Chapter 5

Rheology of Highly Concentrated Fiber Suspensions

5.1. Introduction

Because of their remarkable specific physical and mechanical properties, polymer composites that are reinforced with discontinuous fibers or fiber bundles are suitable materials for many aeronautic, automotive, shipbuilding, electrical, electronic, health and sport applications. Among these materials, Sheet Molding Compounds (SMC), Bulk Molding Compounds (BMC), Glass Mat Thermoplastics (GMT), and Carbon Mat Thermoplastics (CMT), i.e., polymer composites that are reinforced with centimeter length fiber or fiber bundles [ADV 94] [ORG 12a] are the subject of several ongoing development and research programs. Other similar polymer composites that are reinforced with bio-sourced fiber or fiber bundles (e.g. flax, hemp, wood, bamboo, kenaf, sisal, jute) are also short-fiber promising polymer composites.

Most of these composites usually have a high volume fraction $\phi$ of fibers that typically ranges from 0.005 to 0.5 with a high aspect ratio $r = l/d$ that typically ranges from 10 to 1000, where $l$ and $d$ are the characteristic fiber length and diameter, respectively, and with random to quasi-ordered fiber orientations. The microstructure of the fibrous reinforcement of these composites during or after processing/forming operations is a complex network of entangled straight/curved/twisted semi-flexible fibers that form multiple contacts with their

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neighbors. The network connectivity of these systems is so high that the mean number of fiber-fiber contacts per fiber, also known as the mean coordination number $\bar{z}$, is largely above the percolation threshold, and is generally above 2.

Figure 5.1 – Evolution of the mean fiber coordination number $\bar{z}$ as a function of the fiber content $\phi$ and the aspect ratio $r$. Most of polymer composites that are reinforced with discontinuous fibers or fiber bundles behave as concentrated fiber suspensions during their processing, i.e., with $\bar{z} \geq 2$, as shown in the images of the fibrous microstructures: nanocomposite with PEO and Nano Fibrillated Cellulose fibers (a, $\phi = 0.01, r = 300, l = 1.5 \mu m, \bar{z} \approx 4$), PMMA+wood fibers composite (b, $\phi = 0.35, r = 66, l = 2 \text{ mm}, \bar{z} = 35$), PMMA+copper fibers composite (c, $\phi = 0.1, r = 50, l = 10 \text{ mm}, \bar{z} \approx 9$) [ORG 12b], model SMC (d, $\phi = 0.13, r = 13, l = 13 \text{ mm}, \bar{z} \approx 3$) [GUI 12b], model short fiber-reinforced composite (e, $\phi = 0.47, r = 10, l = 3 \text{ mm}, \bar{z} \approx 12$).

The evolution of $\bar{z}$ is shown as a function of $\phi$ and $r$ in Figure 5.1 for the studied composites. This parameter is critical for the properties of these materials. A high connectivity is generally targeted to enhance the end-use physical and mechanical properties of composites [OSS 94][THO 96][VAS 08][ORG 12b]. However, a high connectivity also considerably alters the rheology of composites during processing or forming (film casting, injection molding, compression molding).
Figure 5.2 – Typical patterns observed during the flow of highly concentrated fiber suspensions. (a): coupling between the rheology and the initial fiber orientation for a suspension of molten PMMA reinforced with glass fiber bundles and compressed in a channel (around the stress-strain curves: initial and final top views, orientation distribution function $\psi$ and tensor $A$ of the samples) [DUM 07b]. (b): flow of a similar fiber bundle suspension in a confined geometry: the suspension had to flow around a cylindrical obstacle, and was consequently subjected to high velocity gradients. In both cases, only 10% of the bundles were painted black.
During these operations, the fiber-reinforced polymer composites can be considered to behave as non-Newtonian and highly concentrated fiber suspensions that exhibit a complex rheology that is closely related to:

- their microstructure and its evolution during the suspension flow: spatial position, orientation, geometry of fibers and fiber-fiber contacts.
- the deformation micro-mechanisms arising at the fiber scale: translation, rotation and deformation of fibers, creation, loss, deformation of fiber-fiber contacts,
- the rheology and the (potential) flow of the (non)-Newtonian suspending fluid through the fibrous networks.

This complex rheology results in intricate macroscale flow situations that are still not very well understood and modeled: evolving anisotropy, shear thinning behavior at high strain rates, yield stress at low strain rates, hysteresis, strain hardening or softening, fracture, flocculation and migration phenomena. Figure 5.2 and Figure 5.3 illustrate some of these phenomena. These macroscale flow mechanisms are not very well understood because it is difficult to (i) characterize the rheology of highly concentrated fiber suspensions, in particular because of the high concentration in slender (flexible) elements, (ii) to observe and model the microstructures of the

**Figure 5.3** – Typical patterns observed during the flow of highly concentrated fiber suspensions. (a): Evolution of the flocs kinetics with the shear strain rate during the shearing of a suspension of carbone nanotubes [MA 08]. (b): anisotropic deformation of a model suspension of flax fibers and migration of the suspending fluid (a paraffin gel) during a lubricated compression test at a low strain rate (the red square shows the initial shape of the sample and the yellow arrow the preferred fiber orientation).
suspensions as well as (iii) the deformation micro-mechanisms arising at the fiber scale. One objective of this chapter is to explain how some of the aforementioned experimental difficulties were recently solved.

In addition, it is interesting to notice that most of the models for the rheology of concentrated fiber suspensions are an expansion of the theories established for dilute and semi-dilute systems. Some models are quite sophisticated [TOL 94] [SUN 97] [FAN 98] [SER 99a] [SER 99b] [LEC 04] [DJIA 05] [LEC 05] [DUM 09] [FER 09] [NAT 13]. In these models, the suspension is always considered to be a standard Cauchy continuum and to behave as a single-phase and incompressible fluid. The flow of the suspension is ruled by the following mass and momentum balance equations that are written by assuming quasi-static evolutions and neglecting external volumetric forces:

\[ \nabla \cdot \mathbf{v} = 0 \quad \text{and} \quad \nabla \cdot \mathbf{\sigma} = 0, \] \[ \text{[5.1]} \]

where \( \mathbf{v} \) is the macroscale suspension velocity\(^2\) and where the macroscale symmetric Cauchy stress tensor \( \mathbf{\sigma} \) is usually split into four contributions:

\[ \mathbf{\sigma} = -p \mathbf{\delta} + \mathbf{\sigma}^m + \mathbf{\sigma}^{f/m} + \mathbf{\sigma}^{f}, \] \[ \text{[5.2]} \]

where the pressure \( p \) arises from the fluid incompressibility, \( \mathbf{\delta} \) is the identity tensor, and \( \mathbf{\sigma}^m \) the stress contribution of the suspending fluid:

\[ \mathbf{\sigma}^m = \frac{1}{V} \int_{V^m} \mathbf{\sigma}^m dV, \] \[ \text{[5.3]} \]

where \( V \) and \( V^m \) are the elementary volumes of the suspension and of the suspending fluid, respectively, and where \( \mathbf{\sigma}^{f/m} \) and \( \mathbf{\sigma}^{f} \) denote fluid-fiber interactions and fiber-fiber contacts, respectively. For example, in the case of slender straight fibers, these two contributions can be approximated as follows:

\[ \mathbf{\sigma}^{f/m} = \frac{1}{V} \sum_c \int_{S^c} s_\alpha \mathbf{p}^\alpha \otimes f^{a/m} ds^c, \] \[ \text{[5.4]} \]

and

\[ \mathbf{\sigma}^{f} \approx \frac{1}{V} \sum_c \mathbf{G}^a \mathbf{G}^\beta \otimes f^{\alpha/\beta} = \hat{\varphi} \frac{1}{N_c} \sum_c \mathbf{G}^a \mathbf{G}^\beta \otimes f^{\alpha/\beta} \] \[ \text{[5.5]} \]

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\(^2\) In order to distinguish phenomena arising at the microscale (fiber scale) from those resulting at a macroscale (sample scale), a specific notation for macroscopic quantities is introduced: if \( \mathbf{x} \) is a scalar physical microscale quantity, then \( \mathbf{\bar{x}} \) is its associated macroscale quantity.
In the two last equations, \( F \) and \( C \) are respectively the sets of the \( N_f \) fibers and the \( N_c \) fiber-fiber contacts in the elementary volume \( V \). \( \Gamma_\alpha \) denotes the interface between the suspending fluid and the fiber \( \alpha \) of tangent vector \( \mathbf{p}^\alpha \) and center of mass \( G^\alpha \). \( s_\alpha \) is the curvilinear abscissa along the fiber \( \alpha \). Furthermore, \( f^{\alpha/m} \) and \( f^{\alpha/\beta} \) respectively represent (i) the hydrodynamic force per unit length between the fluid and the matrix at \( s_\alpha \) and (ii) the contact force at a contact between the fiber \( \alpha \) and the fiber \( \beta \). The previous set of equations is often completed with additional equations that describe the evolution of the set \( \mathcal{M} \) of relevant microstructure descriptors:

\[
\frac{d}{dt} \mathcal{M} = F(\mathcal{M}, ...), \tag{5.6}
\]

For example, for homogeneous suspensions of straight fibers, \( \mathcal{M} \) is restricted to the fiber orientation distribution function \( \psi \), and the function \( F \) is based on modified expressions of the well-known Jeffery’s equations [JEF 22] established for the orientation dynamics of an ellipsoid in an infinite Newtonian fluid:

\[
\frac{d}{dt} \psi + \nabla_\mathbf{p} \cdot (\psi \mathbf{p}) = 0 \quad \text{with} \quad \dot{\mathbf{p}} = \mathbf{\Omega} \cdot \mathbf{p} - \lambda (\mathbf{p} \cdot \mathbf{D} \cdot \mathbf{p}) \mathbf{p} - \mathbf{D}_r \cdot \nabla_\mathbf{p} \psi, \tag{5.7}
\]

where \( \lambda = (r^2 - 1)/(r^2 + 1) \), \( \mathbf{\Omega} \) and \( \mathbf{D} \) are the vorticity and strain rate tensors, and where \( \mathbf{D}_r \) is a diffusion tensor accounting for the interactions of the fiber of tangent vector \( \mathbf{p} \) with its neighborhood [FOL 84] [RAH 95] [KOC 95] [PET 00] [NAT 13]. To reduce the calculation times, the function \( \psi \) is often replaced by its first moment, namely the second order fiber orientation tensor \( \mathbf{A} \) [ADV 87]:

\[
\mathbf{A} = \oint \mathbf{p} \otimes \mathbf{p} \psi d\mathbf{p}. \tag{5.8}
\]

Doing so a similar but more compact expression to equation [5.7] is obtained where the fourth order fiber orientation tensor \( \mathbf{A} \) is introduced [ADV 87]:

\[
\dot{\mathbf{A}} = \oint \mathbf{p} \otimes \mathbf{p} \otimes \mathbf{p} \otimes \mathbf{p} \psi d\mathbf{p}. \tag{5.9}
\]

For instance, for an isotropic diffusion term, i.e. for \( \mathbf{D}_r = \mathbf{D}_i \mathbf{I} \), the rate \( \dot{\mathbf{A}} \) of the second order fiber orientation tensor \( \mathbf{A} \) is written as follows:

\[
\dot{\mathbf{A}} = \mathbf{\Omega} \cdot \mathbf{A} - \mathbf{A} \cdot \mathbf{\Omega} + \lambda (\mathbf{A} \cdot \mathbf{D} + \mathbf{D} \cdot \mathbf{A} - 2\mathbf{A} : \mathbf{D}) + 2\mathbf{D}_r (\mathbf{I} - \alpha \mathbf{A}), \tag{5.10}
\]

with \( \alpha = 2 \) or 3 in 2D or 3D dimensions. Closure approximations are then required to write \( \dot{\mathbf{A}} \) as a function of \( \mathbf{A} \) [HAN 62] [HIN 76] [ADV 87] [DUP 99].

The previous set of balance and constitutive equations brings up the following comments:
- When the suspension flow corresponds to this general single-phase and incompressible framework (e.g., the flow situation illustrated in Figure 5.2(a-b)), these equations clearly emphasize the role of the suspensions microstructures $\mathcal{M}$ and deformation micro-mechanisms. Obviously, proper methods to characterize these features would help the building of relevant theories. For example, in order to propose a suitable analytical estimate for $\mathbf{f}$, it would be of great interest to properly characterize and model the set $\mathcal{C}$. Providing relevant expressions for $f^{\alpha/\beta}$ would also be meaningful. As mentioned previously, these points will be further addressed.

- The relevance of the equations used to predict the fiber orientation has been subjected to numerous debates in the literature: is Jeffery’s equation relevant to be the basis of fiber orientation evolution in concentrated systems? If it is actually the case, what is the relevance of $\mathbf{D}_r$, of using orientation tensors, of closure approximations? This will be also discussed in the following sections.

- In many practical situations, the fiber suspensions flow in confined geometries, i.e., in zones (i) which dimensions are of the same order of magnitude than the fiber (or fiber bundles) or (ii) where the suspensions can be highly curved or subjected to severe velocity gradients, as shown in Figure 5.2(b). In these situations, can a concentrated fiber suspension still be considered to be a standard Cauchy medium, i.e., with a symmetric stress tensor? Some comments will be made on this critical point in this chapter.

- The examples given in Figure 5.3(a-b) show that in many situations the flow of highly concentrated fiber suspensions markedly deviates from a single-phase flow, thus showing that the above theoretical background is not appropriate. Thus, a more complete modelling framework is needed to detect, without a priori, whether the suspensions flow according to single-phase or two-phase modes: this problem is addressed in the following sections.

Thus, the objective of this chapter is to give some answers to the aforementioned questions and problems related to the characterization and the modeling of the rheology of highly concentrated fiber or fiber bundle suspensions. This chapter is organized as follows. In section 5.2, first some rheometry difficulties, associated with these concentrated systems, are pointed out. Second, the experimental trends that are commonly observed at the macroscale are shown: influence of the fiber content, aspect ratio and orientation on stress levels, orientation dynamics and migration phenomena. In section 5.3, we further explore the microstructures and the micromechanics of highly concentrated suspensions by combining 3D observations of their microstructures, using X-ray microtomography, and micro-rheometry. Using experimental data collected at the macro and microscales, a rigorous upscaling process is used in section 5.4 to study whether highly concentrated systems can be
considered to be standard or enriched continua. In addition, single-phase rheological models are used to discuss the experimental trends and the relevance of dynamics theories for the fiber orientation. Lastly, in order to account for migration phenomena, an extension of the standard single-phase theoretical framework to a two-phase one is presented in section 5.5.

5.2. Experimental trends observed at macro and mesoscales

5.2.1. Rheometry difficulties

As for other concentrated suspensions [COU 05], several challenges are encountered in properly characterizing the rheology of concentrated fiber suspensions. The problems arise from the highly connected and anisotropic fibrous phases that consist of slender semi-flexible elements of finite size, impregnated by in a (non-)Newtonian suspending fluid. The length of fibers raises the question of the scale separation (see below). The rheology of these suspensions must be characterized in various directions (with respect to the preferred fiber orientation) using various mechanical loading conditions because of their anisotropy. The connectivity of the fibers and the cohesion of the fibrous phase can yield to (i) experimental artefacts at the boundary of samples and to complex deformation modes in the samples (see below), (ii) consolidation and migration phenomena for which drained conditions have to be investigated. Therefore, specific rheometry methods must be developed.

5.2.1.1. The problem of separation of scales

In order to extract from rheometry experiments relevant information (for example to build constitutive theories), one of the most critical points to fulfil is the condition of proper separation of scales. Indeed, the typical macroscale size \( L_c \) of the flowing zone from which the measurements are carried out must be adequately large with respect to the size \( l_c \) of the microscale heterogeneities of the deformed suspension, so that the scale separation parameter \( \varepsilon \) remains sufficiently small [AUR 91]:

\[
\varepsilon = \frac{l_c}{L_c} \ll 1. \tag{5.11}
\]

For example, if a homogeneous concentrated fiber suspension is homogeneously sheared inside a Couette or a plane-plane rheometer, \( L_c \) is the rheometer gap. If the suspension flow becomes heterogeneous, e.g. by exhibiting shear bands, \( L_c \) corresponds to the typical size of shear bands. Similarly, if the suspension microstructure remains homogenous, \( l_c \) is of the order of the fiber length \( l \). However, this condition may be broken for concentrated fibrous systems (and more
particularly for colloidal ones) that often by exhibit flocs or clusters [MA08] such as those emphasized in Figure 5.2. In this case, \( l_c \) is the typical size of the flocs.

Figure 5.4 – (a) Typical evolution of the axial \( \sigma_{33} \) and lateral \( \sigma_{22} \) stresses as a function of the Hencky axial compression strain \( \varepsilon_{33} \), obtained during the plane strain compression (\( \varepsilon_{22} = 0 \)) of a SMC (glass fiber bundles, \( \phi = 0.15, r = 41, l = 25 \text{ mm} \)) at an axial compression strain rate \( \dot{\varepsilon}_{33} = 10^{-2} \text{s}^{-1} \). (b) Influence of the initial length \( l_{10} \) on the local Hencky elongational strain \( \varepsilon_{11} \) [DUM03b].

As for other fibrous materials, the value of the parameter \( \varepsilon \) below which an appropriate scale separation is reached depends on the types of fibers, fibrous architectures and mechanical loadings the suspensions are subjected to [PIC11]. This value was investigated for a standard SMC that was subjected to compression
tests. This material consisted of fiber bundles that had a length \( l \) of 25 mm \([DUM.03b]\). Using large samples, Figure 5.4 shows that the macroscale flow of this type of suspensions proceeds homogeneously during lubricated plane strain compression (providing a proper lubrication of the rheometer surfaces in contact with the SMC). However, a careful inspection of the grid painted onto the upper surface of the sample shows that the flow is not locally homogeneous because of the local translations and rotations of fibers. The flow homogeneity was carefully investigated using an initially regular grid and estimating the elongational Hencky strain \( \varepsilon_{11} \) defined as:

\[
\varepsilon_{11} = \ln \left( \frac{l}{l_0} \right),
\]

where \( l_0 \) and \( l \) are the initial and elongated lengths of the segments picked at a different locations on the grid (\( l_0 \) ranged from 20 to 120 mm). Figure 5.4(b) shows that, when \( l_0 \) is close to the fiber bundle length \( l \), \( \varepsilon_{11} \) exhibits a very large scattering and strongly depends on the segment location. Conversely, for a size of the gauge zone \( l_0 = L_c \) that is greater than four times the fiber bundle length, i.e., for \( \varepsilon < 0.25 \), the scattering is minimized and all the assessed mesoscale elongational strains \( \varepsilon_{11} \) are close to the macroscale elongational strain.

Unfortunately, in many studies of the rheology of concentrated fiber suspensions (often performed using commercial rheometers) the scale separation condition \([5.11]\) is not fulfilled. Hence, both the recorded experimental data and the resulting constitutive theories should be cautiously considered. Other studies took into account this constraint. For that purpose, specially designed rheometers were often used \([LIN.97]\) \([KOT.98]\) \([LIN.99]\) \([LEC.02]\) \([SER.02]\) \([GUI.10]\) \([GUI.12a]\).

5.2.1.2. Coupling rheometry with imaging and kinematical field measurements

The control and/or the assessment of flow boundary conditions together with the bulk flow kinematics of fiber suspensions are also of great importance to obtain relevant data from rheometry experiments. These two points are particularly central when studying concentrated systems that are prone to exhibit complex wall slippages, shear bandings and strain localizations \([PIG.96]\) \([COU.05]\). Without controlling or measuring these phenomena, important misinterpretation of rheograms can be done: this is the case of several experimental works reported in the literature.

Suitable methods to overcome these problems and to access to the “true” rheology of the suspensions consist in combining rheometry experiments with local measurements of their mesoscale deformation. Several techniques can be (or have already been) used such as Magnetic Resonance Imaging (MRI) \([CAL.99]\) \([COU.05]\), Ultra Sonic Velocimetry (USV) \([GAL.13]\), measurement of normal
stresses [GUI 12a], 2D/3D optical/x-ray observations coupled with particle tracking [PIG 96] [DUM 03b] (see Figure 5.4), Particle Image Velocimetry (PIV) or Digital Image Correlation (DIC) [ADR 05] [VIG 11]. The best solution is to couple these mesoscale measurements with techniques that enable a simultaneous analysis of the evolution of the fibrous microstructures using 2D/3D optical/x-ray observations [FOL 84] [PET 98] [PET 00] [DUM 07b] [VAS 07] [LE 08] [YAS 02] [LAT 11] [GUI 12b] [WEG 12] [SAA 14].

5.2.2. Typical trends

Figure 5.5 – (a-b) Stress-strain curves recorded during the lubricated simple compression of a BMC (glass fibers, $\phi = 0.1, r \approx 21, l \approx 0.3$ mm) at various constant or crenelated strain rates $D_{33}$ [ORG 08a]. (c) Stress-strain curves recorded during the simple shearing of a SMC (glass fiber bundles, $\phi = 0.188, r \approx 41, l \approx 25$ mm) [LEC 01].

Figure 5.5(a-b) shows several stress–strain curves that were obtained during the lubricated simple compression of an industrial BMC [ORG 08a]. Note that the shape
of the monotonic stress-strain curves shown in these graphs is frequently obtained during the (simple or plane strain) compression of highly concentrated fiber suspensions (see also Figure 5.2(a) and Figure 5.4(a)). This shape can slightly deviate from that reported in this figure depending on the rheology of the suspending fluids, the geometry and the mechanical properties of fibers, the architecture of the fibrous networks and the mechanics of fiber-fiber contacts. First, the compressive stress $\sigma_{33}$ exhibits a sharp increase which is related to viscoelastic effects that are induced both by the rheology of the polymeric matrix and the elastic deformation of the network of fibers. These effects are still pronounced if the suspension is subjected to strain rates jumps, as evident from the non-monotonic curves shown in the graphs. Then, the stress-strain curves usually reach a threshold stress, above which the suspension flow is easier, but still exhibits a strain hardening induced by the progressive reorientation of the fibrous network along the flow direction and by potential additional elastic deformation of fibers.

Figure 5.5(c) shows the typical response of highly concentrated suspensions during shearing. In this particular example, an annular shear test [LEC 02] was performed using an industrial SMC [LEC 01], i.e., with fiber bundles that exhibit a nearly planar random orientation parallel to the shear direction. First, his figure shows a sharp increase in the shear stress $\sigma_{\theta z}$, similar to the increase that is observed during compression (Figure 5.5(a)). Then, the stress reaches a maximum at a shear strain $\gamma \approx 1$. Up to this limit, the shear deformation is homogeneous through the sample thickness. Then the shear stress exhibits a slow decrease. In this case, this phenomenon was attributed to strain localization. In other systems, the observed peaks can also be related to shear bandings and/or to a progressive alignment of the fibers along the shear direction. It is also interesting to notice that, during shear, a compressive normal axial stress $\sigma_{zz}$ is recorded. The potential origin of this effect is a rearrangement of the fibrous microstructure, which is similar to the rearrangements that are observed in sheared granular systems, leading to dilatancy and consolidation phenomena at the macroscale.

The above examples prove that flow and the microstructure of concentrated fiber suspensions are extremely complex and coupled.

5.2.3. Influence of the strain rate

An increase in the imposed strain rate generally leads to an increase in the suspension stress levels. For example, this effect is illustrated in Figure 5.6(a-b). This phenomenon is mainly ascribed to the increase in the hydrodynamic interactions between (i) the fibers and the suspending fluid (the second and third terms in equation [5.2]) and (ii) the forces between fibers in contacts ($f^{\alpha/\beta}$ in equation [5.5]). For many systems, this increase leads to a shear thinning behavior.
This shear thinning behavior is usually higher than (or close to) that of the suspending fluid at the same macroscale strain rate, as illustrated for an industrial SMC in Figure 5.6(a). The origin of this effect may be attributed to (i) the rheology of the suspending fluid (if the shear thinning of the fluid increases with the strain rate, bearing in mind that the local strain rate in the fluid is higher than the macroscale strain rates in many zones of the suspensions), and (ii) the micro-mechanics of fiber-fiber contacts. Thus, these suspensions behave as non-linear viscoelastic fluids, without any noticeable yield stress. However, other systems can exhibit a yield stress at low strain rates and, thus, behave as visco-elastoplastic fluids, because of colloidal interactions or Coulombic friction forces [SER 99a], [SER 99b] [CAB 07] that both arise from fiber-fiber contacts.

**Figure 5.6** – (a) Evolution of the threshold flow stress $\sigma_{33}$ as a function of the compression strain rate $D_{33}$ during the lubricated simple compression of a SMC at various fiber contents (glass fiber bundles, $r \approx 41, l \approx 25$ mm) [DUM 03b]; the values of the power-law index $n$ that are used to fit the experimental data with a power-law are shown. (b) For the same suspension, evolution of the normalized viscosities for lubricated simple (sc) and plane strain
compressions and for shear (s) as a function of the fiber content $\phi$ [DUM 03b]. (c) Evolution of the normalized compression stress as a function of the aspect ratio $r$ for a fiber reinforced fresh mortar (glass fibers appear black, glass fiber bundles appear white, $\phi = 0.006$) [CHA 10]. Evolution of the component $A_{11}$ of the fiber orientation tensor $A$ as function of the compression strain $\varepsilon_{33}$ during the lubricated plane strain compression of the suspension shown in Figure 5.2(b) at various strain rates $\dot{D}_{33}$ (glass fiber bundles, $\phi = 0.13, r \approx 41, l \approx 25$ mm) [VAS 07].

5.2.3. Influence of the fiber content $\phi$ and aspect ratio $r$ on stress levels

The volume fraction of fiber $\phi$ has a primary effect on the rheology of concentrated fiber suspensions. In most situations, the higher the fiber content, the higher is the suspension stress levels. This increase is different from that of the dilute systems: it is highly non-linear and related both to the intensification of long range hydrodynamic interactions (the second and third terms in Equation [5.2], [LIP 88] [SHA 90]) and short range interactions ($f^{\alpha/\beta}$ in Equation [5.5]). The last contribution is dominant in the highly concentrated regime and is directly connected to the high values of the coordination number $\bar{z}$ (see Figure 5.1). An example of the effect of $\phi$ on the rheology of a SMC is shown in Figure 5.6(b): SMC viscosities are polynomial functions of degree 2 of $\phi$ [DUM 03b].

Increasing the fiber aspect ratio $r$ also leads to an important and non-linear increase in the stress levels. The factors that are responsible for this trend are identical to those that have been reported for $\phi$. Figure 5.6(c) shows this effect for the compression rheology of a fiber-reinforced fresh mortar [CHA 10].

5.2.4. Influence of the fiber orientation on stress levels and orientation dynamics

As for $\phi$ and $r$, the fiber orientation plays a primary role on the rheology of fiber suspensions. In general, the more orientated the fibers along the direction of an elongational flow, the higher are the stress levels required to induce the suspension flow. The opposite trend is expected for shear flows. These phenomenological rules are illustrated in Figure 5.2(a) and Figure 5.6(b), respectively. As shown in Figure 5.2(a) for plane strain compression, stress levels recorded for an initially planar random fiber orientation are higher than those observed when fibers are initially orientated perpendicular to the flow direction. The same phenomenon occurs throughout the compressions, i.e., when both microstructures progressively align along the flow direction from random to highly aligned in the first case and from perpendicular to random in the second case. One potential origin of this phenomenon is the long range hydrodynamic interactions that increase as fibers align in a parallel direction to the elongational flow [LIP 88] [SHA 90]. For short range interactions, this is mainly induced by the evolution of the relative positions of
contacting fibers, which induces preferred orientation of vectors \( \mathbf{G}^\alpha \mathbf{G}^\beta \) in equation [5.5]. Conversely, note that for the same reason, the transverse shear viscosity in Figure 5.6(b), in the case of a concentrated suspension with planar random fiber bundle orientation, is much lower than the viscosities that are recorded for compression perpendicular to the mean fiber orientation. This low value would be higher for an isotropic fibrous microstructure, since the fibers out of the shear plane would contribute to better transmit the shear stress through the sample thickness (the out-of-plane component of \( \mathbf{G}^\alpha \mathbf{G}^\beta \) being higher).

If the suspension fiber orientation has an effect on the mesoscale stress levels, mesoscale mechanical loadings also induce strong variations in the fiber orientation: fibers tend to align along the flow direction. However, compared to dilute systems, the alignment is restrained by long range hydrodynamics interactions (as for semi-dilute systems) but also by short range interactions. Thus, deviations of the experimental evolution of the fiber orientation distribution function \( \psi \) with respect to the prediction of the Jeffery’s equation (equation [5.7] with \( D_r = 0 \)) can arise, as shown in Figure 5.6(d). In this figure, a fiber bundle suspensions with initial planar random orientation (similar to that plotted in Figure 5.2(b)) was subjected to a plane strain compression at various constant strain rates \( D_{33} (\dot{\varepsilon}_{22} = 0) \). The orientation of fiber bundles was also measured to estimate \( \mathbf{A} \) and \( \mathbf{A} \) [VAS 07]: the values of \( A_{11} \) and \( A_{1111} \) were then used to compute the evolution of \( A_{11} \) according to equation [5.7], both assuming that bundles of fibers were very slender (\( \lambda \approx 1 \)) and \( D_r = 0 \): \[
\frac{\partial}{\partial t} A_{11} = 2(A_{11} D_{11} - A_{1111} D_{11}) \quad [5.13]
\]

Figure 5.6(d) shows that the last equation systematically overestimates the reorientation that was experimentally measured.

5.2.5. Migration phenomena

Migration phenomena such as those shown in Figure 5.3(b) for a biosourced concentrated fiber suspension are also known to occur during the forming processes of other fiber-reinforced polymer composites materials [HOJ 87] [YAG 95] [DWE 00] [DAN 05] [DUM 05b] [LE 08] [ORG 08b]. These phenomena are detrimental for the end-use properties of produced parts [DAN 05]. Thus, for some flow conditions and some fibrous architectures of suspensions that are still not well understood and characterized, the suspending fluid is expelled from the fibrous network which is in turn consolidated, \( i.e. \), densified. As a rule of thumb, migration phenomena appear for suspending fluids that exhibit a low viscosity, fibrous networks that exhibit a high permeability and that are stiff because of their high entanglement and their high coordination number \( \bar{z} \). Low strain rates and low interstitial fluid pressure are also prone to induce strong migration phenomena. For
example, Figure 5.7 illustrates the role of the compression strain rate on the fluid migration that was observed during the plane strain compression of a standard GMT [ORG 08b]. This figure shows that the sensitivity of the fluid migration to the strain rate is pronounced, and also depends on the actual compression strain (Figure 5.7(a)) as well as on the sample size (Figure 5.7(b)).

![Figure 5.7](image)

**Figure 5.7** – (a) Lubricated plane strain compression of a GMT (glass fiber bundles, $\phi_0 = 0.33, r \approx 82, l \approx 50$ mm) at various compression strain rates $D_0$ at various compression elongations $\lambda_f = h_f/h_0$ (a) and various initial in-plane lengths $L_0$ of the samples. The graphs show the evolutions of the final fiber content in the samples as a function of the normalized abscissa $x_1/L_0$ of compressed samples [ORG 08b].

### 5.3. Microstructure and micromechanics

#### 5.3.1. Microstructure imaging and modelling

Both the fibrous architecture of concentrated fiber suspensions and the fiber geometry play leading roles for the rheology of these media, and directly affect the stress contributions $\overline{\sigma}^m$, $\overline{\sigma}^f/m$, and $\overline{\sigma}^f$ that are defined in equation [5.2]. For example, the fibrous architecture and the fiber geometry govern (i) the fiber coordination number $\bar{Z}$, i.e., the number of fiber–fiber contacts per unit volume $n_c = \bar{Z}/2$ and the set $C$ of the $N_c = n_cV$ connections in $V$, (ii) and the relative positions $G^\alpha G^\beta$ of contacting fibers $\alpha$ and $\beta$, but also (iii) the corresponding contact surfaces $S_{\alpha/\beta}$ that affect contact forces $f^{\alpha/\beta}$. Therefore, it is crucial to thoroughly characterize the fibrous architectures of these suspensions.
Figure 5.8 – Gray scale (a) and segmented (b) 3D images of a model fiber bundle suspension (glass fiber bundles, \( \phi_0 = 0.13, r \approx 21, l \approx 13 \text{ mm} \)) obtained after scanning the sample using X-ray microtomography and standard X-ray absorption imaging mode. Zooms (c-e) in the same suspension performed using a multi-resolution imaging mode that enables a suitable observation of a bundle-bundle contact zone [GUI 12b].

For that purpose, several experimental studies used 2D observation techniques of to analyze the fibrous architectures and the flow-induced evolution of model dilute and semi-dilute suspensions [FOL 84] [PET 98] [PET 00] [YAS 02]. These studies give valuable information for the validation of suspension rheological models. Fewer studies addressed the case of concentrated suspensions [DUM 07b] [VAS 07], which hinders both the development and the validation of the micromechanical hypotheses of the aforementioned multiscale rheological models. For example, by studying the compression flow of the fiber bundle suspensions shown in Figure 5.2(a), the evolutions of the bundle orientation, the bending and the flattening of fibers that was induced by the suspension flow was analyzed [DUM 07b] [VAS 07]. In particular, as for semi-dilute systems, the predictions of Jeffery-based equation were shown to overestimate the experimental trends (see Figure 5.6). However, the microstructure analyses were restricted to 2D observations. Thus the results gained from these studies remained limited.
More recent studies were performed 3D observations using either Optical Coherence Tomography [SAA 14] or X-ray microtomography. The latter [BAR 06] is particularly appropriate for the 3D analysis of the microstructure of heterogeneous materials with micron to centimeter heterogeneities such as fibers in fibrous materials [MAS 06] [ROL 07] [BAD 08] [WEG 12]:

- Using appropriate X-ray imaging mode and segmentation technique of the 3D images to analyze the fibrous phase (for instance, see Figure 5.8(a,b)), a first set of interesting observations can be made. For example, results obtained using 2D techniques for a model transparent fiber bundle suspensions [DUM 07b] were confirmed and also completed for an opaque industrial SMC [LE 08] using the phase contrast X-ray imaging mode: the spatial distribution of fibers was quantified to gauge migration phenomena, the bundle orientation of the suspension was also assessed, and the flattening of bundles that was induced by the suspension flow was also measured. This flattening phenomenon is not taken into account in current multiscale rheological models for fiber bundle suspensions. However, this phenomenon could induce an increase in the bundle–bundle contact areas, and, consequently an increase in the contact forces $f_{\alpha/\beta}$. For similar model fiber bundle suspensions, using a multi-resolution X-ray imaging mode, the geometry of contact surfaces was also finely investigated (see Figure 5.8(c–e)). This analysis showed that the contact surfaces $S_{\alpha/\beta}$ can be considered to have a rhombus shape that can be expressed as follows [GUI 12b]:

\[
S_{\alpha/\beta} \approx \frac{d_{\text{max}}}{\|p_\alpha \otimes p_\beta\|} \quad [5.14]
\]

where $d_{\text{max}}$ is the principal (largest) dimension of the bundle cross sections.

Figure 5.9 – Examples of images of concentrated fiber and fiber bundle suspensions where a specific algorithm was used to detect and identify fibers (a), fiber bundles (b) and their contacts using 3D images of concentrated fiber or fiber bundle suspensions [VIG 13].
In order to extract automatically from 3D images further relevant microstructure descriptors, additional dedicated image analysis subroutines are necessary to detect and to identify individual fibers and fiber-fiber contacts. For fibers with circular cross sections, this can be performed for 3D segmented images (i) computing the Euclidian distance map on the fibrous phase, (ii) thresholding the resulting map to isolate fibers, (iii) skeletonizing and smoothing the as-thinned fibers to obtain their centerline and then (iv) finding and characterizing fiber-fiber contact surfaces from the distances between the fiber centerlines [LAT 11] [ORG 12b]. For other fibers or fiber bundles, individualizing fibers and fiber-fiber contacts is more complicated. Using both relevant information obtained using a non-Euclidian distance map computed on the fibrous phase [ALT 09] and additional image analysis operations [VIG 13], fibers (fiber bundles) and their contacts were labelled in complex fibrous media, as illustrated in Figure 5.9(a-b). Then, in all cases, this labelling operation enables a fine characterization of the properties of the fiber phase: the number of fibers $N_f$ contained in a volume $V$, the fiber positions, orientations and local curvatures, the number of fiber-fiber contacts $N_c$, the coordination number $z$. 

Figure 5.10 – (a) centerlines (blue) and contact orientation vectors $\mathbf{n}^{\alpha/\beta}$ (red) of the fibrous structure shown in Figure 5.9(a). Unit spheres and corresponding orientation tensors showing the fiber orientation (b) and contact orientation (c) of the fibrous structure (a) [ORG 12b].
the orientation \( \mathbf{n}_{\alpha/\beta} \) and the surface of contacts \( S_{\alpha/\beta} \) can be calculated. Figure 5.10 shows resulting discrete representations of the fiber orientation distribution \( \psi \), fiber orientation tensors \( \mathbf{A} \) and \( \mathbf{A}_\alpha \):

\[
\mathbf{A} = \frac{1}{N_f} \sum_{\mathbf{p}} \mathbf{p}^\alpha \otimes \mathbf{p}^\beta. \tag{5.15}
\]

\[
\mathbf{A}_\alpha = \frac{1}{N_f} \sum_{\mathbf{p}} \mathbf{p}^\alpha \otimes \mathbf{p}^\alpha \otimes \mathbf{p}^\alpha \otimes \mathbf{p}^\alpha. \tag{5.16}
\]

contact orientation tensors \( \mathbf{B} \) and \( \mathbf{B}_\alpha \):

\[
\mathbf{B} = \frac{1}{N_c} \sum_{\mathbf{n}} \mathbf{n}_{\alpha/\beta} \otimes \mathbf{n}_{\alpha/\beta}. \tag{5.17}
\]

\[
\mathbf{B}_\alpha = \frac{1}{N_c} \sum_{\mathbf{n}} \mathbf{n}_{\alpha/\beta} \otimes \mathbf{n}_{\alpha/\beta} \otimes \mathbf{n}_{\alpha/\beta} \otimes \mathbf{n}_{\alpha/\beta}. \tag{5.18}
\]

Notice that it is also possible to estimate interaction tensors proposed in some rheological models [DJA 05] [FER 09] [NAT 13]:

\[
\mathbf{C} = \frac{1}{N_f} \sum_{\mathbf{p}} \sum_{\mathbf{p}} \mathbf{p}^\alpha \otimes \mathbf{p}^\alpha \|\mathbf{p}^\alpha \times \mathbf{p}^\beta\| \tag{5.19}
\]

\[
\mathbf{C}_\alpha = \frac{1}{N_f} \sum_{\mathbf{p}} \sum_{\mathbf{p}} \mathbf{p}^\alpha \otimes \mathbf{p}^\alpha \otimes \mathbf{p}^\alpha \otimes \mathbf{p}^\alpha \|\mathbf{p}^\alpha \times \mathbf{p}^\beta\|. \tag{5.20}
\]

These useful experimental data can also be analyzed to validate microstructure models of the literature, which are necessary to build most of the multiscale analytical rheological models. Microstructure models are often built using excluded volume theories. Among them, the tube model is frequently used for concentrated fiber suspensions [DOI 78] [RAN 91] [TOL 93] [GUI 12b]. Assuming (i) that fibers are straight and homogeneously distributed in space, and (ii) that fibers can overlap (soft core assumption), the tube model gives an estimation of the average coordination number \( z \). For fibers with identical circular cross section and length, from the knowledge of the fiber content and orientation, the tube model describes the average coordination number \( \bar{z} \) as follows:

\[
\bar{z} = 4\phi \left( \frac{2}{\pi} \Phi_1 + \Phi_2 + 1 \right), \tag{5.21}
\]

where the orientation functions \( \Phi_1 \) and \( \Phi_2 \) are expressed as:

\[
\Phi_1 = \frac{1}{N_f} \sum_{\mathbf{p}} \|\mathbf{p}^\alpha \times \mathbf{p}^\beta\| \quad \text{and} \quad \Phi_2 = \frac{1}{N_f} \sum_{\mathbf{p}} \sum_{\mathbf{p}} |\mathbf{p}^\alpha \cdot \mathbf{p}^\beta|. \tag{5.22}
\]
Equation [5.21] was modified for fiber bundles with planar fiber bundle orientation, by replacing the expression of the aspect ratio \( r = d/l \) by \( r = d_{\text{max}}/l \) [GUI 12b]. Thus, the use of 3D images of fiber or fiber bundle suspensions enables the relevance of equation [5.21] to be demonstrated for concentrated networks of nearly straight fibers or fiber bundles with planar orientations [GUI 12b] [ORG 12b]. Conversely, the tube model was found to give irrelevant predictions for concentrated suspensions with nearly aligned fibers [LAT 11]. However, in this particular case, the predictions of this model were significantly enhanced by relaxing the assumption of spatial homogeneity of the fiber positions (see Figure 5.12(b)).

5.3.2. Micromechanics

As for the microstructures, determining suitable expressions of local forces \( f_{\alpha/m} \) and \( f_{\alpha/\beta} \) involved in equation [5.2] is a critical point to build proper multiscale rheological models. Hence, to characterize and then model contact micromechanics between contacting fibers (or fiber bundles), in several studies, pull-out experiments, consisting of extracting a straight continuous fiber (or fiber bundle) initially embedded in the fiber suspension were performed (see as Figure 5.11(a)).

![Figure 5.11](image)

Figure 5.11 – (a) 3D view inside a model fiber bundle suspension similar to that in Figure 5.8 (a-b), where (i) a continuous fiber bundle (green) inserted in the suspension was extracted during the pull-out tests, and (ii) its contacting fiber bundles (red). (b) Typical curves that show the evolution of the pull-out force \( f_{e} \) as a function as the extracted length of bundle for various fiber contents \( \phi \) at constant extraction velocity \( v_{e} \) of the continuous bundle and constant confining stress \( \sigma_{n} \) [GUI 12b].

These studies were performed using concentrated fibrous suspensions such as industrial GMTs that were reinforced with fibers [SER 99a] or fiber bundles [SER 99b], CMT [CAB 07] and model concentrated fiber bundle suspensions [GUI 12b]. For instance, for GMTs and CMTs, the measured pull-out force
exhibited a velocity independent yield force at very low pull-out velocities. On the contrary, for the model fiber bundle suspensions [GUI 12b], the pull-out force always exhibited a noticeable viscous behavior, following a Carreau-Yasuda’s law of the pull-out velocities. This viscous behavior was also observed for the GMTs and the CMTs at higher velocities, following power-law [SER 99b] or Carreau-Yasuda’s law [SER 99a] [CAB 07]. In all cases, pull-out forces significantly increased with the fiber content \( \phi \) (see for instance Figure 5.11(b)): this increase was related to the increase in the coordination number \( \bar{z} \) with \( \phi \) (see equation \[5.21\]). The role of the confining pressure was also investigated, leading to an affine increase in the pull-out forces as a function of the confining pressure [GUI 12b]. Accounting [GUI 12b] or not [SER 99a] [SER 99b] [CAB 07] for hydrodynamic interactions between the extracted fiber or fiber bundle, several expressions of the tangential component \( f_t^{\alpha/\beta} \) of contact force were established. For that purpose, the tube model was used to analyze the experimental results. For example, in the case of a model fiber bundle suspension, Guiraud et al. [GUI 12b] describe the contact force as follows:

\[
 f_t^{\alpha/\beta} = \frac{\mu_0}{e} \left( \frac{d_{\text{max}}}{p^a p^b} \right) \left( 1 + \frac{p}{p_0} \right) \left( 1 + \left( \frac{1}{\gamma_c} \right) \frac{\Delta v_t^{\alpha/\beta}}{e} \right)^{n-1} \Delta v_t^{\alpha/\beta}, \tag{5.23}
\]

where \( \mu_0, \gamma_c, n \) are the constitutive parameters of the Carreau-Yasuda suspending fluid, \( p \) the fluid confining pressure and \( p_0 \) its associated constitutive parameter, \( e \) the equivalent thickness of the suspending fluid sheared in the contact zone at the shear rate \( \left\| \Delta v_t^{\alpha/\beta} \right\| / e \), \( \Delta v_t^{\alpha/\beta} \) being the tangent relative velocity of fiber bundles \( \alpha \) and \( \beta \) at their contact point.

5.3.2. Simultaneous micromechanics and microstructure imaging

The work carried out in [GUI 12b] was interesting since it combined pull-out tests with 3D ex situ observations of the microstructures of the suspensions. The imaging enabled the tube model to be validated. This information was used to extract the contact force [5.23].

The current experimental trends consist of 3D in situ observations of the microstructures, i.e., by coupling simultaneously high resolution 3D imaging and micro-rheology [LAT 11] [WEG 12]. For example, Figure 5.12 shows the compression of a concentrated suspension of quasi-aligned polymer fiber immersed in olive oil that was studied using this approach [LAT 11]. Migration of the fluid occurred because of the low compression strain rate, and the severe consolidation of the fibrous network was analyzed (see Figure 5.12). The evolution of the
compression stress during consolidation was fitted using commonly established models for the packing of dry fibrous materials [VAN 46] [TOL 98a]:

$$\sigma \propto \phi^m - \phi_0^m,$$  \( [5.24] \)

where the exponent \( m \) was high (14.25) and close to those observed for this type of fibrous architectures. The microstructure analysis showed that for these types of suspensions: (i) the fiber orientation almost remained invariant, (ii) the fiber local curvature increased with the consolidation of the fibrous network, (iii) the coordination number \( Z \) increased during the consolidation (because this phenomenon is associated to an increase in \( \phi \)), but its value was lower than the prediction of the tube model \([5.21]\), (iv) the contact orientation progressively aligned along the compression direction, (v) and the contact length and deformation decreased and increased, respectively, during compression. Similarly, by studying the shear of fibrous media (without suspending fluid) with moderately slender rods, the orientation dynamics of the rods was studied: dilatancy was shown to occur during the shear of these structures [WEG 12]. Obviously, this type of advanced experimental studies brings very useful information to build relevant constitutive theories.

![Figure 5.12](image)

**Figure 5.12** – Compression of a concentrated suspension of quasi-ordered fibers with 3D in situ observations of its deformation [LAT 11]. (a) Evolution of the compression stress \( \sigma \) as a function of the fiber content \( \phi \) and corresponding evolution of the fibrous microstructure, showing its consolidation (the marks represent the experimental data and the continuous line the prediction of equation [5.24], the inset shows the deviatoric strain as a function of the volumetric strain of the fibrous phase). (b) Corresponding evolution of the fiber coordination number \( Z \) as a function of the fiber content \( \phi \): the marks represent the experimental data, the continuous line the prediction given by equation [5.21], the dotted line the prediction of the tube model without the spatial homogeneity assumption.
5.4. Rheological models: single-phase approaches

5.4.1. Macroscale vs. multi-scale approaches

To model the rheology of concentrated fiber suspensions from the above experimental evidences, two approaches can be followed. First, strategy constitutive equations can be directly deduced from rheological data that were obtained at the macroscale [LIN 97] [LIN 99] [DUM 03b] [GUI 10]. The resulting models are 3D, anisotropic, and describe the non-linear strain rate and strain dependencies of the suspension rheology, as well as the effect of microstructural parameters such as the volume fraction of fibers \( \phi \). For example, this type of macroscale phenomenological approach was used to describe the rheology of highly concentrated fiber bundle suspensions with planar fiber orientation such as SMCs or GMTs, that were considered to be single-phase incompressible transversely isotropic fluid [DUM 03b] [GUI 10]. Thus, the overall suspension viscous stress \( \bar{\sigma}_v \), i.e., the three last terms in equation [5.2], was sought as the gradient of a viscous dissipation potential \( \varpi_v \), which is a function of a scalar equivalent strain rate \( D_{eq} \), with respect to the strain rate tensor \( \tilde{D} \):

\[
\bar{\sigma}_v = \frac{\partial \varpi_v}{\partial \tilde{D}} = \sigma_{eq} \frac{\partial D_{eq}}{\partial \tilde{D}} = \eta_{eq}(D_{eq}) \frac{\partial D_{eq}}{\partial \tilde{D}}, \tag{5.25}
\]

Using the theory of representation of anisotropic tensor functions [BOE 87], the following quadratic form was proposed for \( D_{eq} \):

\[
D_{eq}^2 = \alpha_0 (\tilde{D} : \tilde{D}) + \alpha_1 (M : \tilde{D})^2 + \alpha_2 (\tilde{D} : M) : \tilde{D}, \tag{5.26}
\]

where the \( \alpha_i \)'s are rheological functions that depend on the fiber content \( \phi \), and \( M = e \otimes e \) is a structure tensor characterizing the transverse isotropy of the suspension, of normal axis \( e \). The equivalent viscosity \( \eta_{eq} \) is a scalar function of \( D_{eq} \), e.g. a power-law function [DUM 03b]. Figure 5.13 shows that this model allows a nice fit of flow stresses of SMCs for various mechanical loadings and fiber contents.

To model the strain hardening observed in some systems (for example, see Figure 5.5), note that \( \eta_{eq} \) can also be expressed as a scalar function of an equivalent strain Hencky \( \epsilon_{eq} \) [GUI 10] as follows:

\[
\epsilon_{eq} = \int_{\alpha}^{t} D_{eq} \, dt, \tag{5.27}
\]
Further, this type of mechanical model can be weakly coupled with a set of equations, e.g., equation [5.10] [GUI 10], that are used to predict the evolution of fiber orientation during the suspension flow.

Figure 5.13 – Comparison between experimental results and the prediction of the model [5.25][5.26] for an industrial formulation of SMC for various fiber contents and mechanical loadings [DUM 03b].

The main advantages of these macroscale approaches are their easy identification and implementation into numerical simulation softwares to predict the forming processes of fiber reinforced polymer composites [DUM 07a] [GUI 10]. Their main drawback is that they contain a poor microscale information. On the opposite, the last point is the main benefit brought by the second category of rheological models,
i.e., the so-called multiscale models (although none of them have yet been implemented into simulation softwares). Multi-scale models are built using microstructural and micromechanical considerations which are introduced in upscaling processes [TOL 94] [SUN 97] [FAN 98] [SER 99a] [SER 99b] [DJA 05] [LEC 05] [DUM 09] [FER 09] [NAT 13]. Some of them have already been introduced in this book. Another model dedicated to the rheology of fiber bundle suspensions is presented hereafter [LEC 04] [LEC 05] [DUM 09].

5.4.2. Revisiting the validity domain of multiscale single-phase approaches

As mentioned in the introduction, current multi-scale rheological models systematically lead to the same macroscale description: the suspension is a single-phase and incompressible visco-elastoplastic Cauchy medium. We addressed and revisited the key question of the standard “Cauchy” nature of concentrated fiber suspension, by using both a proper micromechanical description of fiber-fiber interaction and a rigorous upscaling process. This approach does not require a priori assumptions on the properties of the macroscale equivalent continua [LEC 04]. The analysis was carried out for thin fiber bundle suspensions with planar bundle orientation. However, general results deduced from this analysis could be extended to other types of fibrous suspensions. In the following section, we briefly summarize and discuss them.

5.4.2.1. Microstructure and micro-mechanics

The considered suspensions look like standard industrial SMC or GMT formulations. These suspensions can be considered to be sheets of thickness $h$ in which slender fiber-bundles of length $l$ ($h < l$) are immersed in an incompressible non-Newtonian fluid. The X-ray micrographs in Figure 5.8 and Figure 5.9(b) show that fiber bundles form connected networks, i.e., with no isolated bundles or groups of bundles. The fiber-bundles have elliptical cross-section with an area $\pi d_{\text{max}} d_{\text{min}} / 4$. Further, the main axis $d_{\text{max}} \gg d_{\text{min}}$ of the fiber bundle cross sections lies in the plane of the sheets $P \equiv (e_1, e_2)$. X-ray microtomography images show that the mean orientation $\mathbf{p}^{\alpha}$ of a bundle $\alpha$ is mainly contained in $P$ [DUM 07b] [LE 08] [GUI 12b], even if bundles can be slightly bent or wavy around their major axis, because of their slenderness and the high fiber contents. Likewise, as reported by [GUI 12b] and [DUM 07b] [LE 08], bending efforts along the thickness of the sheet and bending of the bundle in $P$ can be neglected as a first approximation. Thus, for the considered suspensions, contact forces in the $e_3$-direction can be neglected. For more concentrated planar systems, this assumption should be reconsidered. The studied systems are also considered to be sufficiently concentrated so that, except the incompressibility constraint brought by the suspending fluid, micro-mechanical efforts are only related to the deformation of the fiber bundle networks [SER 99a] [SER 99b] [GUI 12b]. For the sake of
simplicity, only in-plane flows are considered. Thus, the motion of a bundle \( b \) is contained in \( P \), and characterized both by the in-plane translational velocity of its center of mass \( G^\alpha \):

\[
v^\alpha = v_1^\alpha e_1 + v_2^\alpha e_2.
\]  

[5.28]

and by its angular velocity:

\[
\omega^\alpha = \omega^\alpha e_3.
\]  

[5.29]

In accordance with the experimental observations of [GUI 12b], contacting bundles \( \alpha \) and \( \beta \) interact with viscous efforts. Interaction forces \( f^{\alpha/\beta} \) and moments \( m^{\alpha/\beta} \) occur at the contact zone \( \alpha/\beta \) located at curvilinear abscissa \( s_\alpha \) and \( s_\beta \) on the bundles \( \alpha \) and \( \beta \), respectively. These interactions are supposed to be induced by the deformation of a small amount of a fluid entrapped in these regions. Such a complex situation is assumed to be equivalent to the shearing of thin prism of height \( e \), which in-plane dimensions and orientation depend on \( d_{\text{max}} \) and the relative orientation of the bundles \( \beta_\alpha \) and \( \beta_\beta \). These assumptions are supported by the 3D observations of bundle-bundle bonds (see Figure 5.8). Hence, during the relative motion of the bundles \( \alpha \) and \( \beta \), the entrapped fluid is subjected to (i) a simple shear induced by the difference of in-plane translational velocities

\[
\Delta v^{\alpha/\beta} = v^\alpha - v^\beta + s_\alpha \omega^\alpha \times \beta_\alpha - s_\beta \omega^\beta \times \beta_\beta
\]  

[5.30]

and (ii) a torsion induced by the difference of angular velocities:

\[
\Delta \omega^{\alpha/\beta} = \omega^\alpha - \omega^\beta
\]  

[5.31]

The form proposed for the viscous interaction force \( f^{\alpha/\beta} \) is non-linear and has the same structure than that established experimentally (see equation [5.23]):

\[
f^{\alpha/\beta} = \frac{\mu_0 d_{\text{max}}}{e} \frac{d_{\text{max}}}{||p^\alpha \times p^\beta||} \left( \frac{||\Delta v^{\alpha/\beta}||}{e} \right)^n (n-1) \Delta v^{\alpha/\beta}.
\]  

[5.32]

The viscous interaction moment \( m^{\alpha/\beta} \) are expressed as:

\[
m^{\alpha/\beta} = \frac{\mu_0 \pi d_{\text{max}}^3}{e} \frac{d_{\text{max}}}{n+3} \frac{1}{||p^\alpha \times p^\beta||} \left( \frac{||\Delta \omega^{\alpha/\beta}||}{e} ||p^\alpha \times p^\beta|| \right)^n (n-1) \Delta \omega^{\alpha/\beta}.
\]  

[5.33]

By introducing dimensionless and characteristic quantities using the exponent ”*” and the subscript ”c” (for example, \( s^*_\alpha = s_\alpha / l_c \)), respectively, the microscale equilibrium of bundle \( \alpha \) is defined as follows:

\[
\sum_{c,\alpha} f^{\alpha/\beta*} = 0.
\]  

[5.34]
\[
\mathcal{M} \sum_{\alpha} \mathbf{m}^{\alpha/\beta} = \sum_{\alpha} \mathbf{f}^{\alpha/\beta} \times \mathbf{p}^{\alpha}, \quad [5.35]
\]

where \( C_\alpha \) is the set of contacts of the bundle \( \alpha \), and where \( \mathcal{M} \) is the following dimensionless number:

\[
\mathcal{M} = \pi \frac{n+1}{n+3} \left( \frac{d_{\max}}{\| \mathbf{p}^{\alpha} \times \mathbf{p}^{\beta} \|} \right)^{n+1} \frac{\| \mathbf{\omega}^{\alpha/\beta} \|^{n}}{l_\epsilon \| \mathbf{\omega}^{\alpha/\beta} \|^{n}}, \quad [5.36]
\]

which gauges the magnitude of local interaction moments \( \mathbf{m}^{\alpha/\beta} \) with respect to the magnitude of moments \( \mathbf{s}^{\alpha/\beta} \mathbf{f}^{\alpha/\beta} \times \mathbf{p}^{\alpha} \) induced by interaction forces \( \mathbf{f}^{\alpha/\beta} \). Notice that such a dimensionless number would probably be a key number for other fiber suspensions with other types of interaction forces and moments.

5.4.2.2. Upscaling process

To determine whether the aforementioned microstructure and micromechanics can be homogenized or not, the method of homogenization with multiple scale asymptotic expansions was used [BEN 78] [SAN 80] [AUR 91]. This interesting deterministic upscaling technique was used because it relies upon the possibility of (i) avoiding prerequisites at the macroscopic scale (ii) determining whether the considered heterogeneous media can be homogenized or not (iii) providing the domains of validity of the macroscopic models, if they exist. Due to the discrete nature of the considered fibrous networks, the discrete version of the method was used [MOR 95] [MOR 98] [TOL 98b]. Briefly, the method consists first in introducing the following asymptotic expansions of the discrete bundle linear and angular velocities [LEC 04]:

\[
\mathbf{v}^{\alpha} = \mathbf{v}^{\alpha(0)} + \varepsilon \mathbf{v}^{\alpha(1)} + \varepsilon^2 \mathbf{v}^{\alpha(2)} + \ldots \quad [5.37]
\]

\[
\mathbf{\omega}^{\alpha} = \mathbf{\omega}^{\alpha(0)} + \varepsilon \mathbf{\omega}^{\alpha(1)} + \varepsilon^2 \mathbf{\omega}^{\alpha(2)} + \ldots \quad [5.38]
\]

where \( \varepsilon \) is the scale separation parameter introduced in equation [5.11] and where the velocity fields \( \mathbf{v}^{\alpha(i)} \) and \( \mathbf{\omega}^{\alpha(j)} \) are continuous functions of the same order of magnitude. Such asymptotic expansions are then introduced into the local equilibrium of each bundle [5.35], and problems arising at various \( \varepsilon \)-orders are solved to analyze the existence and the properties of macroscale descriptions. The main results deduced from this analysis are given in the following paragraph.

5.4.2.3. General results and discussion

Depending on the order of magnitude of the dimensionless number \( \mathcal{M} \) with respect to \( \varepsilon \), the studied fibrous networks exhibit two types of macroscale continuous equivalent media [LEC 04]:
For $\mathcal{M} = O(1)$, i.e. when interaction moments $\mathbf{m}^{a/\beta}$ are of the same order of magnitude than moments $s_t f^{a/\beta} \times \mathbf{p}^a$ induced by interaction forces $f^{a/\beta}$, the fibrous networks of the suspension behave as Cosserat fluid [COS 09] [TRU 65] [ERI 68]. Besides, the first order linear or angular velocities do not depend on the considered fiber bundle $\alpha$ so that the fibrous network behaves as a single-phase medium with two kinematical variables:

$$\forall \alpha, \quad \mathbf{v}^{\alpha(0)} = \bar{\mathbf{v}} \quad \text{and} \quad \omega^{\alpha(0)} = \bar{\omega}. \quad [5.39]$$

The two associated momentum balance equations of the equivalent medium are expressed as follow:

$$\nabla \cdot \bar{\sigma}^f = 0, \quad [5.40]$$

$$\nabla \cdot \bar{\kappa}^f - \bar{\xi}^f = 0, \quad [5.41]$$

where the stress tensor $\bar{\sigma}^f(\mathbf{v}, \bar{\omega}, \bar{\theta})$ is no more symmetric, and where $\bar{\kappa}^f(\mathbf{v}, \bar{\omega})$ and $\bar{\xi}^f(\mathbf{v}, \bar{\omega})$ are the couple stress tensor and the micro-stress vector, respectively. These three quantities are defined as follows:

$$\bar{\sigma}^f = \bar{z} \sum_{e} \mathbf{G}^a \mathbf{G}^\beta \otimes f^{a/\beta(0)},$$

$$\bar{\kappa}^f = \bar{z} \sum_{e} \mathbf{G}^a \mathbf{G}^\beta \otimes m^{a/\beta(0)},$$

$$\bar{\xi}^f = \bar{z} \sum_{e} f^{a/\beta(0)} \times \mathbf{G}^a \mathbf{G}^\beta,$$

where the first order forces $f^{a/\beta(0)}$ and moments $m^{a/\beta(0)}$ are expressed as in equations [5.32] and [5.33], replacing the relative linear and angular velocities by their first order approximations $\Delta \mathbf{v}^{a/\beta(1)}$ and $\Delta \omega^{a/\beta(1)}$:

$$\Delta \mathbf{v}^{a/\beta(1)} = \mathbf{v}^{a(1)} - \mathbf{v}^{\beta(1)} + \nabla \bar{\mathbf{v}} \cdot \mathbf{G}^a \mathbf{G}^\beta + \bar{\omega} \times \mathbf{G}^a \mathbf{G}^\beta,$$

$$\Delta \omega^{a/\beta(1)} = \omega^{a(1)} - \omega^{\beta(1)} + \nabla \bar{\omega} \cdot \mathbf{G}^a \mathbf{G}^\beta.$$  

In the case of microscale power-law interactions (see equations [5.32] and [5.33]), the homogeneity of degree $n$ of $\bar{\sigma}^f$, $\bar{\kappa}^f$ and $\bar{\xi}^f$ with respect to the macroscale kinematical variables is also proved so that $\forall \lambda$:

$$\bar{\sigma}^f(\lambda \nabla \bar{\mathbf{v}}, \lambda \bar{\omega}) = \lambda^n \bar{\sigma}^f(\mathbf{v}, \bar{\omega}),$$  

$$\bar{\kappa}^f(\lambda \nabla \bar{\omega}) = \lambda^n \bar{\kappa}^f(\mathbf{v}, \bar{\omega}),$$  

$$\bar{\xi}^f(\lambda \nabla \bar{\mathbf{v}}, \lambda \bar{\omega}) = \lambda^n \bar{\xi}^f(\mathbf{v}, \bar{\omega}).$$
For $\mathcal{M} = \mathcal{O}(\varepsilon)$ or $\mathcal{M} = \mathcal{O}(\varepsilon^2)$, i.e., for interaction moments that are one or two orders of magnitude lower than moments induced by interaction forces, the fibrous network of the suspension is a standard Cauchy fluid (i) with a standard momentum balance, (ii) for which the first order velocity field $\mathbf{v}$ does not depend on the considered bundle (as in equation [5.39]), (iii) with a symmetric stress tensor $\mathbf{\tilde{\sigma}}^f$ that is an homogeneous function of the macroscale strain rate tensor $\mathbf{D}$:

$$\forall \lambda, \mathbf{\tilde{\sigma}}^f(\lambda \mathbf{D}) = \lambda^n \mathbf{\tilde{\sigma}}^f(\mathbf{D}).$$  \hspace{1cm} [5.50]

The expression of $\mathbf{\tilde{\sigma}}^f$ is identical to that of equation [5.42]. However, the first order interaction force $\mathbf{f}^{\alpha/\beta(0)}$ is different because of the expression $\Delta \mathbf{v}^{\alpha/\beta(1)}$. For $\mathcal{M} = \mathcal{O}(\varepsilon^2)$, $\Delta \mathbf{v}^{\alpha/\beta(1)}$ is defined as follows:

$$\Delta \mathbf{v}^{\alpha/\beta(1)} = \mathbf{v}^{\alpha(1)} - \mathbf{v}^{\beta(1)} + \nabla \mathbf{v} \cdot \mathbf{G}^a \mathbf{G}^b + s_\alpha \mathbf{w}^{\alpha(0)} \times \mathbf{p}^\alpha - s_\beta \mathbf{w}^{\beta(0)} \times \mathbf{p}^\beta$$ \hspace{1cm} [5.51]

Note again that the above results have been established without any a priori assumptions stated at the macroscale. One may expect that they are not restricted to fiber bundle suspensions with planar fiber orientation, and that they can be extended to a larger set of concentrated fiber suspensions, i.e., those for which non negligible interaction moments are likely to occur. These results show that in some flow situations the rheology of concentrated fiber suspensions exhibit a non-standard mechanics because of the mechanics of their fibrous networks. Such situations could probably occur in flow zones where the suspensions are curved, twisted and where the scale separation is poor, as illustrated in Figure 5.2(b). In this case, the existing rheological models would thus fail to predict the suspension flow accurately: a new class of Cosserat rheological models such as the first type of model shown above should have to be developed.

### 5.4.3. Application to fiber bundle suspensions

In flow zones where the suspension rheology can be modeled by a standard Cauchy formalism, the second type of aforementioned model was used to model the rheology of concentrated fiber bundle suspensions such as SMCs [LEC 05] [DUM 09]. For that purpose, representative elementary volumes (REVs) of the fiber bundle suspension were first generated, using a generation procedure consistent with the predictions given by the tube model. One of these REVs is shown in Figure 5.14(a). If long range hydrodynamic interactions are sufficiently weak, the macroscale stress of the suspensions [5.2] can be recasted in the following semi-analytical form:
\[
\sigma \approx -p\delta + \frac{4\mu(2z_1 + z_2 + 1)}{\pi e^3 N_c} \sum_{\ell} \frac{||\Delta v^{\alpha/\beta(1)}||^{\ell-1}}{||\mu^{\alpha/\beta}||} \cdot G^{\alpha} G^{\beta} \otimes \Delta v^{\alpha/\beta(1)} [5.52]
\]

**Figure 5.14** – (a) Typical Representative Elementary Volume showing the centerlines (blue) and the bundle-bundle contacts (green) of a concentrated fiber bundle suspension similar to a SMC. (b-e) Comparison between experimental results and the predictions of the multiscale model of equation [5.52] during the simple and plane strain compressions of an industrial SMC. (b) Evolution of the experimental and numerical axial \(\sigma_{33}^{ps}\) and lateral \(\sigma_{22}^{ps}\) stresses, and the numerical component \(A_{11}\) of the fiber orientation tensor \(A\) as a function of the compression strain \(\varepsilon\). Evolution as a function of the fiber bundle content \(\phi\) of the axial (c-d) and lateral (e) viscosities that were observed and predicted during the simple (c) or plane strain (d-e) compressions of SMCs [LEC 05].

where the relative velocities \(\Delta v^{\alpha/\beta(1)}\) are given by equation [5.51] in the studied cases \(M = O(e^2)\). This form clearly shows the role of the fibrous microstructure and the rheology of the contact zones on the macroscale suspension stress. Additional assumptions on the form \(\Delta v^{\alpha/\beta(1)}\), e.g. affinity of the velocity of the center of mass \(G^a\) with the macroscale velocity gradient \(\bar{\nabla} \bar{V}\), would lead to an
analytical expression of the last equation. To quantitatively estimate the macroscale suspension stress, another method was used [LEC 05] [DUM 09]. It consists in numerically solving (using a discrete elements code) the self-equilibrium of the REVs when they are subjected to given macroscale velocity gradients $\vec{v}$, i.e. solving the set of non-linear equilibrium equations at the first order to compute the $N_f$ linear and angular velocities $\vec{v}^{a(1)}$ and $\vec{w}^{a(0)}$ involved in equation [5.51].

Doing so, it was possible to model the rheology of SMC, see Figure 5.14 [LEC 05]: this model allows a nice fit of stress-strain curves and well captures the effect of the fiber bundle content on the suspension flow stress levels, regardless of the considered in-plane mechanical loading. Note also that the model predictions fail if the fiber bundle content is below 0.1: this restriction could be relaxed by taking into account long range hydrodynamic interactions in the model.

Discrete element simulations were also used to discuss the relevance of the fiber orientation models of the literature (based on equations [5.7] [5.10]) in the case of Newtonian interaction at bundle-bundle contacts ($n = 1$ in equation [5.23]) [DUM 09]. The following main results were obtained:

- The bundle orientation rate was found to weakly depend on the fiber bundle content $\phi$. This result is in accordance with the experimental trends reported for similar fiber bundle suspensions [VAS 07]. On the contrary, the current fiber orientation and the shape of its orientation distribution lead to significant changes on the orientation rate. In spite of its different micro-mechanics, the predictions of Jeffery’s equation were in accordance with the results of the discrete element simulations. Closure approximations were also shown to yield inaccurate predictions of the orientation rate except for some very particular cases of orientation states. Likewise, models that use hydrodynamic diffusion terms $D_r$ fail to reproduce the results of the discrete element simulations.

- By updating the position and orientation of each fiber bundle of the REVs, microstructures having orientation distributions with sharp orientation peaks were obtained. Surprisingly, similar microstructures could only be obtained by replacing, for each bundle of the REV, the rotation deduced from the simulation by the prediction of the Jeffery’s equation. Thus, considering the complexity of fiber bundle orientation states in concentrated fiber bundle suspensions, the use of equation [5.10] based on orientation tensors $\mathbf{A}$ and their closure approximations of the orientation tensor $\mathbf{A}$ should be avoided to compute accurately the evolution of the fiber bundle orientation. Instead, other cost-efficient strategies should be used to solve equation [5.7], such as some recent decomposition techniques [AMM 06].
These results should be completed for concentrated fiber bundle suspensions with non-linear interactions. This was initiated [DUM 03a] [LEC 05], but has to be further investigated.

5.5. Rheological models: a two-phase approach

Most of the rheological models dedicated to concentrated fiber or fiber bundle suspensions are based on the assumption that the fibrous network \( f \) and the suspending fluid \( m \) exhibit the same macroscale velocity field:

\[
\vec{v}(f) = \vec{v}(m) = \vec{v}.
\]  

[5.53]

Thus, these single-phase models cannot be used for the prediction of the pronounced migration phenomena that are shown in Figure 5.3 and Figure 5.7. Indeed, using this type of approaches, no suitable physical option can be used to induce these effects with the above restricting condition. A proper alternative consists of using two-phase models, e.g., based on the mixture theory. We briefly recall the basis of these models and present their potential use for concentrated fiber suspensions.

5.5.1. General principles of a two-phase model

The theory of mixtures was first developed in a pioneering study by Truesdell and Toupin [TRU 60]. Later, Bowen [BOW 76] established a complete formalism. Within such a theoretical framework, the following basic assumptions are stated:

- The suspension is considered to be the superposition of two continuous media. Each of them represents immiscible phase of the material, i.e. the fiber network \( f \) and the fluid (or matrix) \( m \). Thus, each macroscale material point \( M \) of the mixture is simultaneously occupied by macroscale material points \( M^{(m)} \) and \( M^{(f)} \) of the phases \( m \) and \( f \), respectively.

- Each elementary macroscale volume \( \delta V \) of the mixture (elementary mass \( \delta \tilde{m} \) and density \( \tilde{\rho} = \delta \tilde{m}/\delta V \) ) is simultaneously occupied by the phases \( \phi \) (\( m \) and \( f \) ). The elementary macroscopic mass \( \delta \tilde{m}^{(\phi)} \) occupies an elementary macroscale volume \( \delta V^{(\phi)} \) included in \( \delta V \). Thus, (i) the macroscale \( \tilde{\rho}^{(\phi)} \) and the microscale \( \rho^{(\phi)} \) densities, and (ii) the volume fraction \( \phi^{(\phi)} \) of phase \( (\phi) \) are defined as follows:

\[
\delta \tilde{m}^{(\phi)} = \tilde{\rho}^{(\phi)} \delta V = \int_{\delta V^{(\phi)}} \rho^{(\phi)} \, dV \quad \text{and} \quad \phi^{(\phi)} = \frac{\delta V^{(\phi)}}{\delta V} = \frac{\tilde{\rho}^{(\phi)}}{\tilde{\rho}} \quad [5.54]
\]
- For the sake of simplicity, the suspending fluid and the fibers will be considered to be incompressible media and the suspension will be considered to be saturated, which yields the following saturation condition:

\[
\bar{\phi}^{(m)} + \bar{\phi}^{(f)} = 1 \tag{5.55}
\]

Doing so, only the fiber content \(\bar{\phi}^{(f)}\) will be used and denoted \(\phi\) in the following paragraphs.

### 5.5.2. Mass and momentum balance equations

Taking into account the previous assumptions, the local mass balance equation for a given material point \(M^{(\phi)}\) for each phase \((\phi)\) is:

\[
\frac{D^{(\phi)}}{Dt} \bar{\phi}^{(\phi)} + \bar{\phi}^{(\phi)} \mathbf{v} \cdot \mathbf{v}^{(\phi)} = 0, \tag{5.56}
\]

where \(D^{(\phi)}/Dt\) is the material time derivative following the material point \(M^{(\phi)}\) of velocity \(\mathbf{v}^{(\phi)}\). Summing the last mass balance equations for the suspending fluid and the fibers, and accounting both for the microscale incompressibility of the phases and for the saturation condition [5.55] lead to an incompressibility condition for the mixture:

\[
\mathbf{\nabla} \cdot (\phi \mathbf{v}^{(f)} + (1 - \phi) \mathbf{v}^{(m)}) = 0. \tag{5.57}
\]

The mixture theory defines the concept of partial stress vector \(\mathbf{t}^{(\phi)}\) and partial stress tensor \(\mathbf{\Xi}^{(\phi)}\) for each phase \((\phi)\), writing the total force \(\delta f\) exerted onto the surface element \(\delta S\) as:

\[
\delta f = \delta f^{(f)} + \delta f^{(m)} = (\mathbf{t}^{(f)} + \mathbf{t}^{(m)}) \cdot \mathbf{n} \delta S = (\mathbf{\Xi}^{(f)} + \mathbf{\Xi}^{(m)}) \cdot \mathbf{n} \delta S, \tag{5.58}
\]

where \(\mathbf{n}\) is the outward normal vector to \(\delta S\). Thus, (i) assuming that each phase behaves as a standard Cauchy medium, and (ii) neglecting both dynamic effects and external volumetric forces, the first momentum balance for each phase is expressed as follows:

\[
\mathbf{\nabla} \cdot (\phi \mathbf{\Xi}^{(f)}) - \mathbf{\Pi}^{(f/m)} = \mathbf{0}, \tag{5.59}
\]

\[
\mathbf{\nabla} \cdot \mathbf{\Xi}^{(m)} + \mathbf{\Pi}^{(f/m)} = \mathbf{0}, \tag{5.60}
\]

where \(\mathbf{\Pi}^{(f/m)}\) represents a volumetric momentum exchange exerted by the fibrous phase onto the suspending fluid.
5.5.3. Constitutive relations

5.3.3.1. Stresses \( \mathbf{\sigma}^{(f)} \) and \( \mathbf{\sigma}^{(m)} \)

The mixture theory offers a sufficiently large framework to propose suitable expressions for the partial stresses \( \mathbf{\sigma}^{(f)} \) and \( \mathbf{\sigma}^{(m)} \) [BOW 76]. When the phases are incompressible at the local scale, partial stresses can be decomposed as follows:

\[
\mathbf{\sigma}^{(f)} = -\phi p_i \mathbf{\delta} + \mathbf{\sigma}^{(f)e},
\]

\[
\mathbf{\sigma}^{(m)} = -(1 - \phi) p_i \mathbf{\delta} + \mathbf{\sigma}^{(m)e},
\]

where \( p_i \) is the interstitial fluid pressure, and where \( \mathbf{\sigma}^{(f)e} \) and \( \mathbf{\sigma}^{(m)e} \) are extra stresses contributions induced by the fibrous network and the suspending fluid, respectively. Various expressions can be adopted for the extra stresses using results from macroscale rheological experiments or multiscale analyses. For example, based on experimental data obtained for the polymer matrix of SMCs or GMTs, the following simple phenomenological form was established for \( \mathbf{\sigma}^{(m)e} \) [DUM 05a] [DUM 05b]:

\[
\mathbf{\sigma}^{(m)} = 2(1 - \phi)\mu(\dot{\gamma}_{eq}^{(m)}) \mathbf{D}^{(m)},
\]

where \( \mu(\dot{\gamma}_{eq}^{(m)}) \) is the suspending fluid viscosity, and where the shear strain rate \( \dot{\gamma}_{eq}^{(m)} \) is defined as:

\[
\dot{\gamma}_{eq}^{(m)^2} = 2\mathbf{D}^{(m)}: \mathbf{D}^{(m)},
\]

Similarly, a straightforward extension of the single-phase macroscale model of equations [5.25] and [5.26] could also be used for \( \mathbf{\sigma}^{(f)e} \), replacing \( \mathbf{D} \) by \( \mathbf{D}^{(f)} \) in these equations [DUM 05a] [DUM 05b]. Still, by replacing \( \mathbf{D} \) by \( \mathbf{D}^{(f)} \), note that, for the same fiber bundle suspensions, the viscous contribution of the multiscale model [5.52] for concentrated suspensions is also an appropriate expression for \( \mathbf{\sigma}^{(f)e} \).

5.3.3.2 Momentum exchange \( \mathbf{\pi}^{(f/m)} \)

As for the partial stresses, in the framework of the mixture theory, a general expression is specified for the momentum exchange \( \mathbf{\pi}^{(f/m)} \) [BOW 76]. In the case of the local phase incompressibility, \( \mathbf{\pi}^{(f/m)} \) is defined as follows:

\[
\mathbf{\pi}^{(f/m)} = -p_i \mathbf{\nabla} \phi + \mathbf{\pi}^{(f/m)},
\]

so that, accounting for equations [5.61] and [5.62], the momentum balance equations [5.59] and [5.60] become:
\[ \mathbf{v} \cdot \mathbf{\sigma}^{(f)e} - \phi \mathbf{v} \mathbf{p}_1 - \mathbf{\Pi}^{(f/m)} = 0, \quad [5.66] \]
\[ \mathbf{v} \cdot \mathbf{\sigma}^{(m)e} - (1 - \phi) \mathbf{v} \mathbf{p}_1 + \mathbf{\Pi}^{(f/m)} = 0, \quad [5.67] \]

The momentum exchange \( \mathbf{\Pi}^{(f/m)} \), which characterizes the flow of the suspending fluid through the fibrous network, is a function of the relative velocity \( \mathbf{v}^r \):

\[ \mathbf{v}^r = \mathbf{v}^{(m)} - \mathbf{v}^{(f)}. \quad [5.68] \]

In the case of Newtonian suspending fluids, the momentum exchange \( \mathbf{\Pi}^{(f/m)} \) is such that the momentum balance [5.67] reduces to the well-known Darcy's law [DAR 56], when the macroscale deformation of both phases and the velocity of the fibrous phase are zero-valued:

\[ \mathbf{\Pi}^{(f/m)} = -(1 - \phi)^2 \mu \mathbf{K}^{-1} \cdot \mathbf{v}^r, \quad [5.69] \]

where \( \mathbf{K} \) is the permeability tensor of the fibrous network. Several analytical estimates of \( \mathbf{K} \) are reported for ordered fibrous media [JAC 86] [BOU 00]. However, the problem is still open for the fibrous microstructures of concentrated fiber suspensions: there is no unified accurate analytical estimate of \( \mathbf{K} \) as a function of \( \phi, r, A \ldots \)

When the suspending fluids are non-Newtonian, the problem is more complicated: the flows of these fluids through fibrous media exhibit severe deviation from the case of Newtonian fluids [BRU 93] [WOO 03] [IDR 04]. Consequently, equation [5.69] is not valid and \( \mathbf{\Pi}^{(f/m)} \) is a non-linear anisotropic function of \( \mathbf{v}^r \).

The literature dedicated to the flow of non-Newtonian fluids through rigid isotropic porous media is rather well-documented [CHH 01]: thus, suitable forms of \( \mathbf{\Pi}^{(f/m)} \) can be used for these media. Unfortunately, much less is known for the flow properties of non-Newtonian fluids through anisotropic fibrous media such as those of concentrated fiber suspensions. This problem was investigated for power-law fluids [AUR 02] [ORG 06] and generalized Newtonian fluids [ORG 07] using the method of homogenization with multiple scale asymptotic expansions [BEN 78] [SAN 80] [AUR 91]. These theoretical and numerical studies showed that, for rigid fibrous networks, \( \mathbf{\Pi}^{(f/m)} \) is the gradient of a macroscale viscous dissipation potential \( \langle \Phi^v \rangle \) (i.e., the volume average of the microscale viscous dissipations of the flowing fluid \( \Phi^v \)), that is a function of a scalar equivalent velocity \( \nu_{eq} \), with respect to the velocity field \( \mathbf{v} = (1 - \phi) \mathbf{v}^r \):

\[ \mathbf{\Pi}^{(f/m)} = \nabla \langle \Phi^v \rangle, \]

\[ \langle \Phi^v \rangle = \int_0^1 \Phi^v(\nu) \, d\nu. \]

Notice that when \( \mathbf{v}^r = 0 \), i.e. when the mixture behaves as a single-phase suspension, the mass balance equation of the mixture [5.57] and the sum of the two momentum balances [5.66] and [5.67] reduce to the mass balance and the momentum balance [5.1], respectively.
The scalar equivalent volumetric drag force \( f_{eq} \) is a non-linear function of \( v_{eq} \) and follows the property of the suspending fluid (e.g. power-law, Carreau-Yasuda, regularized Bingham models). \( f_{eq} \) is closely linked with the microscale flow of the suspending fluid of viscosity \( \mu(\dot{\gamma}) \) through the fibrous network. When the fluid is sheared at a characteristic local shear strain rate \( \dot{\gamma}_c = \frac{v_c}{l_c} \), \( v_c \) and \( l_c \) being the characteristic microscale velocity and thickness of the sheared suspending fluid, respectively, \( f_{eq} \) is expressed as follows [ORG 07]:

\[
f_{eq} = \frac{1}{l_c} \mu \left( \frac{v_c}{l_c} \right) \frac{v_c}{l_c} = \frac{1}{l_c} \mu \left( \frac{v_{eq}}{\phi_c^{(m)} l_c} \right) \frac{v_{eq}}{\phi_c^{(m)} l_c}
\]

In the last expression, \( \phi_c^{(m)} \) represents the characteristic volume fraction of fluid effectively contributing to the drag force \( f_{eq} \) and links the characteristic microscale velocity \( v_c \) with the macroscopic equivalent velocity \( v_{eq} \), i.e. \( v_{eq} = \phi_c^{(m)} v_c \) [LOI 08].

In the case of isotropic fibrous networks, the flow of the suspending fluid is isotropic. Thus, \( v_{eq} \) is equal to the Euclidean norm \( \| (\mathbf{v}) \| \). For orthotropic fibrous networks, i.e., the most common situation encountered in the case of concentrated fiber suspensions, \( v_{eq} \) is a function of the following scalar velocity invariants [BOE 87] [AUR 02] [ORG 07]:

\[
V_i = (\mathbf{v}) \cdot \mathbf{e}_i \otimes \mathbf{e}_i \cdot (\mathbf{v}), \quad i = I,II,III,
\]

\[\text{Figure 5.15 -- Typical evolution of the equivalent velocity} \ v_{eq} \ \text{[5.73]-[5.74], i.e. the isodissipation surface, plotted in the invariant velocity space in the case of orthotropic flow of generalized Newtonian fluids, for various values of its constitutive parameters} \ [\text{ORG 07}].\]
where the $\mathbf{e}_i$’s are the normal unit vectors of the orthogonal symmetry planes of the fibrous network, i.e. the principal directions of $\mathbf{A}$. The following form of $v_{eq}$ was then established for these situations [ORG 07]:

$$v_{eq}^m = v_a^m + v_b^m,$$  \[5.73\]

with

$$v_a^m = V_i^m + \left( \frac{V_i}{A} \right)^m, \quad v_b^m = \left( \frac{V_m}{\theta} \right)^m \quad \text{and} \quad m = \frac{m_a V_i + m_b V_m}{V_i^2 + V_m^2}. \quad [5.74]$$

The expression for $v_{eq}$ requires five constitutive parameters to be identified, i.e., $B$, $m_a$, $m_b$ and $m_c$. The evolution of $v_{eq}$ in the invariant velocity space is shown in Figure 5.15 for various characteristic values of these parameters. This expression was shown to properly describe the flow of power-law or Carreau-Yasuda fluids through orthotropic fibrous media [ORG 06] [ORG 07] [LOI 09]. Notice that for a Newtonian suspending fluid, $m_a = m_b = m_c = 2$. In this case, the expression [5.73] of $v_{eq}$ is quadratic and simpler:

$$v_{eq}^2 = V_i^2 + \left( \frac{V_i}{A} \right)^2 + \left( \frac{V_m}{\theta} \right)^2,$$ \[5.75\]

the momentum exchange [5.70] is then reduced to its linear Darcy-like form [5.69], and the principal values $k_i$ of the permeability tensor $\mathbf{K}$ are expressed as:

$$k_i = \phi_c (m) \varepsilon_i^2, \quad k_{II} = \phi_c (m) (A \varepsilon_c)^2 \quad k_{III} = \phi_c (m) (B \varepsilon_c)^2.$$ \[5.76\]

### 5.5.4. Application to migration phenomena in fiber bundle suspensions

To model migration phenomena in fiber bundles suspensions such as GMTs or SMCs, the mass balances for the fibrous phase [5.56] and for the mixture [5.57], and the momentum balances [5.66] and [5.67] were implemented in a specially designed finite elements code [DUM 05a]. The constitutive equations for the partial stress of the suspending fluid were given by equation [5.63], where $\mu (\dot{\gamma}_{eq}^{(m)})$ is a power-law function of $\dot{\gamma}_{eq}^{(m)}$ with a power-law exponent $n^{(m)}$. The constitutive equations for the fibrous phase corresponded to the two-phase version of [5.25] and [5.26] with a power-law exponent $n^{(f)}$. The permeation law was compatible with the general formulation [5.70]. The scalar equivalent drag force $f_{eq}$ [5.71] was a power-law function of $v_{eq}$ with a power-law exponent $n^{(m)}$. The simulation of the plane strain compression of such a mixture allowed the role of the fiber content, the permeability of the fibrous network and the compression strain rate on the migration of the suspending fluid to be studied. In particular, when $\delta n = n^{(m)} - n^{(f)} = 0$, the strain rate had no effect. For the other tested situations, it was found that the lower the
strain rate, the higher was the suspending fluid migration. The same effect was also enhanced with an increase in $\delta n$. These trends are illustrated in Figure 5.16.

Figure 5.16 – Numerical simulations of plane strain compression for various compression strain rates for SMC samples of length $L$ modeled as a two-phase medium. Evolution of the segregation rate $D_{\text{seg}}$ with the imposed axial compression strain rate $D_{33}$, and for different values of the power-law indices pairs $\{n^{(m)}, n^{(f)}\}$: (a) $\{0.58, 0.44\}$, (b) $\{1, 0.44\}$, (c) $\{1, 1\}$.

In this figure, a segregation rate $D_{\text{seg}}$ was introduced to gauge migration phenomena during the compression of the suspension at an axial compression strain rate $D_{33}$:

$$D_{\text{seg}} = \frac{\nabla \psi^{(f)}}{D_{33}} = -\frac{1}{\phi D_{33}} \frac{\text{D}(f)}{\text{D}t} \phi,$$

so that the higher the value of $D_{\text{seg}}$, the higher the migration of the suspending fluid (conversely, a single-phase behavior of the suspension is observed when $D_{\text{seg}} = 0$). The simulation results were also successfully compared to compression experiments achieved with GMTs [DUM 05b].

5.6. Conclusion

In this chapter, we showed that the current modeling framework for highly concentrated suspensions is still mainly inherited from the strategies that have been used for dilute and semi-dilute simple fibrous suspensions. Accordingly, the rheological and orientational models are based on the description of the motion of rigid slender rods in a flowing Newtonian fluid. Note that for semi-dilute suspensions, the randomization effect of long range hydrodynamic interactions between fibers or the effect of sparse fiber-fiber contacts on the fiber orientation evolution can be described by various modifications of the evolution rate of the second order orientation tensor.
The description of the rheology of highly concentrated fiber suspensions is however more complex because of their particular microstructural features. In these suspensions, the number of fiber-fiber contacts is well above the mechanical percolation threshold. Therefore, they have to be considered as consisting of a network of intricate fibers, where fiber-fiber contacts play a dominant role on the rheological response. Hence, the understanding and modeling of the rheology of these suspensions requires the use of appropriate experimental techniques and the development of specific models.

Experimental macroscale studies of the rheology of highly concentrated model or industrial fiber suspensions showed several specific features of their rheology:

(i) Inherent microstructural heterogeneity of these suspensions induces important scale effects that must be taken into account for a proper experimental identification,
(ii) Strong effects of the microstructure (fiber volume fraction, orientation and aspect ratio, presence of clusters),
(iii) Flow-induced microstructural evolutions (reorientation, matrix migration, flocculation),
(iv) Drastic evolution of the suspension behavior in confined zones.

The necessity of microscale characterization studies was also pointed out. We showed that several suitable experimental techniques can provide a precise description of the geometry and of the topology, as well as of their evolution when the suspension is submitted to a macroscopic mechanical loading. In this field, three-dimensional X-ray imaging techniques, associated both to the use of micro-rheometry experimental devices and to automated numerical image analysis algorithms, are particularly promising experimental approaches. Large database on the suspension microstructural properties can be acquired using these techniques. This approach is particularly important for providing multiscale models with relevant experimental data.

Lastly, the pertinence of developing specific multiscale models was shown here. For that purpose, the continuous or discrete methods of homogenization with multiple scale asymptotic expansions were shown to be efficient and rigorous theoretical frameworks. The resulting models can be used in enriched single-phase or two-phase macroscopic rheological models.

If a large amount of works has already been done to gain a better understanding of highly concentrated fiber suspensions at the different scales of interest, several difficult problems still must be investigated. Among them, we would like to mention:
- The problem of the formation and disaggregation of aggregates or flocs. Starting from an initially almost homogeneous fiber network, this problem results in a strong degradation of the final mechanical properties and in a drastic change of the orientational behavior during flow. A possible theoretical framework to tackle this problem was presented in another chapter of this book.

- The integration of enriched microscale behaviors, including for example unilateral contacts, fibers flexibility effects, and non-linear rheology of the matrix.

- The scale effects are often present in practical applications. To account for such effects in continuous models for highly concentrated suspensions, Cosserat-like models, as presented in this chapter, should be further investigated.

References


