. Conclusions

The work is aimed at evaluating the combined effects of fibre orientation evolution and fibre concentration migration on shear stress predictions. Steady shear rheological testing on the filled composites displayed a stress overshoot which decays to a steady value. This is likely due fibre orientation evolution and fibre concentration migration. The RSC model was used to obtain for fibre orientation evolution predictions and compared with experimental fibre orientation obtained from X-CT. Likewise; the SBM was used to validate fibre concentration migration observed experimentally during simple shear. Finally, the fibre orientation and concentration values obtained from these models were incorporated into the Lipscomb stress equation to obtain stress predictions. The stress predictions were compared with steady shear results obtained from a parallel plate rheometer which showed a reasonable agreement. Further refinements need to be performed to enhance the capabilities of this model.

References

1. T. Köpplmayr, I. Milosavljevic, M. Aigner, R. Hasslacher, B. Plank, D. Salaberger and J. Miethlinger: Influence of fiber orientation and length distribution on the rheological characterization of glass-fiber-filled polypropylene. *Polymer Testing*, 32(3), 535-544 (2013)

2. G. M. Vélez-García, P. Wapperom, D. G. Baird, A. O. Aning and V. Kunc: Unambiguous orientation in short fiber composites over small sampling area in a center-gated disk. *Composites Part A: Applied Science and Manufacturing*, 43(1), 104-113 (2012)

3. V. Perumal, R. K. Gupta, S. N. Bhattacharya and F. S. Costa: Fiber orientation prediction in nylon-6 glass fiber composites using transient rheology and 3-dimensional x-ray computed tomography. *Polymer Composites*, 0(0) (2018)

4. M. J. Cieslinski, P. Wapperom and D. G. Baird: Fiber orientation evolution in simple shear flow from a repeatable initial fiber orientation. *Journal of Non-Newtonian Fluid Mechanics*, 237, 65-75 (2016)

5. A. P. R. Eberle, D. G. Baird, P. Wapperom and G. M. Vélez-García: Using transient shear rheology to determine material parameters in fiber suspension theory. *Journal of Rheology*, 53(3), 685-705 (2009)

6. G. B. Jeffery: The motion of ellipsoidal particles immersed in a viscous fluid. In: *Proceedings of the royal society of London A: Mathematical, physical and engineering sciences*. The Royal Society, (1922)

7. F. Folgar and C. L. Tucker: Orientation Behavior of Fibers in Concentrated Suspensions. *Journal of Reinforced Plastics and Composites*, 3(2), 98-119 (1984)

8. J. Wang, J. F. O'Gara and C. L. Tucker: An objective model for slow orientation kinetics in concentrated fiber suspensions: Theory and rheological evidence. *Journal of Rheology*, 52(5), 1179-1200 (2008)

9. R. J. Phillips, R. C. Armstrong, R. A. Brown, A. L. Graham and J. R. Abbott: A constitutive equation for concentrated suspensions that accounts for shear-induced particle migration. *Physics of Fluids A: Fluid Dynamics*, 4(1), 30-40 (1992)

10. P. R. Nott and J. F. Brady: Pressure-driven flow of suspensions: simulation and theory. *Journal of Fluid Mechanics*, 275, 157-199 (1994)

11. J. F. Morris and F. Boulay: Curvilinear flows of noncolloidal suspensions: The role of normal stresses. *Journal of Rheology*, 43(5), 1213-1237 (1999)

12. N. Phan-Thien, X. J. Fan, R. I. Tanner and R. Zheng: Folgar–Tucker constant for a fibre suspension in a Newtonian fluid. *Journal of Non-Newtonian Fluid Mechanics*, 103(2–3), 251-260 (2002)

13. R. M. Miller, J. P. Singh and J. F. Morris: Suspension flow modeling for general geometries. *Chemical Engineering Science*, 64(22), 4597-4610 (2009)

14. G. G. Lipscomb, M. M. Denn, D. Hur and D. V. Boger: The flow of fiber suspensions in complex geometries. *Journal of Non-Newtonian Fluid Mechanics*, 26(3), 297-325 (1988)