Electronic Supplementary Material (ESI) for Soft Matter.

Supplementary Materials

-

Micro-mechanics of electrostatically stabilized suspensions of cellulose nanofibrils under steady state shear flow

F. Martoïa[1,2,3,4,5], P.J.J. Dumont[1,2,3,**], L. Orgéas[4,5], M.N. Belgacem[1,2,3], J.-L. Putaux[6,7]*

[1]Univ. Grenoble Alpes, LGP2, F-38000 Grenoble, France

[2]CNRS, LGP2, F-38000 Grenoble, France

[3]Agefpi, F-38000 Grenoble, France

[4]Univ. Grenoble Alpes, 3SR, F-38000 Grenoble, France

[5]CNRS, 3SR, F-38000 Grenoble, France

[6]Univ. Grenoble Alpes, CERMAV, F-38000 Grenoble; France

[7]CNRS, CERMAV, F-38000 Grenoble, France

[*]Corresponding author's email: pierre.dumont@insa-lyon.fr

[**]Present address: Université de Lyon, INSA-Lyon, LaMCoS CNRS UMR5259, F-69621 Villeurbanne cedex, France

Appendix S1

For a prolate ellipsoid with an aspect ratio $r' = a/b$ (where a and b are the major and minor axes of the prolate ellipsoid, respectively), the shape factor writes $\lambda = (r'^2 - 1)/(r'^2 + 1)$[1]. For more complicated particles such as the studied NFCs, the estimate of λ is not straightforward[2]. For simplicity sake, as shown in Fig. 8, a first order estimation of λ was used. Hence, the coefficient λ was calculated from the prolate ellipsoid that displays the same inertia axes and moments as NFC i.

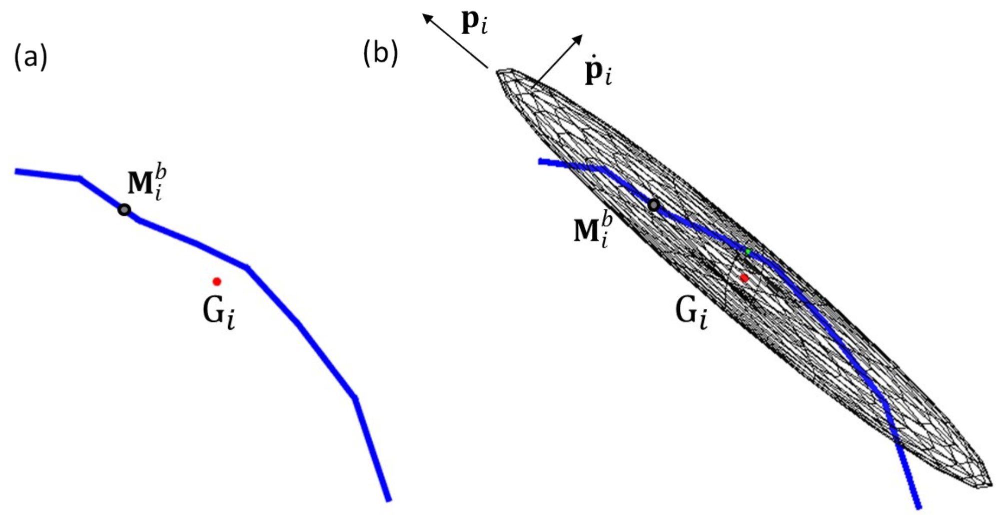

Figure 8. Sketch that shows (a) the NFC i and (b) its associated equivalent prolate ellipsoid with the same inertia axes and moments.

Appendix S2

Each tortuous nanofiber i of diameter d and length l contained n_{seg} straight segments of length l_{seg} that had a random relative misorientation θ_{seg}. Their center of mass G_i was randomly placed in the elementary volume Ω. Following this procedure, various fibrous networks with various nanofiber contents ϕ were generated with random orientation of nanofibers, *i.e.*, the generated microstructures exhibited isotropic fiber orientation with second order orientation tensors $A = \frac{1}{3} e_i \otimes e_i$ (Fig. 5a and Fig. 5c for the corresponding pole figure of orientation). This type of microstructures was chosen because it constituted a reference state, potentially close to that of NFC suspensions at rest (where Brownian motion presumably induces particle dispersion[3,4]).

To mimic microstructures with a pronounced NFC orientation along the flow direction[3] under shear flow, *i.e.*, for $\nabla v = \dot{\gamma} e_1 \otimes e_2$, the evolution of the mean orientation vector p_i of each NFC i was computed up to the steady state overall orientation of the fibrous networks, solving the Jeffery-based model (eqn (17)) with $D_r = C_I \sqrt{2D:D}$ and $C_I = 10^{-3}$[5,6]. For convenience, calculations were done using the expansion in spheric harmonics up to the fourth order[7] of the orientation distribution . Hence, the following approximation was used[8]:

$$\frac{1}{\psi}\frac{\partial \psi(p)}{\partial p} \approx 4\frac{-21A \cdot p + 63p \cdot A : p \otimes p}{3 - 42A : p \otimes p + 63 p \otimes p : A : p \otimes p}, \quad (22)$$

All the simulations were run using Matlab and a suitable time step $\Delta t = \Delta\gamma / \dot{\gamma}$ such that $\Delta\gamma = 0.01$ until reaching a steady state, which was close to a shear strain $\gamma = 100$. Then, the simulation procedure led to orientated fibrous microstructures with $A \approx 0.9 e_1 \otimes e_1 + 0.05\,(e_2 \otimes e_2 + e_3 \otimes e_3)$ (Fig. 5b and Fig. 5d for the corresponding pole figures of orientation). In accordance with the Jeffery-based models, it is worth to note that the resulting fiber orientation only depended on initial orientation state and on the applied shear strain, but not on the shear rate $\dot{\gamma}$.

References

1 G. B. Jeffery, Proc. R. Soc. Lond. Math. Phys. Eng. Sci., 1922, 102, 161–179.

2 F. P. Bretherton, J. Fluid Mech., 1962, 14, 284–304.

3 K. M. O. Håkansson, A. B. Fall, F. Lundell, S. Yu, C. Krywka, S. V. Roth, G. Santoro, M. Kvick, L. Prahl Wittberg, L. Wågberg and L. D. Söderberg, Nat. Commun., 2014, 5:4018.

4 R. G. L. Larson, The Structure and Rheology of Complex Fluids, Oxford University Press, New York, 1 edition., 1998.

5 F. Folgar and C. L. Tucker, J. Reinf. Plast. Compos., 1984, 3, 98–119.

6 S. G. Advani, Flow and Rheology in Polymer Composites Manufacturing, Elsevier, 1994.

7 S. G. Advani and C. L. Tucker. II, J. Rheol., 1987, 31, 751–784.

8 M. Djalili-Moghaddam and S. Toll, J. Non-Newton. Fluid Mech., 2005, 132, 73–83.