Electronic Supplementary Material (ESI) for Soft Matter.

Inelastic behaviour of cellulose microfibril networks

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Supplementary Information

Note 1: Comparison of rheology for smooth and sand-blasted top plate to test for wall-slip effects.

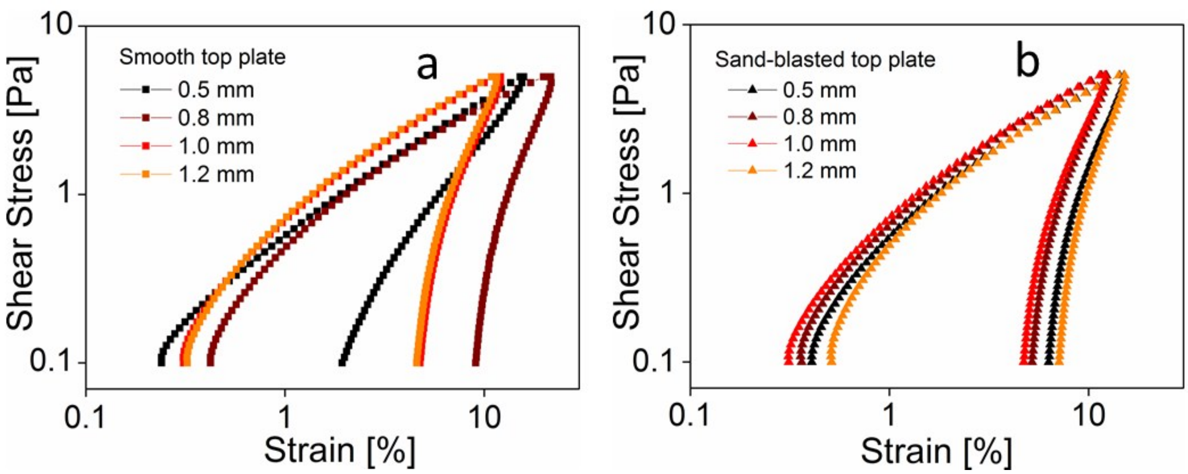

Fig S1: Strain response of ϕ= 0.33 % CMF networks to an increasing and decreasing stress ramp, showing inconsistencies at different gap sizes when (a) a smooth top plate was used. A more uniform response was recorded with (b) a sand-blasted top plate, confirming the absence of wall-slip in this case.

Note 2: Determination of fractal dimension of single CMFs in DMSO.

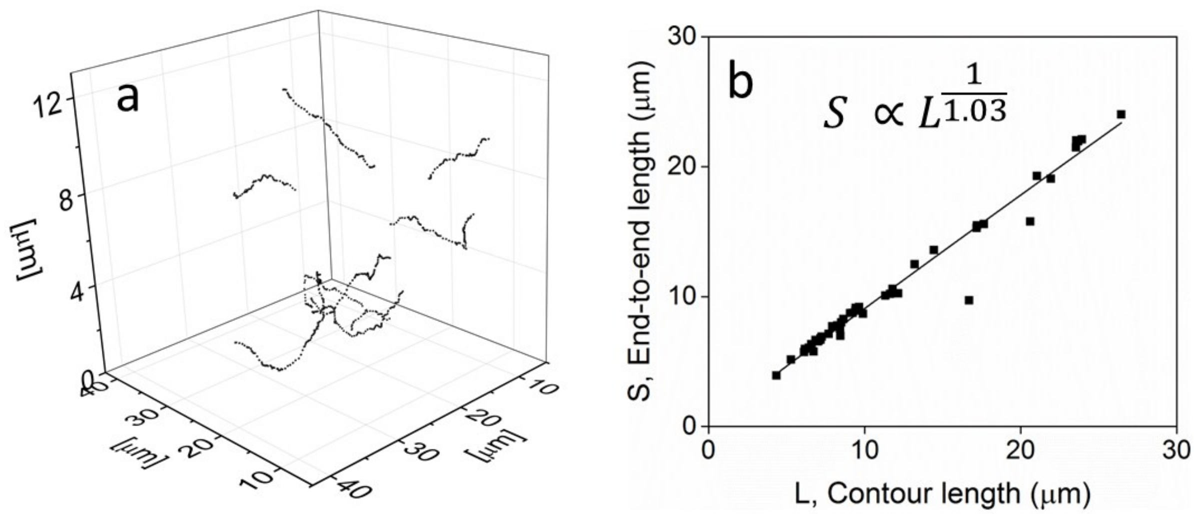

Fig. S2: (a) 3D coordinates of single fibrils dispersed in DMSO traced in confocal image stacks using VAA3D.[1] (b) Esimation of fractal dimension from the length parameters of the traced fibrils (N= 50).

A dilute suspension (with a volume fraction of 0.01%) of CMFs in DMSO was sonicated using a bath ultrasonicator (Branson 8800, 40 kHz, 90 s) to disperse the fibrils. The suspension was then imaged in 3D in a 512 × 512 pixel format with an average voxel size of $80 \times 80 \times 200$ nm^3, using a reflectance confocal microscope (Leica TCS SP8) in resonant scanner mode. We used a 100× NA 1.4 objective (n_{oil} = 1.515). From the volume image, the coordinates (with sub-pixel resolution) of the individual fibrils were obtained by semi-automatic tracing, using VAA3D software (version 3.20).[1] From the fibril coordinate data, both the fibril contour length (L) and end-to-end length (S) were obtained, from which a fractal dimension of 1.03 ± 0.03 was deduced, using the expression $S \propto L^{\frac{1}{D_F}}$.[2]

Note 3: Constant maximum stress cycle using cone-plate geometry

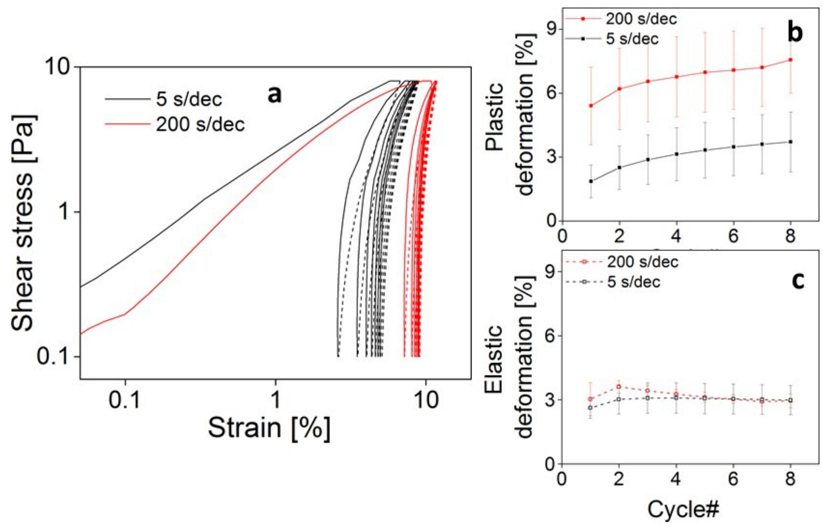

Fig. S3: (a) Strain response of CMF networks (φ= 0.33%) to the application of stress cycles with a constant maximum of 8 Pa in a cone-plate measuring system at two different loading rates as indicated. (b, c) The recorded plastic and elastic component of the deformation with respect to the number of stress cycles.

Note 4: Cyclic constant linear strain ramp measurements

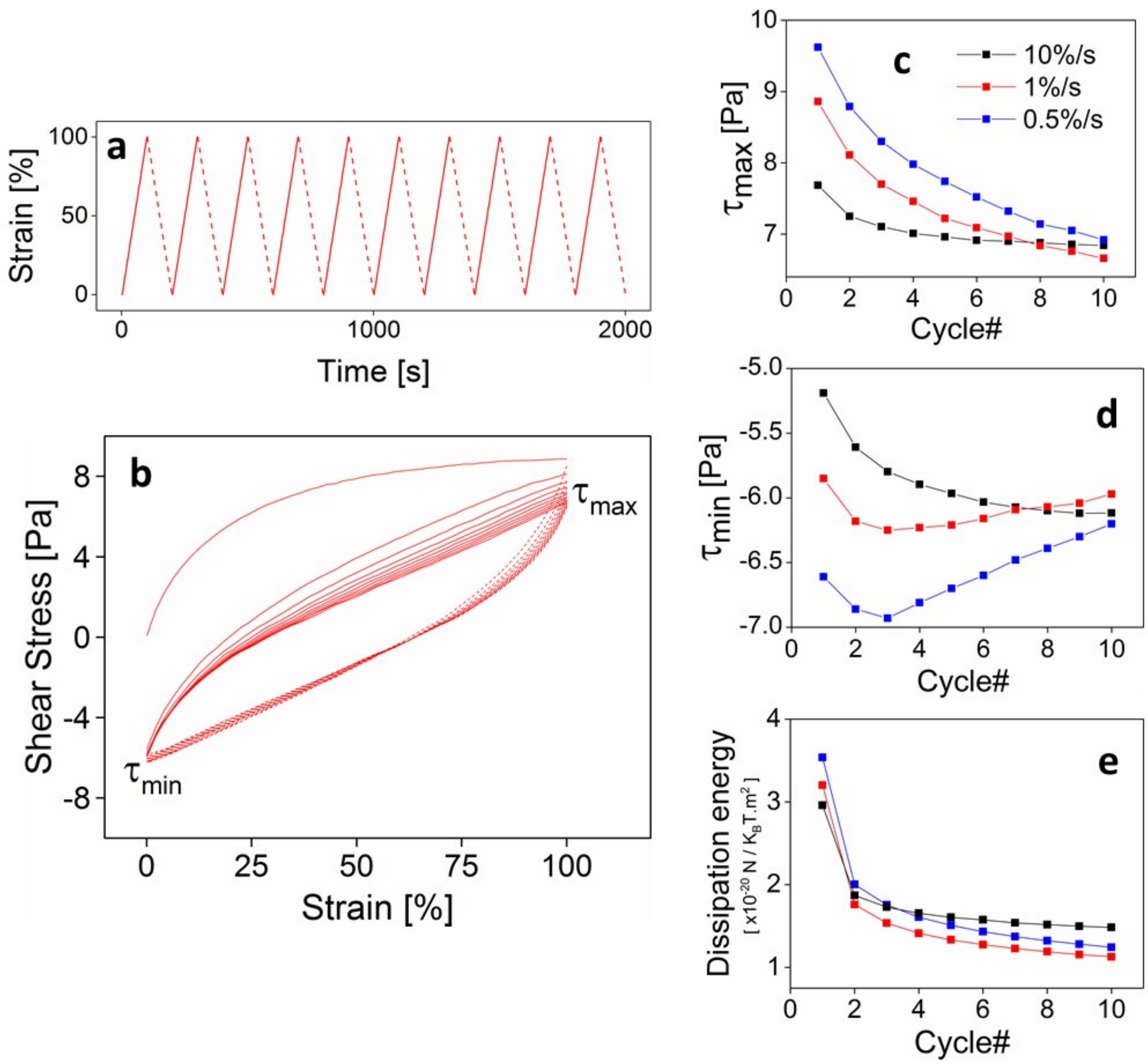

Fig. S4: (a) Constant maximum strain ramp protocol. (b) Stress response of ϕ= 0.33 % CMF network to the applied strain ramp at a strain rate of 1%/s. (c, d) The stress at the maximum applied strain of 100% (τ_{max}) and back at 0% strain (τ_{min}) at the end of every cycle, both shown as a function of strain cycles. (e) Dissipation energy which corresponds to the hysteresis loop area (normalized to k_BT) plotted as a function of the number of applied strain cycles.

References

1. H. Peng, Z. Ruan, F. Long, J. H. Simpson and E. W. Myers, *Nat. Biotechnol.*, 2010, **28**, 348.
2. J.-M. Guenet, *J. Rheol.*, 2000, **44**, 947-960.