Microstructures of fiber suspensions in complex geometry

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Abstract: Evolutions of molecular conformation and fiber orientation in fiber suspensions are investigated by collocated finite volume method on unstructured triangular meshes. FENE-P (Finite Extensible Nonlinear Elastic Dumbbell model with Peterlin’s approximation) model which is microstructure-based is chosen to describe the polymeric matrix and TIF (transversely isotropic fluid) model is used to calculate the fiber contribution in order to realize the coupling of flow and fiber orientation. Microstructures of molecule and fiber are obtained by analyzing the information of molecular conformation tensor and second-order fiber orientation tensor respectively. Two numerical examples are considered, namely, a planar contraction flow and a planar flow past a confined cylinder. Present results are hoping to give more insight into the microscopic details of complex flows and thus be more helpful for industrial application.

Introduction

Short fiber reinforced polymeric composite is one of polymer-based composites which are the most widely used in injection or compression moldings. The mechanical properties of these composites are primarily dependent upon the following three factors [1]: (1) strength and modulus of the fiber; (2) strength and modulus of the matrix; (3) effectiveness of the bond between fiber and matrix in transferring stress across the interface. These factors are basically determined by the microstrutural details of these materials, including crystallinity, transcrystalline, fiber orientation, etc. Since these microstrutural details have the important relationship with the manufacture processes, studying the behaviour of fiber suspensions is significant.

Pervious studies, including theoretical and numerical works, have been devoted to the rheological behavior of fiber suspensions. Reviews of these works can be found in Papathanasiou et al. [2] and Petrie [3]. In the scope of fiber suspensions' study, most researchers focus on the Newtonian solution, the viscoelastic nature of polymeric matrix, which is more correlated to the modern composite manufactory, has received relatively modest attention. Several works have been carried out to improve this issue. As for the rheological model of fiber suspensions in polymeric fluids, some important formulations have been constructed with the help of GENERIC modeling. Ait-Kadi and Grmela [4] developed fiber suspension model in FENE-P fluid by using two internal conformation tensors. Ramazani et al. [5, 6] developed a GENERIC structure to take into account the contribution of polymeric matrix and rigid fiber interaction. Rajabian et al. [7,8] formulated the suspensions of semiflexible fibers in polymeric fluids with GENERIC structure. Eslami et al. [9, 10] later used the similar structure to model the polymer/layered silicate nanocomposites. As for the numerical studies of fiber suspensions in polymeric fluids, Azaiez et al. [11]
investigated the fiber orientation and vortex size with different polymeric matrix model in the planner contraction. Azaiez [12] as well as Sharifi and Azaiez [13] studied the reduction and the vortex dynamics in the fiber-laden free shear flows. As far as we know, molecular morphology of the polymeric matrix has not yet been investigated, in contrary to the extensively studied microstructures of fiber. This is probability due to the fact that the pervious works are mainly concentrated on the suspensions with Newtonian solution. However, for the polymer-based composites or especially the semicrystalline polymer-based composites, the study of morphology of the polymer chains during the process becomes urgent since the deformed structures of matrix can change crystallization forms [14] and result in different crystallinity as well as the different morphology of transcrystalline [15] in solid state. Relating the final mechanical properties to the processing parameters is not an obvious task. However, understanding how molecules and fibers evolve with flow during the processes can considerably help to predict the microstructures and the ultimate properties.

The purpose of this study is to investigate the microstructures of the molecule and fiber during the processes. We want to understand better the connections between microstructures change and the macroscopic behavior of the flow. To do this, Finite Extensible Nonlinear Elastic Dumbbell model with Peterlin’s approximation (FENE-P model) [16] which is microstructure-based is chosen to describe the polymeric matrix; transversely isotropic fluid (TIF) model [2] is used to calculate the fiber contribution in order to realize the coupling between flow and fiber orientation, meanwhile, the Natural approximation is selected to close the fourth-order fiber orientation tensor. The present paper is organized as follows: in section 2, we give the governing equations; in section 3, the brief computational approach is presented; numerical simulation of fiber suspensions in simple shear flow, elongational flow, contraction flow as well as flow past a confined cylinder are investigated in section 4; and finally the conclusions are presented in section 5.

**Governing equations**

For incompressible, isothermal suspension flow, the conservation equations for mass and momentum can be expressed as follows, respectively

\[ \nabla \cdot \mathbf{u} = 0 \quad (1) \]

\[ \rho \frac{D\mathbf{u}}{Dt} = -\nabla p + \nabla \cdot \mathbf{\tau} \quad (2) \]

where \( \mathbf{u} \) denotes the velocity field, \( \rho \) is the density, \( p \) is the pressure field, \( \mathbf{\tau} \) is the extra-stress which can be divided into three parts, defined as

\[ \mathbf{\tau} = \mathbf{\tau}_s + \mathbf{\tau}_p + \mathbf{\tau}_f \quad (3) \]

where \( \mathbf{\tau}_s = \eta_s \dot{\gamma} \) is the Newtonian solvent contribution, \( \eta_s \) is the Newtonian contribution viscosity, \( \dot{\gamma} \) is the rate of strain tensor, \( \mathbf{\tau}_p \) is the polymer contribution stress, \( \mathbf{\tau}_f \) is the fiber contribution stress.

We use FENE-P model to describe the polymeric matrix. This model treats the macromolecules as dumbbells consisting of two point-mass beads connected by a Warner spring with its maximum extension equal to \( b \) and immersed in a Newtonian solvent. We shall mention that we assume the suspensions are dilute, hence, the polymer-fiber interactions are neglected.
The constitutive equation of the FENE-P model can be written as follows [16]
\[ \dot{\lambda} C = -\frac{C}{1 - \text{tr}C/b} + \delta \]  
(4)
\[ \tau_p = \frac{\eta_p}{\lambda} \left( \frac{C}{1 - \text{tr}C/b} - \delta \right) \]  
(5)

Here \( C \) is a dimensionless conformation tensor, \( \lambda \) is the relaxation time of the polymer chains, \( \nabla \) is the upper-convected derivative, defined as \( \nabla = \frac{DC}{Dt} = C \cdot \nabla - (\nabla u)^T \cdot C \), \( \delta \) is the identity tensor, \( \eta_p \) is the polymer-contributed viscosity. As pointed by Beris et al. [16], the conformation tensor is symmetric, positive-definite and can be treated as a generalized deformation tensor. It can be used to describe the morphology of the polymer chains. From the physical point of view, the first invariant of tensor \( C \), the trace of conformation tensor, may represent the extension of the polymer chains [16]. Moreover, the orientation of the polymer chains can be described through the eigenvalues and eigenvectors of the conformation tensor.

For the fiber contribution to the extra stress tensor, we shall apply TIF model [2] and take the polymer contribution into account to obtain the following expression
\[ \tau_f = (\eta_s + \eta_p) \phi \left[ A \dot{\gamma} : a_4 + B (\dot{\gamma} \cdot a_2 + a_2 \cdot \dot{\gamma}) + C \dot{\gamma} + 2FC_I [\vec{\gamma} | a_2] \right] \]  
(6)
in which \( \phi \) is the fiber volume fraction, \( A = \frac{re^2}{2(\ln(2re) - 1.5)} \), \( B = \frac{6\ln(2re) - 11}{re^2} \), \( C = 2 \), \( F = \frac{3re^2}{\ln(2re) - 0.5} \), \( C_I \) is the fiber interaction coefficient which is a function of fiber length and diameter, fiber concentration as well as fiber orientation, \( |\vec{\gamma}| = \sqrt{\frac{1}{2} \gamma_{ij} \gamma_{ji}} \) is the scalar magnitude of \( \dot{\gamma} \), \( a_2 \) and \( a_4 \) are the second- and fourth-order orientation tensors respectively. When the cylinder fiber with random orientation is considered \( A = \frac{re^2}{3\ln(\pi/2re)} \), meanwhile, the “B term” and “F term” are neglected. The corresponding fiber stress expression becomes
\[ \tau_f = (\eta_s + \eta_p) \phi \left[ \frac{re^2}{3\ln(\pi/2re)} \dot{\gamma} : a_4 + 2\dot{\gamma} \right] \]  
(7)

The evolution of the second-order orientation tensor which can be derived from the continuity equation of probability distribution function is written as follows [2, 17]
\[ \frac{D a_2}{Dt} = -\frac{1}{2} (\omega \cdot a_2 - a_2 \cdot \omega) + \frac{\chi}{2} (\dot{\gamma} \cdot a_2 + a_2 \cdot \dot{\gamma} - 2\dot{\gamma} : a_4) + 2C_I |\vec{\gamma}| (\delta - ma_2) \]  
(8)
where \( \omega \) is the vorticity tensor, \( \chi = (re^2 - 1)/(re^2 + 1) \) with \( re \) the fiber aspect ratio, \( m \) is a constant equal to 2 for two-dimensional problems, and 3 for three-dimensional problems to satisfy the condition \( ma_2 \) = 1.
Note that the fourth-order orientation tensor $a_4$ appears in equation (8). Hence, the closure approximation is needed. Here, we choose the natural closure [11], since this kind of closure is more accurate than the hybrid one [17] and its cost is cheaper than that of the Eigenvalue Based Orthotropic Fitted closure (EBOF) [18]. The closure approximation in two-dimensional problem is given by

$$a_{ijkl} = \frac{1}{6} \text{det}(a_{ij}) (\delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) + \frac{1}{3} (a_{ij}a_{kl} + a_{ik}a_{jl} + a_{il}a_{jk})$$

(9)

where $a_{ijkl}$, $a_{ij}$, $\delta_{ij}$ are the components of $a_4$, $a_2$, $\delta$, respectively.

Thus, the equations (1)(2)(3) together with the polymer contribution equation (5), evolution of molecular conformation equation (4), the fiber contribution equation (7) and the evolution of fiber orientation equation (8), are the whole governing equations. It should be noted that when the suspensions are semi-dilute or concentrated, the interactions between fibers and molecules are significant. Therefore, evolution of molecular conformation equation (4) together with evolution of fiber orientation equation (8) becomes more complex [5-10].

**Numerical method**

In this study, continuity equation (1), momentum equation (2) together with evolution of molecular conformation equation (4) and evolution of fiber orientation equation (8) are coupled and simultaneously solved by collocated finite volume method on fully triangular meshes. This is due to the fact that this method has several advantages, e.g., it is not only very easy to carry out on irregular geometries and convenient to deal with the location of variables, but also has cheap computational cost [19]. During the implementation of the above progress, the Semi-Implicit Method for Pressure Linked Equations Consistent (SIMPLEC) algorithm is employed to solve the equations, we particularly adopt the technique provided by Mathur and Murthy [20] to discrete the general transport equation and then the robust solutions are obtained. Additionally, Rhie and Chow [21] type interpolation is used to overcome the pressure-velocity as well as the stress-velocity decoupling. Meanwhile, the convection terms are approximated using either upwind or deferred correction scheme depending on the equation types. More details of the method can be obtained from our previous work [22].

**Results and discussion**

We start with simple shear and elongational flow; before that we investigate the microstructures of fiber suspensions in contraction and cylinder flows. We introduce the following characteristic numbers: the Reynolds number $Re = \rho U_0 L / \eta_0$, the Deborah number $De = \lambda U_0 / L$, the parameter for the viscosity $\beta = \eta_t / \eta_0$, in which $L$ is the characteristic dimension of the domain, $U_0$ is the characteristic velocity and $\eta_0 = \eta_t + \eta_p$ is the total viscosity. We use the scalar $f_w = 1 - m^m \text{det}(C/\text{tr}C)$ to represent the degree of molecular orientation/deformation, $f_f = 1 - m^m \text{det} a_4$ to denote the degree of fiber orientation, with $m$ the dimension of space. Since all the flows we study here are two-dimensional, $m$ is equal to 2. The factor $f_f$ is derived from the hybrid closure [17] of fiber orientation tensor. When $f_f \to 0$, the random
orientation is obtained, while \( f_f \to 1 \), fairly aligned orientation is achieved. Similar principle can be also applied to molecules, expect that they can be stretched. That is why we introduce \( C/\text{tr} C \), which is a traceless tensor exactly like \( a_2 \). Note that in all our works the orientation ellipsoids of molecule are plotted by \( C/\text{tr} C \).

**Simple flow field**

As we know the molecular conformation tensor is highly dependent on shear/elongational rate, while fiber orientation tensor is significantly influenced by the shear/elongational strain. In the following, molecular conformation is tested while varying shear/elongational rate, meanwhile, fiber orientation is investigated with changing shear/elongational strain.

Fig. 1 shows the molecular extension and orientation degree with changing the shear/elongational rate. As we can see, the extension of molecules \( \text{tr} C \) and the orientation factor \( f_m \) climb rapidly with increase of shear/elongational rate, especially for elongational rate.

![Fig. 1](image)

**Fig. 1.** Molecular extension and orientation degree in simple flow.

Fig. 2 describes the relationship between the fiber orientation factor and the shear/elongational strain. We test two different fiber aspect ratio: \( re = 10 \), \( re = 20 \). In the shear flow, when the fiber-fiber interaction is neglected \( (C_f = 0) \), which reduces to the Jeffery’s model, fibers rotate periodically with the period about \( 2\pi re / |\dot{\gamma}| \). When moderate fiber-fiber interaction is considered \( (C_f = 0.001) \), fibers tumble in the vicinity of the period. But for the larger fiber aspect ratio case, fibers become harder to tumble. Moreover, when the fiber-fiber interaction is larger \( (C_f = 0.01) \), fibers become steady oriented after a transitory higher aligned state. In the elongational flow, fibers do not rotate in a wide range of fiber-fiber interaction, and soon get aligned with the flow direction.
In this section, we study the flow through planner contraction. We particularly focus on four regions noted A-D, see Fig. 3. The flow near the upstream wall far from the corner (zone A) and downstream wall (zone D) is shear dominated, whereas near the centerline in the vicinity of the entry region (zone C) it is extensional flow dominated. In zone B, the flow is more complex and mainly rotational.

Fig. 3. Contraction flow geometry.

The suspensions studied here have finite fiber aspect ratio and moderate fiber-fiber interaction. Parameters we used in the simulation are $Re_0 = 0$, $b = 10$, $\beta = 0.3$, $re = 10$, $C_f = 0.001$, $\phi = 0.5\%$. Initialization of fiber orientation is set to be random, that is,
\( a_2 = \delta / 2 \). As to the polymer, initial condition is \( \tau_p = 0 \), which means \( C = b\delta / (b + 2) \). Boundary of fiber orientation tensor in the inlet is set to be random. The velocity in the inlet is \( u = 3U_0 (1 - (y/4L)^2) / 8, v = 0 \).

We test for two cases, \( De=2.625 \) and \( De=7.875 \), by varying the characteristic velocity \( U_0 \). Fig. 4 shows the molecular orientation ellipsoids, orientation degree and extension, respectively. As we can see from the orientation ellipsoids, the orientation becomes significantly distinct at different regions. At the downstream where the flow is mainly dominated by the higher shear flow, ellipsoids change into pipe and orient with the flow direction. At the entry region, the flow is mainly extensional, the ellipsoids are parallel to the flow direction. However, the upstream including the vortex zone is not highly oriented because of the lower shear rate. The trace of the conformation tensor which represents the extension of the molecule, gains the maximum value near the wall downstream and minimum value at the centerline near the inlet as well as the vortex region. And the value ranges from \( 2b / (b + 2) \) to \( b \). The relationship between the Deborah number and the orientation degree is that: the higher Deborah number is, the higher oriented/deformed structure is obtained. This is reasonable since the higher Deborah number relates to higher shear rate.

For the orientation of fiber, we present the result of \( De=2.625 \). Similar results can be obtained at different Deborah numbers. In Fig. 5, pictures (a)-(f) give the evolution of fiber orientation. Fibers in the downstream near the wall and at the entry region first orient along the flow direction (a). This structure is similar as molecule obtained in the above segment. When the time increases, fibers upstream become oriented (b) and then fibers near the wall downstream start tumbling (c)-(d). Picture (e) gives the whole regions except for vortex region which are highly oriented, while picture (f) gives the final structure that all the fibers are oriented to the flow direction.

**Fig. 4.** Conformation of molecule at different \( De \) numbers (from up to down: orientation ellipsoids, orientation degree, molecular extension).
Fig. 5. Orientation of fiber at different time.

Fig. 6. Fiber orientation factor with changing shear strain at different zones.
The final fiber orientation is similar to the results obtained by Azaiez et al. [11] where a more widely parametric study was used. The structure is also similar to the experimental work of expansion geometry by Yasuda et al. [23] and the numerical work of backward-facing step channel by Chiba and Nakamura [24].

To be more clear with the fiber evolution, we now test for its behavior in four different zones (see Fig. 3): zone A: A_1(3, 3.9), A_2(4, 3.6), A_3(3, 3.6); zone B: B_1(5.5, 3.5), B_2(5.5, 3), B_3(5, 3.95); zone C: C_1(6, 0.5), C_2(6, 0), C_3(7, 0); zone D: D_1(8, 0.98), D_2(11, 0.98), D_3(8, 0.93), D_4(11, 0.93). We can see from Fig. 6 that fibers tumble at upstream far away from the vortex (zone A), in the vortex (zone B) as well as downstream near the wall (zone D). When the shear strain is large enough, the orientation becomes steady. Among these three zones, fibers near the wall downstream exhibit the most similar behavior as we derive in the simple shear flow. In the extension zone C, points C_1, C_2 do not tumble, and exhibit exactly the elongational flow behavior; however point C_3 becomes slightly over aligned then keeps steady.

**Flow past a confined cylinder**

The flow past a cylinder is investigated here. This kind of flow has some significant characters, see Fig. 7, the flow in the gap between the cylinder and channel walls (zone C) is primarily a shear flow, and the flow near the axis of symmetry upstream and in the wake of the cylinder (zone D) has a high extensional component, moreover, the flow upstream (zone A) and downstream (zone B) near the wall is basically a shear flow. Parameters we used in this simulation are the same as those in the contraction flow. The velocity in the inlet is \( u = 3U_0(1 - (y/2L)^2)/2, v = 0 \).

Fig. 7. Cylinder flow geometry.

Fig. 8 describes the molecular conformation at different Deborah numbers. As we can see from Fig. 8, the orientation of molecules is more or less along the flow direction: in the upstream region of the cylinder, material is compressed, so the molecular orientation is perpendicular to the flow; the accelerating flow between the side wall and the cylinder, brings a good alignment along the stream; at the centerline behind the cylinder, owing to higher extension rate, molecule orient along the centreline; at the centerline far away ahead/behind the cylinder where the flow is fully developed, because of the loss of shear rate, molecule becomes less oriented/deformed. We now focus on the stretch of the molecules \( nC \). There are four regions with high polymer chain extension. There are: a short distance upstream of the cylinder; at the sides of the cylinder; downstream of the rear stagnation point and at the channel walls nearest to the cylinder. These results are the same as [25], in which a FENE-P constitutive equation is used to investigate the flow past a periodic, linear array of cylinders.
Fig. 8. Conformation of molecule at different De numbers (from up to down: orientation ellipsoids, orientation degree, molecular extension).

Orientation of fiber at different time is described in Fig. 9. Fibers tend to the flow direction headmost in the regions with higher shear rate (a). Later on, fiber near the channel wall tumbles (b-e). Picture (f) gives the final orientation of fiber, which is more or less along the flow direction. This is also in agreement with the experimental study of Yasuda et al. [26], where they investigated different suspensions past a cylinder in a slit channel.

Fig. 9. Orientation of fiber at different time.

We also test for four different zones’ fiber behaviors (see Fig. 7) as we do in the contraction example. Zone A: $A_1(-5, 1.95)$, $A_2(-3, 1.95)$, $A_3(-4, 1.8)$; zone B: $B_1(3,$
1.95), B_2(5, 1.95), B_3(4, 1.8); zone C: C_1(0, 1.95), C_2(0, 1.05); zone D: D_1(−1.5, 0), D_2(1.5, 0). Zone A and zone B belong to shear flow dominated regions, zone C experiences both shear and elongational strain, while zone D belongs to elongational flow dominated region. Fig. 10 shows the degree of fiber orientation at different points. As we can see, fibers tumble at zone A, B, C, and become steady when the shear stain is large enough. Fibers in zone D, take on a slight higher orientation then become steady; behavior more like the elongational flow.

Fig. 10. Fiber orientation factor with changing shear strain at different zones.

Conclusions

Microstructures of fiber suspensions including molecular conformation and fiber orientation are investigated in this paper. Microstructures of the polymeric matrix, including the molecular orientation, deformation, extension, are derived by analyzing the information of conformation tensor. In the same way, microstructures of fiber, such as orientation and orientation degree, are obtained by the information of second-order fiber orientation tensor.

Simple shear and elongational flow are studied first to get a general cognition of microstructures of fiber suspensions. Orientation/deformation and extension of molecule increase with the increase of shear/elongational rate, especially for elongational rate. Orientation of fiber with different fiber-fiber interaction and different fiber aspect ratio are tested through the change of shear/elongational strain. In the case of shear flow, when no fiber-fiber interaction is considered (\( C_f = 0 \)), fibers (with finite aspect ratio) rotate periodically; when moderate fiber-fiber interaction is taken into account (\( C_f = 0.001 \)), fibers with small aspect ratio exhibit a periodic damp
tumbling motion, however, this become harder for those with larger aspect ratio; when fiber-fiber interaction is large (\( C_I = 0.01 \)), no tumble is observed. In the elongational flow, fibers soon get aligned with the flow direction, and the smaller fiber-fiber interaction, the more aligned the fiber orientation.

Fiber suspensions with finite aspect ratio and moderate fiber-fiber interaction are investigated under the condition of contraction or a confined cylinder latter. In these two geometries, molecules orient more or less with the flow direction, and gain the maximum extension in the regions with highest shear/elongational rate. Almost at the same places, molecules are highly deformed. Due to the different shear/elongational rate at different regions, fibers exhibit distinct behaviors. Fibers in the regions with higher shear/elongational rate orient first. With the increase of time, fibers in the regions dominated by the shear flow tumblering, however when the time is enough, orientations become steady and are along the flow direction more or less. Fibers in the region dominated by extension soon orient along the flow direction and do not change with the increase of time. In summary, molecules and fibers exhibit different behavior depending on the nature of the flow in complex geometries.

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