

Supporting Information for

Surface Structuring and Water Interactions of Nanocellulose Filaments Modified with Organosilanes toward Wearable Materials

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This Supporting Information document contains two figures and two tables in three pages.

Thermogravimetric analysis (TGA). TGA was carried out using a TA Instruments Q500 Thermogravimetric Analyzer. Filament samples of *ca.* 2 mg were heated from 40 to 800 °C at 10 °C.min⁻¹ in an inert environment provided by a 20 mL.min⁻¹ nitrogen purge.

Thermograms (Figure S1) showed that the unmodified filament exhibited the typical thermal degradation behavior of unmodified cellulose nanofibrils, presenting a single main step of thermal degradation, with maximum decomposition rate ($T_{d_{max}}$) at 308 °C. The modified filaments were shown to be slightly less stable (except the TCf5 sample), since they started decomposing at lower temperatures, albeit with a less abrupt decay, probably due to a corresponding lower heat transfer to cellulose, resulting from the presence of the inorganic coating around the filament. Moreover, their thermograms revealed a slightly more complex degradation behavior, with additional decomposition steps (compared to that of the unmodified filament). The first step, with $T_{d_{max1}}$ around 220-240 °C, is attributed to the decomposition of the organic functions (methyl groups) in the hybrid coatings. Additionally, the $T_{d_{max2}}$ for the second decomposition step decreased, compared to the equivalent $T_{d_{max}}$ in the precursor CNF

filament, varying in the range 292-300 °C (except for the TCf5 sample, to which it was 314 °C). Nonetheless, the higher amount of final residue at *ca.* 800 °C for the modified filaments corroborates the presence of an inorganic environment around them.

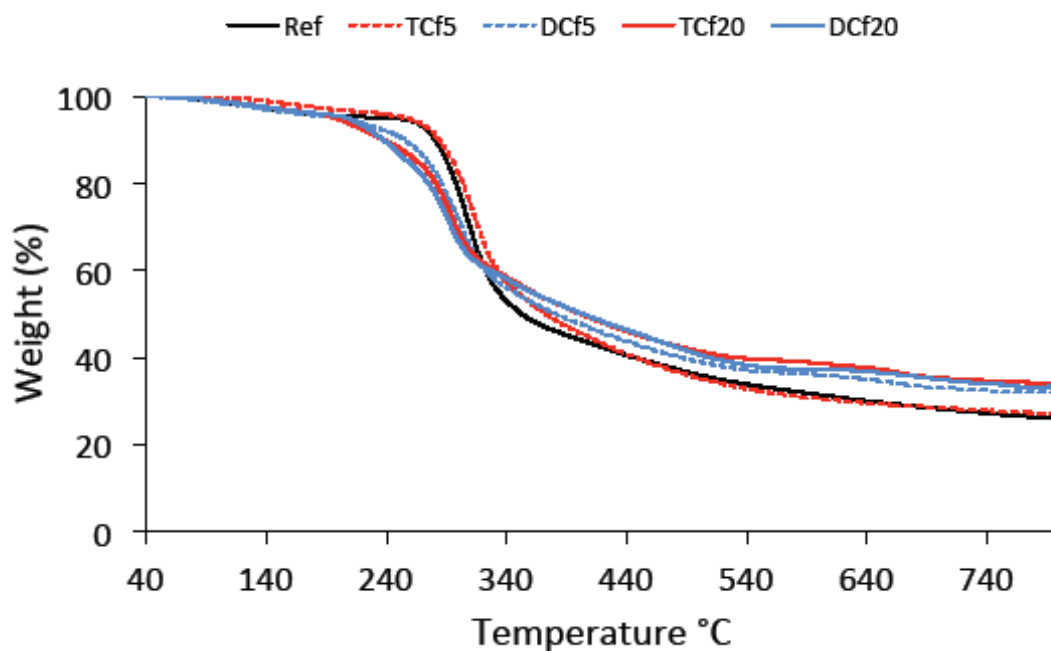


Figure S1. TGA thermograms of unmodified and organosilane-modified CNF filaments.

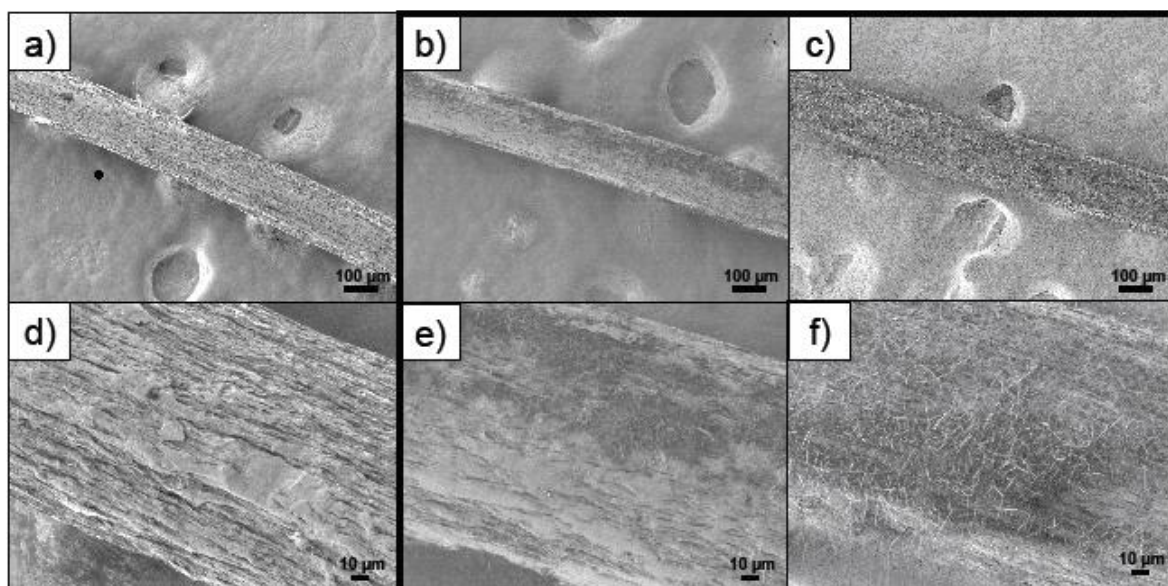


Figure S2. SEM micrographs at different magnifications of unmodified and organosilane-modified CNF filaments: Ref (a and d), TCf5 (b and e) and TCf20 (c and f) at 100x (a, b and c) and 500x (d, e and f) magnification.

Contact angle

Table S1. Contact angles of given fluids on unmodified and organosilane-modified CNF filaments with the calculated surface energy.

	Contact angle (°)			Surface energy (mJ.m ⁻²)		
	Water	Formamide	Diiodomethane	Polar contribution	Dispersive contribution	Total
Ref	57 ± 3	37 ± 6	61 ± 4	19.48	27.87	47.35
TCf5	85 ± 5	91 ± 2	79 ± 10	7.80	13.53	21.34
TCf10	96 ± 9	94 ± 2	87 ± 5	4.52	11.81	16.34
TCf15	100 ± 1	99 ± 5	90 ± 5	3.60	10.45	14.05
TCf20	116 ± 3	102 ± 2	100 ± 10	0.88	8.90	9.77
DCf5	94 ± 6	99 ± 3	75 ± 4	3.38	14.86	18.23
DCf10	94 ± 3	98 ± 5	78 ± 3	3.82	13.94	17.76
DCf15	98 ± 3	98 ± 2	77 ± 6	2.42	15.00	17.43
DCf20	96 ± 3	98 ± 4	80 ± 5	3.43	13.53	16.97

Tensile strength

Table S2. Tensile strength and Young's modulus of unmodified and organosilane-modified CNF filaments in dry and wet conditions.

	Dry conditions		Wet conditions	
	Tensile strength (MPa)	Young's Modulus (GPa)	Tensile strength (MPa)	Young's Modulus (MPa)
Ref	161.9 ± 17.9	9.1 ± 0.6	1.9 ± 0.2	184.2 ± 29.6
TCf5	179.4 ± 14.8	11.1 ± 0.2	3.0 ± 0.4	158.3 ± 14.4
TCf20	147.9 ± 14.2	8.3 ± 1.0	3.2 ± 0.4	185.7 ± 12.0
DCf5	162.4 ± 7.1	9.1 ± 0.7	3.0 ± 0.5	236.1 ± 60.3
DCf20	127 ± 17.1	7.5 ± 1.0	4.9 ± 0.7	280.6 ± 78.6