

Supporting information

Elastoplastic behavior of anisotropic, physically crosslinked hydrogel networks comprising stiff, charged fibrils in an electrolyte

*Rebecca Östmans,^{1, 2, +} Maria F. Cortes Ruiz,^{1, 2, +} Jowan Rostami,¹ Farhiya Alex Sellman,^{1, 2} Lars Wågberg,^{1, 2} Stefan B. Lindström³ and Tobias Bensefelt,^{1, 4, *}*

¹ Department of Fibre and Polymer Technology, Division of Fibre Technology, KTH Royal Institute of Technology, 100 44 Stockholm, Sweden.

² Wallenberg Wood Science Center, 100 44 Stockholm, Sweden

³ FSCN Research Center, Mid Sweden University, 851 70 Sundsvall, Sweden

⁴ School of Materials Science and Engineering, Nanyang Technological University, 639798 Singapore

⁺ These authors contributed equally to this work

^{*} Correspondence to: bense@kth.se or gustaftobias.b@ntu.edu.sg

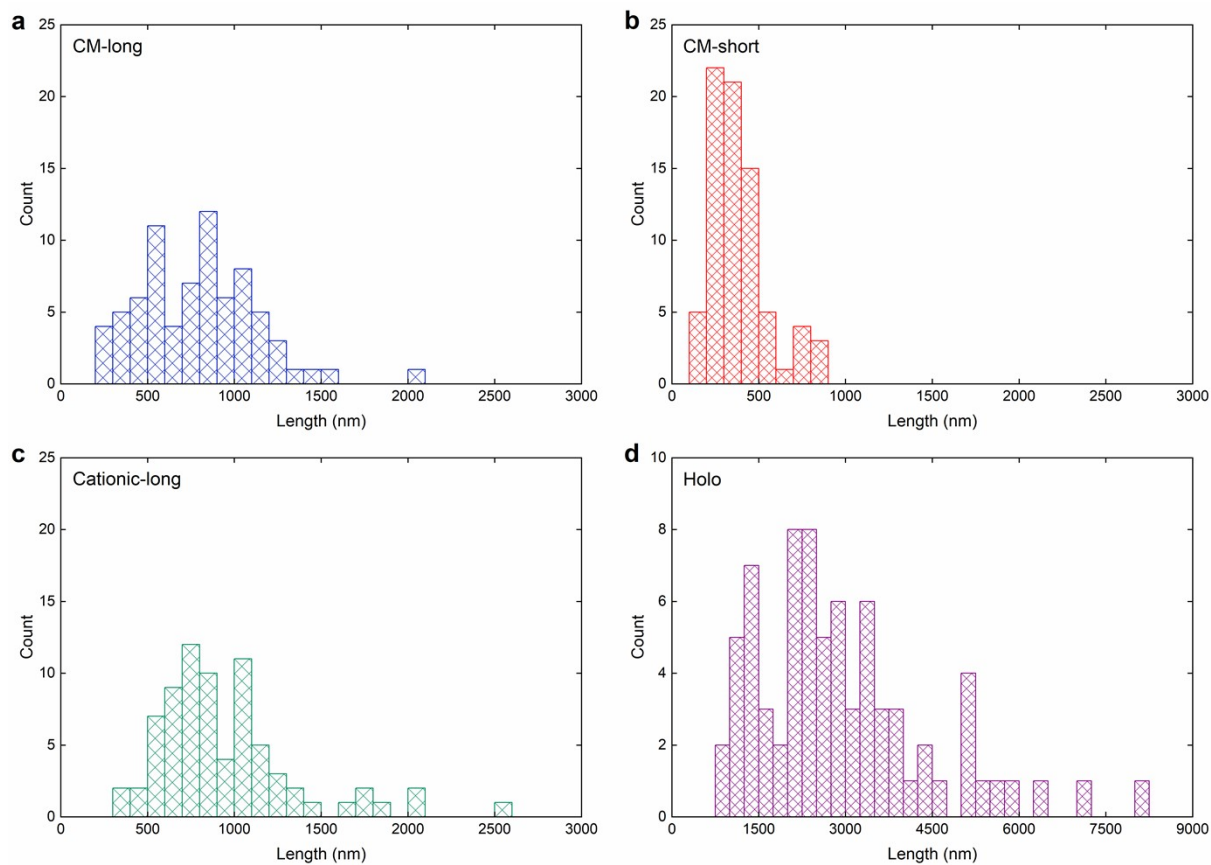


Fig. S1. Length distribution for a) CM-long, b) CM-short, c) Cat-long, and d) Holo CNFs.

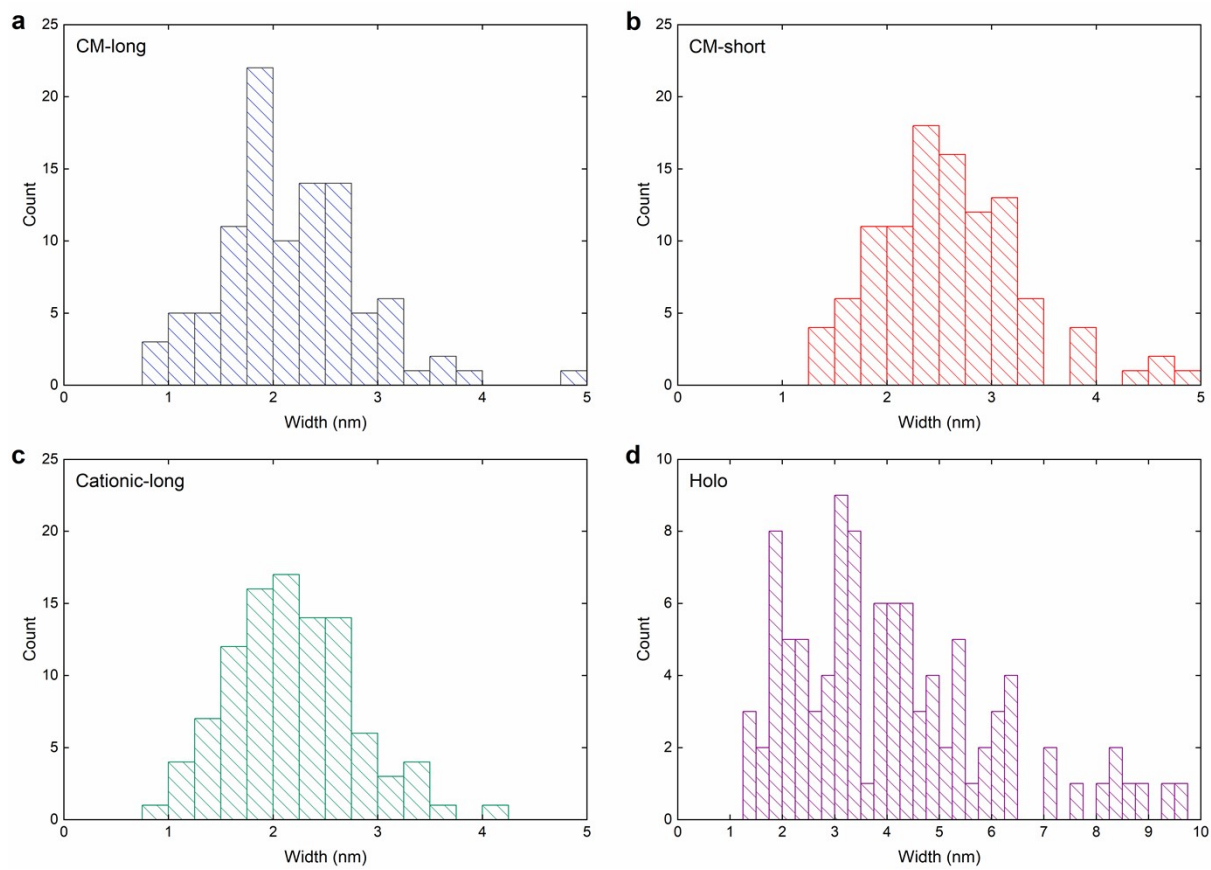


Fig. S2. Width distribution for a) CM-long, b) CM-short, c) Cat-long, and d) Holo CNFs.

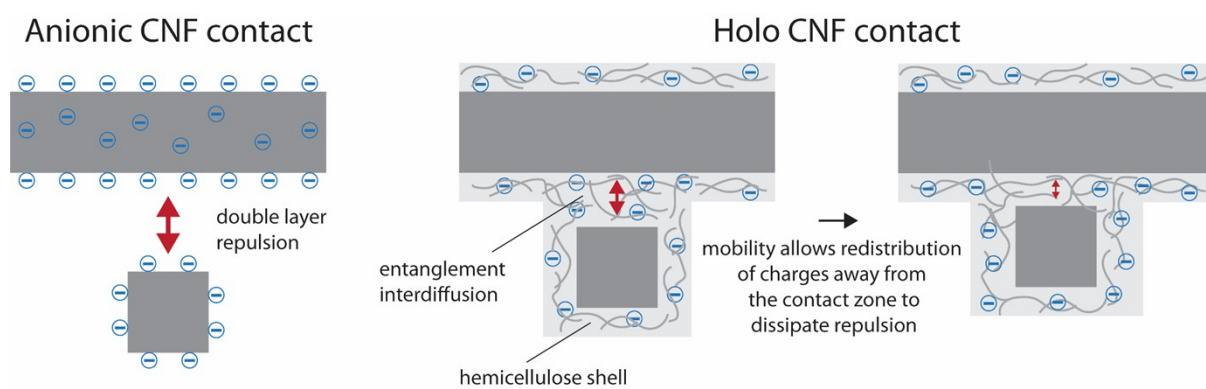


Fig. S3. Hypothetical mechanism for Holo-CNF interfaces by entanglement, interdiffusion, and redistribution of charges to dissipate repulsion.

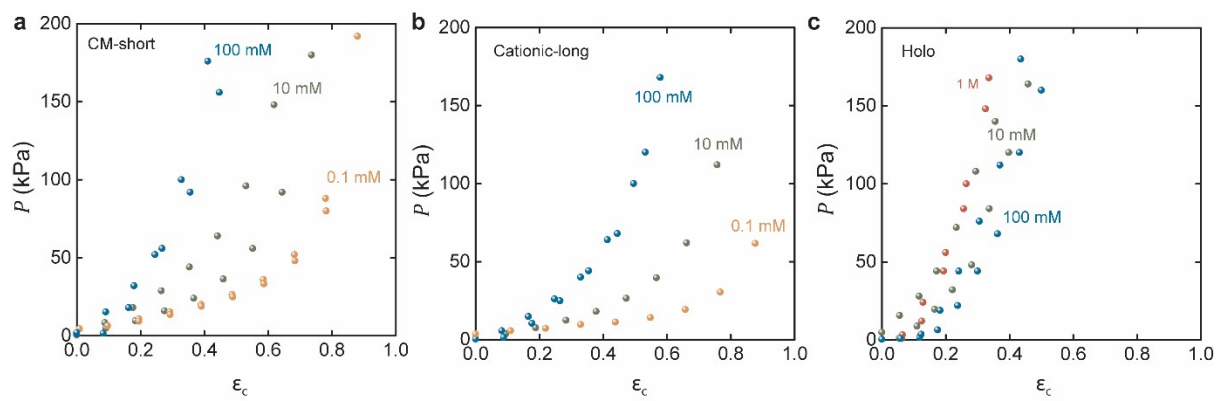


Fig. S4. Plate pressure (P) as a function of compressive strain for a) CM-short, b) Cat-long, and c) Holo CNFs.

Table S1. Steps and relaxation time during compression measurements.

| STEP | COMPRESSION RATE ($\mu\text{m/s}$) | RELAXATION TIME (min) |
|-------------|--|----------------------------------|
| 1 | 0.3 | 20 |
| 2 | 0.3 | 20 |
| 3 | 0.3 | 20 |
| 4 | 0.3 | 20 |
| 5 | 0.2 | 20 |
| 6 | 0.2 | 20 |
| 7 | 0.2 | 40 |
| 8 | 0.1 | 40 |
| 9 | 0.1 | 60 |
| 10 | 0.1 | 60 |