## **Supplementary Information (SI)**

## High throughput tailoring of nanocellulose films: from complex biobased materials to defined multifunctional architectures

Alexey Khakalo<sup>†</sup>\*, Tapio Mäkelä<sup>‡</sup>, Leena-Sisko Johansson<sup>§</sup>, Hannes Orelma<sup>†</sup>, Tekla Tammelin<sup>†</sup>\*

<sup>†</sup> VTT Technical Research Centre of Finland Ltd., Tietotie 4E, P.O. Box 1000, FI-02044 Espoo, Finland

‡ VTT Technical Research Centre of Finland Ltd., Tietotie 3, FI-02150 Espoo, Finland

§ Department of Bioproducts and Biosystems, School of Chemical Engineering, Aalto University, P.O. Box

16300, FI-00076 Aalto, Finland

\*Corresponding authors: alexey.khakalo@vtt.fi, Tel. +358504018149

tekla.tammelin@vtt.fi, Tel. +358207224632



**Figure S1.** Surface energies (a) and water contact angles (b) of NIL surface patterned and APTES and hybrid APTES+HMDSO plasma deposited TEMPO-CNF films.

**Table S1.** Surface free energy parameters for the liquid probes and calculated surface energy components and affinity to water  $\Delta G_{sws}^{IFE}$  for plasma deposited TEMPO-CNF films with and without NIL surface patterning.

	$\gamma^+$	γ	$\gamma^p$	$\gamma^d$	<i>7s</i>	$\gamma / \gamma^+$	$\Delta G_{sws}^{IFE}$
Liquid probes							
Water	25.5	25.5	51	21.8	72.8	-	-
Formamide	2.28	39.6	19	39	58	-	-
Diiodomethane	0	0	0	50.8	50.8	-	-
Ethylene glycol	1.92	47	18.99	29	47.99	-	-
Plasma-deposited TEMPO-CNF film							
Reference	0.6	53.3	11.5	38.9	50.4	85.9	33.4
HMDSO	1.0	0.3	1.0	27.7	28.7	0.3	-74.6
APTES	1.7	17.5	10.9	41.8	52.7	10.3	-19.4
APTES+HMDSO	2.2	0.8	2.7	19.6	22.3	0.4	-59.5
NIL patterned TEMPO-CNF films after plasma deposition							
Reference	1.3	8.5	6.7	33.6	40.3	6.5	-35.9
HMDSO	0.0	0.4	0.2	21.4	21.6	-	-86.6
APTES	0.0	18.1	1.2	36.1	37.3	-	-19.2
APTES+HMDSO	0.1	2.5	1.0	21.5	22.4	27.3	-66.1

 $\Delta G_{sws}^{IFE}$  is given in mJ/m<sup>2</sup>. The standard deviation in the data is less than 5%.