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X-RAY MICROTOMOGRAPHY AND PULL-OUT TESTS TO ANALYSE MICROSTRUCTURES AND FIBRE-FIBRE INTERACTIONS IN CONCENTRATED FIBRE SUSPENSIONS

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ABSTRACT: During their processing short fibre reinforced polymer composites behave as non-Newtonian concentrated fibre suspensions, the rheology of which highly depends on both the microstructure of the fibrous networks and deformation micro-mechanisms arising at fibre-fibre contacts. In this work, these two aspects are studied by using model concentrated fibre suspensions made up short glass fibre bundles impregnated in a viscous paraffin gel. X-ray microtomography was used to analyse fibrous microstructures, showing that the studied suspensions exhibit a planar fibre orientation and that the fibre bundle connectivity can be described by the geometrical statistical tube model. Bundle-bundle contacts forces are analyzed by pull-out tests. These tests allow us to quantify the influence of the pull-out velocity, the confining stress and the volume fraction of fibre bundles on the pull-out force. These different results are combined to propose a bundle-bundle contact model.

KEYWORDS: Concentrated fibre suspensions, pull out tests, X-rays microtomography.

INTRODUCTION

Short fibre bundles reinforced polymer composites such as Sheet Moulding Compounds (SMC), Glass Mats Thermoplastics (GMT), Carbon Mat Thermoplastics (CMT)… are extensively used in the automotive and electric industries. These composites usually display a high volume fraction of fibres (typically ranging from 5 to 20%), so that each fibre bundle inside the composite exhibits multiple contact points with its neighbours. This highly affects physical and mechanical properties of these materials. This also drastically affects their rheology during their forming processes (*i.e.* compression or injection mouldings): such composites materials behave as non-Newtonian and highly concentrated fibre suspensions, exhibiting a complex rheology which is largely affected by bundle-bundle interactions occurring in contact zones during the suspension flow. Such interactions constitute the basis of most of rheological models dedicated to these suspensions [1-5]. In order to characterise contact micromechanics between touching fibres or fibre bundles, several authors have carried out pull-out tests [2,6]. These studies were performed using industrial GMT [2] or CMT [6] and they are rather complete. However, the studied fibrous microstructures remain restrained and the influence of the confining stress, which is non negligible during the forming process, was not studied. Furthermore, important *a priori* hypotheses concerning the fibrous architectures were assumed in order (i) to deduce contact forces from pull-out experiments and (ii) to build rheological models for these fibre suspensions.

From these observations, this study completes the aforementioned works [2,6]. For that purpose, various model fibre bundle suspensions exhibiting various fibrous microstructures were processed, and finely characterised by using x-ray microtomography. Therefrom, such suspensions were subjected to pull-out experiments at various confining stresses and pull-out velocities. These different results are combined to propose a bundle-bundle contact model.

MATERIALS AND PULL-OUT DEVICE

The polymer matrix of the studied model fibre suspensions is a transparent paraffin gel (Versagel, Penreco). This matrix saturates fibrous networks made up of industrial glass fibre bundles (Owens Corning) which are generally used for SMC and which are composed of approximately 200 cylindrical glass fibres (diameter d_f =15.4 µm, length l_f = 12 mm). Produced samples are plates of dimensions $L \times l \times h$ = 170 mm × 80 mm × 6 mm). They are solid at a room temperature and exhibit a highly viscous behaviour at 50°C, similar to those displayed by industrial compounds. Their processing provides a very good control of their fibrous microstructure [7], *i.e.* with (i) a reasonable homogeneity of the spatial distribution of bundles, (ii) a prescribed fibre content and (iii) a prescribed overall orientation of the fibrous network. For this study, the generated fibrous networks have a planar random orientation in the principal plane of the plate samples and display fibre contents ϕ ranging from 0% to 13%. As shown from figure 1, please notice that five straight continuous and parallel fibre bundles were inserted during the suspension processing.

Figure 1 also depicts the oversimplified scheme of the specially designed pull-out device. Briefly, a suspension sample is inserted inside a 50°C heated rectangular mould of in-plane dimension $L \times l$ (thus limiting the in-plane flow of the suspension). The upper rectangular plate of the mould (dimension $L \times l$) exerts a prescribed confining force F_n (or a confining normal stress $\sigma_n = F_n/Ll$) which is kept constant during the experiment. A pull-out experiment consists in extracting one of the five straight and continuous fibre bundles at a constant pull-out velocity v_e over a pull-out length L_e. During such an extraction, the pull-out force F_e is recorded. Therefrom, the pull-out force per unit of length $f_e = F_e/L_e$ together with the drag coefficient $\lambda_e = f_e/v_e$ can be estimated. Doing so, various tests have been achieved by following this procedure, with various confining stress σ_n and various pull-out velocities v_e, ranging from 0 to 18000 Pa, and from to 10^{-6} to 10^{-1} m s^{-1}, respectively. Such values were estimated from those encountered by the considered industrial composites during their forming processes.

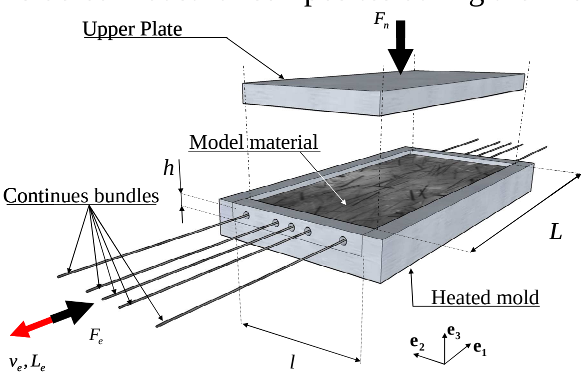

Fig. 1 – Over-simplified scheme of the pull-out device.

MICROSTRUCTURE CHARACTERISATION

The characterisation of the fibrous networks was performed by using x-ray microtomography. Hence, several small samples (dimensions are 10 mm × 10 mm × 6 mm) were cut from the produced plates. They were imaged by using the microtomograph of the ID19 beamline (European Synchrotron Facility Radiation): 1500 x-ray projections were achieved at different orientations of the sample (with a CCD detector, 2048×2048 pixels). After suitable reconstruction and image analysis procedures, it was possible to obtain 3D representations of the fibrous networks, with a voxel size of 7×7×7 μm^3, as shown for example in figure 2(a). From such type of volume, estimations of relevant descriptors of the considered fibrous networks could be obtained:

- The centrelines of the N bundles contained in a scanned volume (N = 150 in figure 2) were detected by using a manual procedure similar to that adopted previously for SMC [9]. It was then possible to gauge the tortuosity of each bundle, defined as length of the bundle centreline divided by the chord joining the two extremities of the same centreline. Doing so, the average value of the tortuosity of the bundles of the studied suspensions was found to be very close to 1 (1.005 in the example of figure 2a). Thus, fibre bundles can be considered as straight bundles and can be ascribed a unique orientation vector: it is then shown that all these vectors nearly lie in the produced plates' midplanes ($\mathbf{e}_1$, $\mathbf{e}_2$), within which they are randomly oriented.
- The analysis of the scanned volumes proved that it was reasonable to assume that fibre bundles exhibit elliptical cross section with major and minor axes of dimensions d_{max} = 0.5±0.007 mm and d_{min} = 0.05±0.007 mm, respectively. Furthermore, it was shown that the major axes mainly lie in the midplane ($\mathbf{e}_1$, $\mathbf{e}_2$) of the plates.
- Therefrom, the reconstruction of each bundle around its centreline was carried out (see figure 2(b)), by assuming constant elliptical cross section (d_{max}, d_{min}). A contact detection algorithm was also developed in order to quantify the average number of bundle-bundle contacts per bundle n_c within the scanned volumes. In the example shown in figure 2, $n_c = 5.7$. This value is very close to the value of 5.8 which is predicted by the statistical tube model [4, 11]. The same trend was observed for other fibrous microstructures, showing the relevance of this model to describe the fibrous connectivity in the case of the considered fibre suspensions.

PULL-OUT RESULTS

Figure 3(a) shows a collection of five f_e-L_e curves showing the pull-out response obtained with the matrix without bundle (ϕ = 0), with a fairly nice reproducibility (≈ ± 10%). A fast increase of the pull-out force f_e is observed until a steady state threshold value f_{ep}. Its corresponding drag coefficient $\lambda_{ep} = f_{ep}/v_e$ is a Carreau type function of the pull-out velocity v_e, as shows the graph of figure 3(c).

Figure 3(b) shows typical f_e-L_e curves obtained during the pull-out with fibre bundles suspensions. After a fast increase of the pull-out force f_e, similar to the one observed in figure 3(a), a decrease of f_e is observed after a peak f_{ep}. It is attributed to the damage of the matrix within bundle-bundle contact zones. Such a decrease diminishes and disappears (i) as the confining pressure σ_n is increased (see figure 3(b)), (ii) as v_e is decreased, (iii) as ϕ is decreased. In addition, whatever the confining stress σ_n and the fibre content ϕ, the drag coefficient λ_{ep} at the peak is a power-law function of the pull-out velocity v_e, the power-law exponent n of which is identical to that observed for the matrix in the power-law regime (see figure 3(c)). Furthermore, we have reported in figure 3(d) the evolution of the drag coefficient λ_{ep} with respect to the fibre content ϕ: whatever the confining stress σ_n, λ_{ep} increases linearly with ϕ. This is directly linked with the increase of the number of bundle-bundle contacts along the extracted continuous bundle. Notice that this figure also underlines a non-negligible contribution of the matrix to the overall pull-out force. Lastly, figure 3(e) proves that the evolution of the drag coefficient λ_{ep} is a linear function of the confining stress σ_n. Such a trend is linked with the increase of the contact pressure and/or the contact areas as the confining stress is increased.

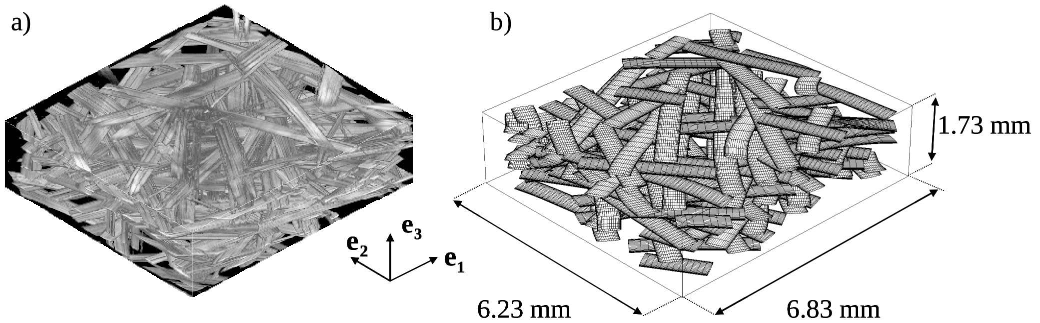

Fig. 2 (a) 3D image of the fibrous network obtained from X-ray microtomography (voxel size of 7×7×7 μm^3, ESRF, ID19) of a model suspension sample ($\phi\in$= 0.133). (b) Reconstruction of all the bundles contained in the volume shown in (a).

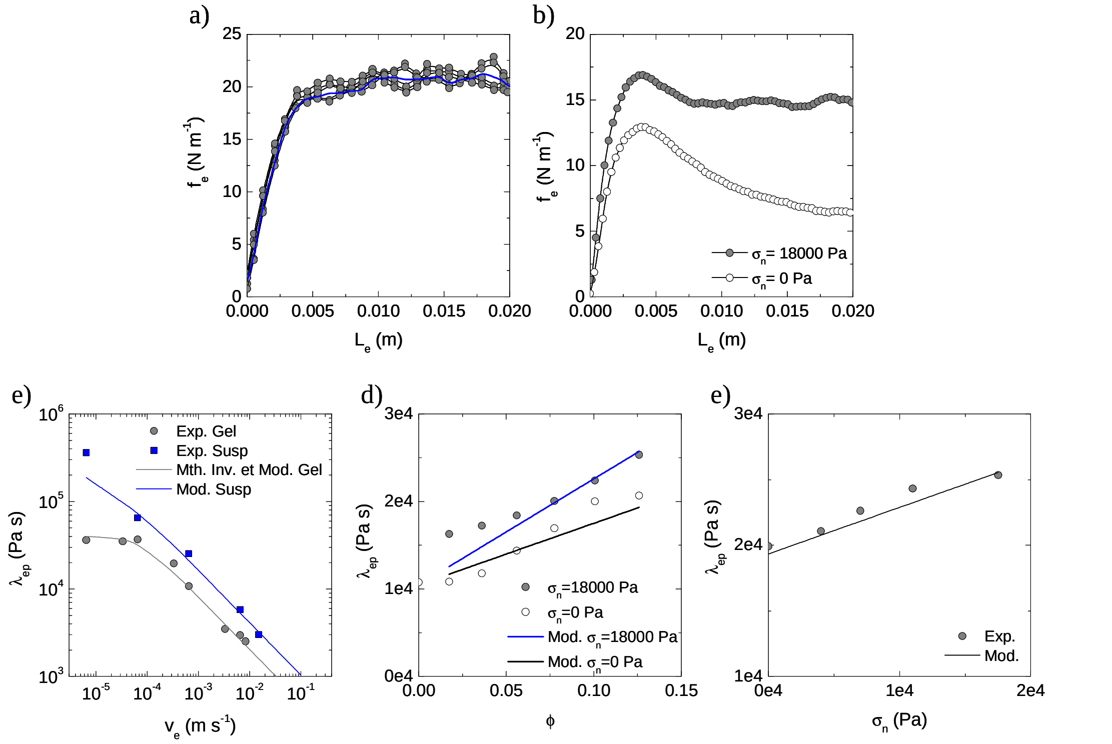

Fig. 3 General aspect of f_e-L_e curves recorded (a) for the matrix alone (v_e = 8.3 10^{-3} m s^{-1}) and (b) for a bundle suspension for two different confining stress σ_n (v_e = 6.5 10^{-4} m s^{-1} and ϕ= 0.133). Evolution of the drag coefficient λ_{ep} with (c) v_e (for the gel σ_n = 0 Pa and the suspension with ϕ = 0.133 and σ_n = 18000 Pa), (d) the volume fraction ϕ of fibres and (e) the confining stress σ_n (with ϕ = 0.133). Solid lines plotted in graphs (c,d,e) are the pull-out model predictions.

DISCUSSION AND CONCLUSION

Results obtained in the two previous sections bring us precious information about the microstructures and deformation micro-mechanisms associated with concentrated fibre bundle suspensions. Firstly, we have proved from x-ray microtomography and image analysis that the bundle-bundle connectivity of the studied suspensions could be reasonably predicted by the simple statistical tube model, validating by the way a strong assumption which is usually and *a priori* stated in rheological models [1-5]. Secondly, from the pull-out experiments, it was also possible to analyse precisely micro-mechanisms arising at the bundle scale during the relative motion of fibre bundles, by quantifying the role of both the confining stress and the extraction velocity, for a wide range of fibre content. If the second effect is already taken into account in microrheological model, the first one is not despite its non-negligible impact. Hence, as a first step towards the proposition of a suitable micromechanical model of bundle-bundle contacts in concentrated bundle suspensions, a simple pull-out model is proposed from the experimental results gained in this work. This model is first based on the capability of the tube model to predict the bundle connectivity in the studied suspension. Therewith, the overall pull-out force is seen as the superposition of two contributions, *i.e.* one contribution accounting for the resistance induced by the matrix (far from contact zones), and another one that reflects the resistance induced by the contacts the C short bundles that touch the continuous bundle (C being directly deduced from the tube model). The first contribution is such that its drag coefficient is a Carreau function of the extraction velocity, in accordance with results observed in figure 3(c). The second contribution involves a drag coefficient that is a linear function of C, the confining normal stress and a power-law function of the pull-out velocity. From the knowledge of the matrix rheology, the geometry, content and orientation of fibre bundles, this model requires only one additional constitutive parameter which gauges the effect of the confining stress. As shown from the continuous lines plotted in the graphs of figure 3, such a simple model allows a fairly nice prediction of pull-out forces.

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