Supporting information

The Influence of Drying Routes on the Properties of Anisotropic All-Cellulose Composite Foams from Post-Consumer Cotton Textiles

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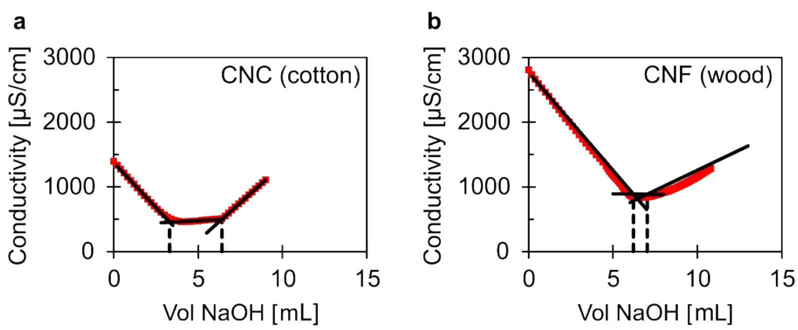

Figure S 1: Exemplary conductometric titration curves of (a) cotton textile-derived CNCs and (b) wood-derived CNFs. The flat region corresponds to the titration of carboxylic groups.

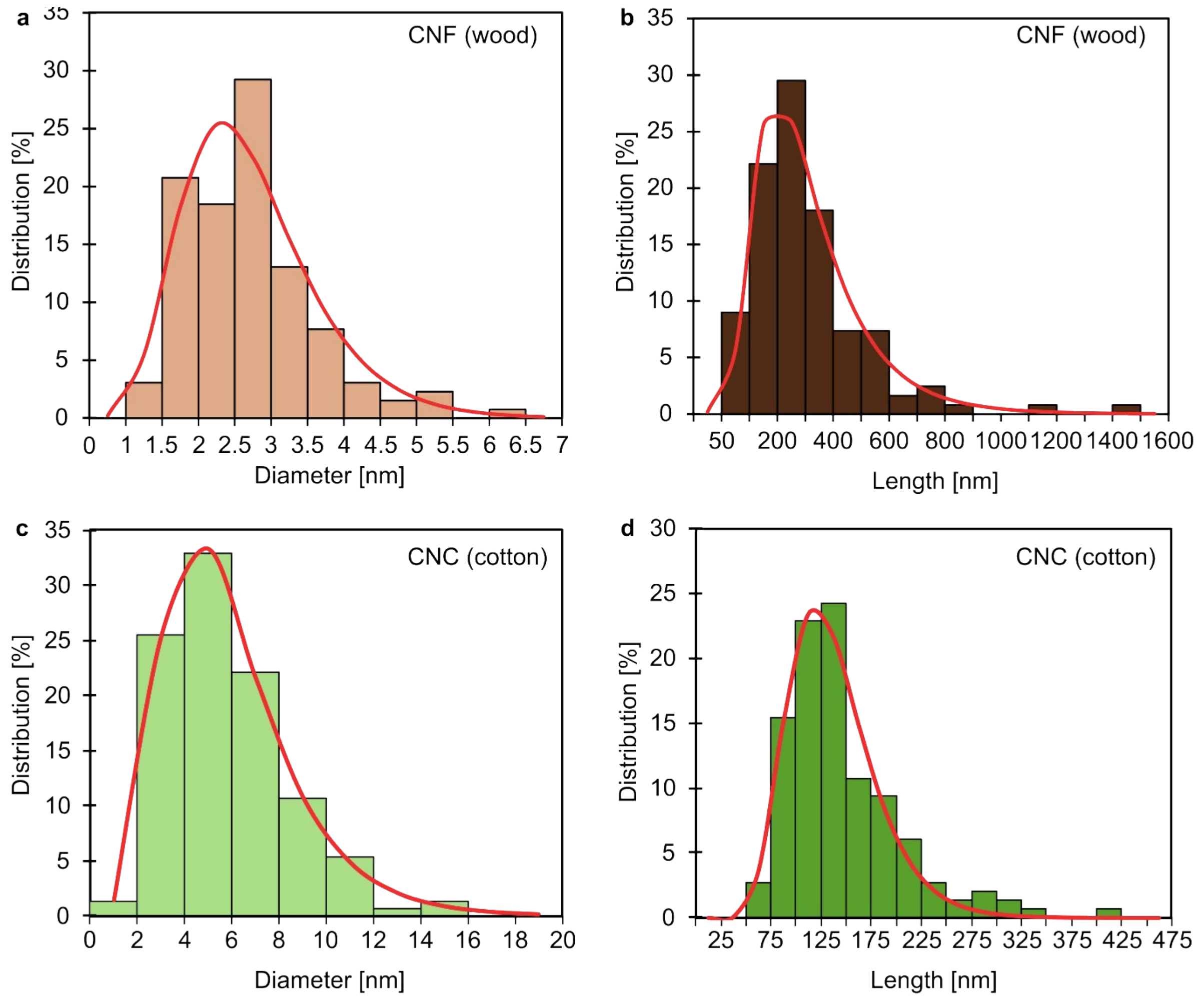

Figure S 2: Particle diameter and length distribution of cotton textile-CNCs and wood-CNFs with the fitting of the lognormal distribution.

Table S 1: Composition and properties of the foams

Label	CNC:CNF ratio	Density at 50% RH [kg m^{-3}]	Porosity [%]	Solid content [wt%]	Drying technique
CNC:CNF-1:1_FD	1:1	13 ± 0.4	99	1	FD
CNC:CNF-5:1_FD	5:1	34 ± 1.2	98	3	FD
CNC:CNF-9:1_FD	9:1	52 ± 0.8	96	5	FD
CNC:CNF-1:1_SCD	1:1	24 ±1.3	98	1	SCD
CNC:CNF-5:1_SCD	5:1	44 ± 1.6	98	3	SCD
CNC:CNF-9:1_SCD	9:1	66 ± 1.6	96	5	SCD

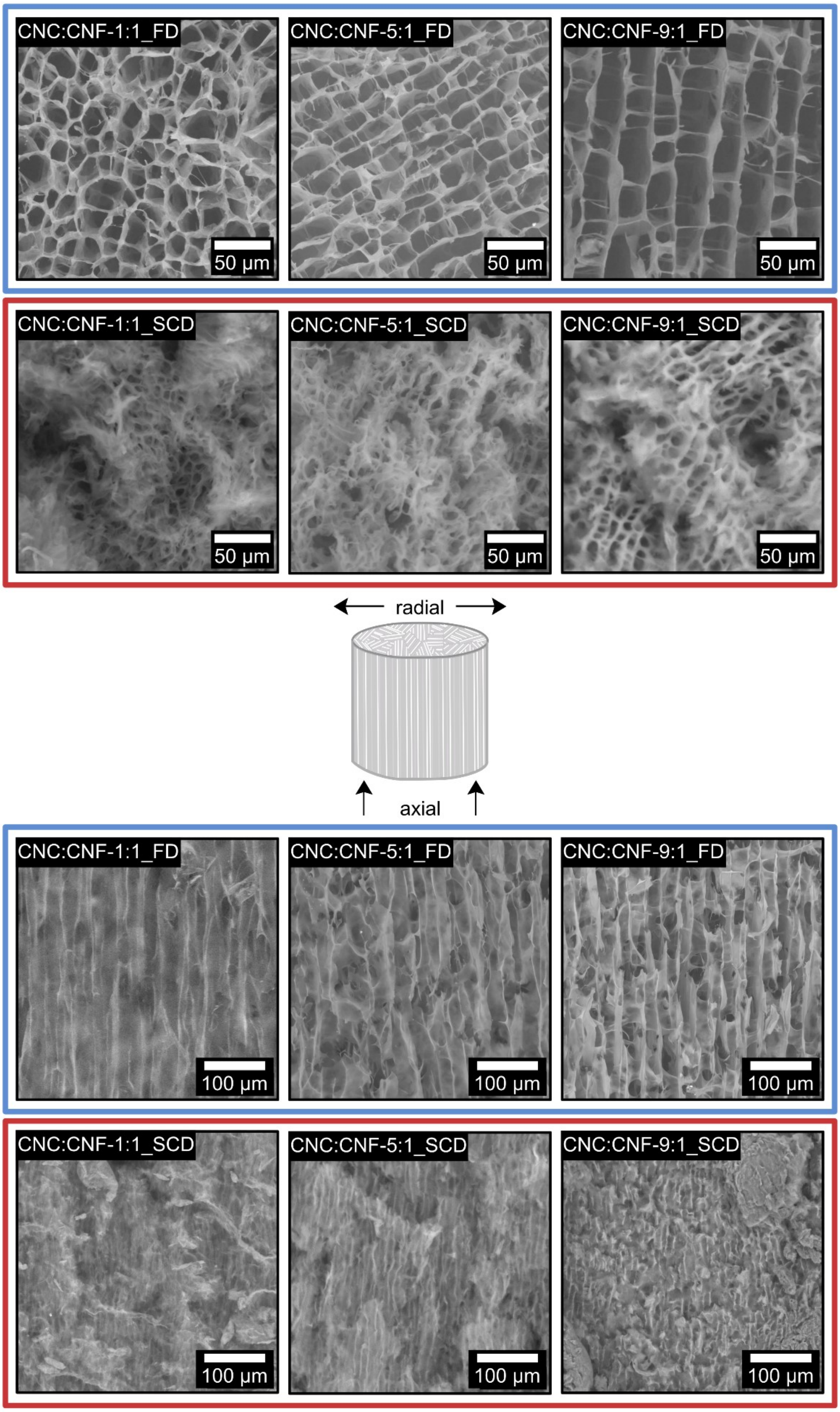

Figure S 3: Scanning electron microscopy images of the eight different configurations along the radial (top) and axial (bottom) direction.

Table S 2: Properties of the foam pores, surface area and CO_2 adsorption

Label	Macropore size [µm] via SEM n=120	Macropore size [µm] via Tomography n=120	Surface area [m^2 g^{-1}] via BET	Share of macropores (≥ 300 nm) [%]	Share of nanosized pores (≤ 300 nm) [%]	CO_2 adsorption [mmol g^{-1}]
CNC:CNF-1:1_FD	28 ± 8	19 ± 4	19	99.92	0.08	-
CNC:CNF-5:1_FD	26 ± 8	16 ± 4	8	99.92	0.08	-
CNC:CNF-9:1_FD	27 ± 8	13 ± 3	6	99.90	0.10	-
CNC:CNF-1:1_SCD	9 ± 2	-	234	98.76	1.24	-
CNC:CNF-5:1_SCD	9 ± 4	9 ± 2	263	97.60	2.40	0.385
CNC:CNF-9:1_SCD	10 ± 3	15 ± 3	262	96.87	3.13	0.383

Table S 3: Scale factor C derived from the Porod model at different relative humidity (RH)

Label	Scale Factor C (x 10^{-6}) At 50% RH	Scale Factor C (x 10^{-6}) At 80% RH
CNC:CNF-1:1_FD	2.53 ± 0.03	2.28 ± 0.03
CNC:CNF-5:1_FD	1.76 ± 0.03	1.60 ± 0.03
CNC:CNF-1:1_SCD	107 ± 0.18	109 ± 0.35
CNC:CNF-5:1_SCD	347 ± 0.40	428 ± 0.40

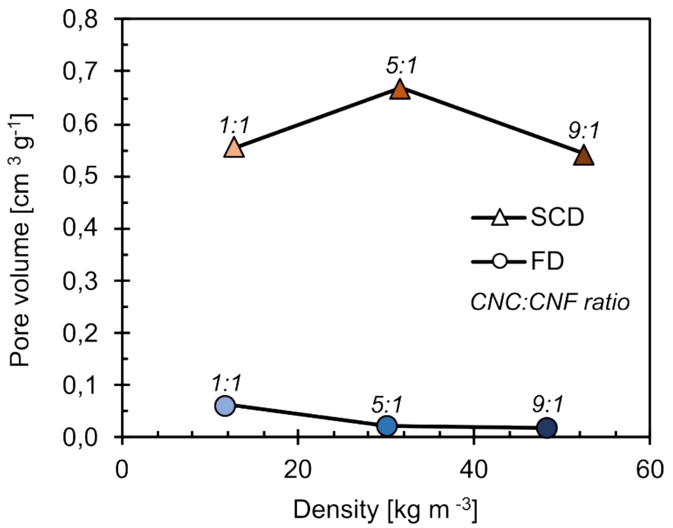

Figure S 4: Pore volume of pores between 1.7 and 300 nm as estimated from N_2 sorption using the Barrett–Joyner–Halenda (BJH) model.

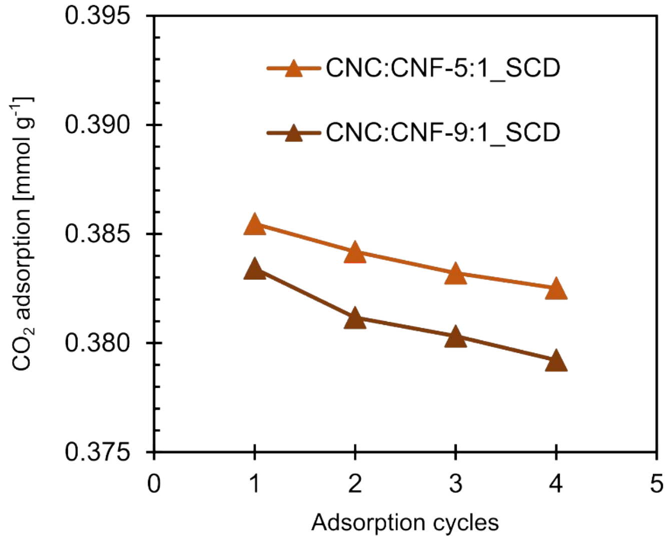

Figure S 5: CO_2 adsorption capacity at STP and 0°C of CNC:CNF-5:1_SCD and CNC:CNF-9:1_SCD as a function of CO_2 adsorption cycles.

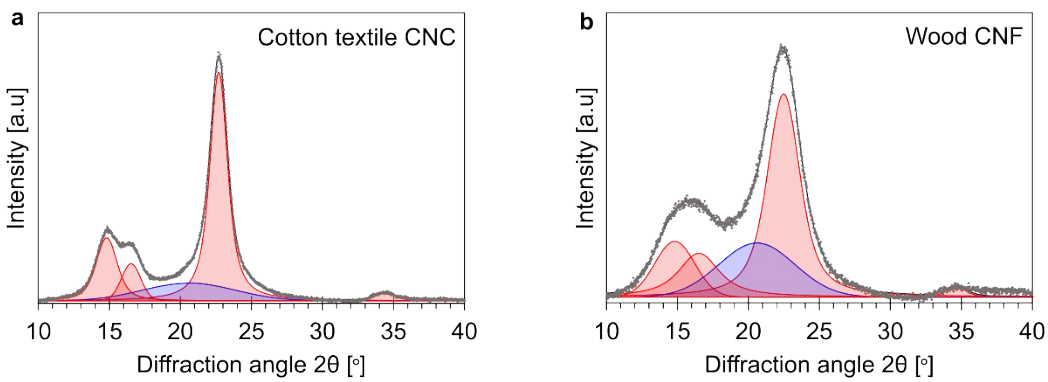

Figure S 6: XRD profiles of (a) cotton textile CNCs and (b) wood CNFs. The dotted curve corresponds to the experimental data. The black line is the fitted model. the red curves are the crystalline peaks. and the blue curve is the amorphous peak. The crystallinity indexes derived from these figures are stated in Table S2.

Table S 4: Crystallinity index (CI) of cotton textile CNCs and wood CNFs

Label	CI deconvolution [%]	CI Segal [%]
Cotton textile CNC	83	92
Wood CNF	77	78

Table S 5: Orientation degree for the foam walls and the particles

Label	Macropore alignment SEM + OrientationJ n=3	Particle alignment 50%RH SAXS	Particle alignment 80%RH SAXS	Particle alignment WAXS n=3
CNC:CNF-1:1_FD	0.93 ± 0.01	0.92	0.92	0.70 ± 0.01
CNC:CNF-5:1_FD	0.91 ± 0.01	0.89	0.91	0.62 ± 0.02
CNC:CNF-9:1_FD	0.92 ± 0.01	-	-	0.71 ± 0.02
CNC:CNF-1:1_SCD	0.85 ± 0.02	-	-	-
CNC:CNF-5:1_SCD	0.88 ± 0.02	-	-	-
CNC:CNF-9:1_SCD	0.78 ± 0.04	-	-	-

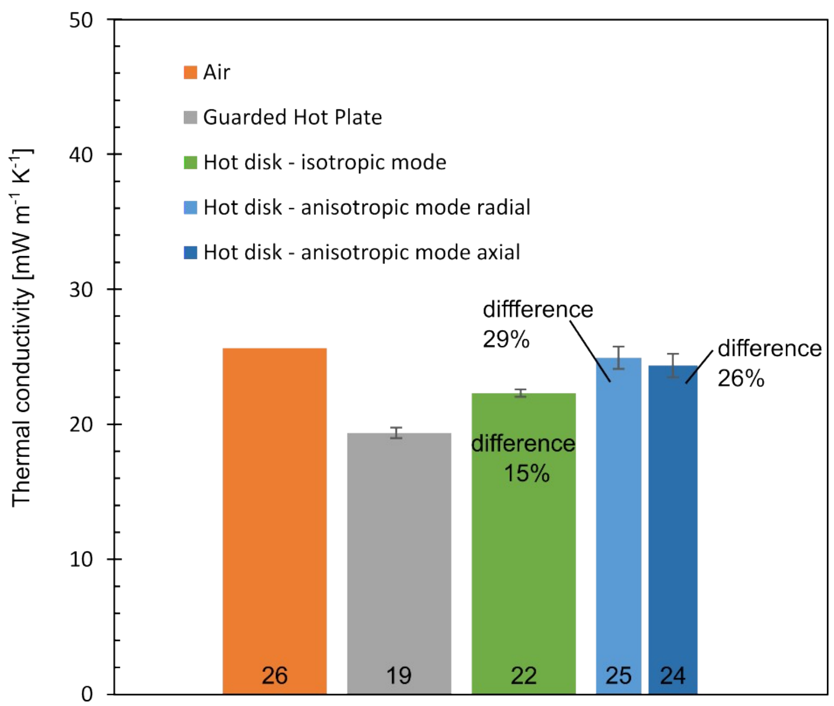

Figure S 7: Thermal conductivity of a super-insulating reference material (PU aerogel) measured with the Hot disk in isotropic and anisotropic mode compared to a reference measurement on the Guarded Hot Plate apparatus.

Table S 6: Mechanical properties of the foams

Label	Young's Modulus [kPa] n=5	Specific Young's Modulus [kNm kg^{-1}] n=5	Toughness [kJ m^{-3}] n=5	Specific Toughness [kJ kg^{-1}] n=5
CNC:CNF-1:1_FD	440 ± 140	35 ± 10	210 ± 70	17 ± 5
CNC:CNF-5:1_FD	2600 ± 550	76 ± 17	1600 ± 300	48 ± 8
CNC:CNF-9:1_FD	4200 ± 400	81 ± 8	2100 ± 780	41 ± 15
CNC:CNF-1:1_SCD	330 ± 57	14 ± 2	340 ± 160	14 ± 6
CNC:CNF-5:1_SCD	560 ± 120	13 ± 2	280 ± 74	6 ± 2
CNC:CNF-9:1_SCD	950 ± 180	14 ± 3	330 ± 120	5 ±2

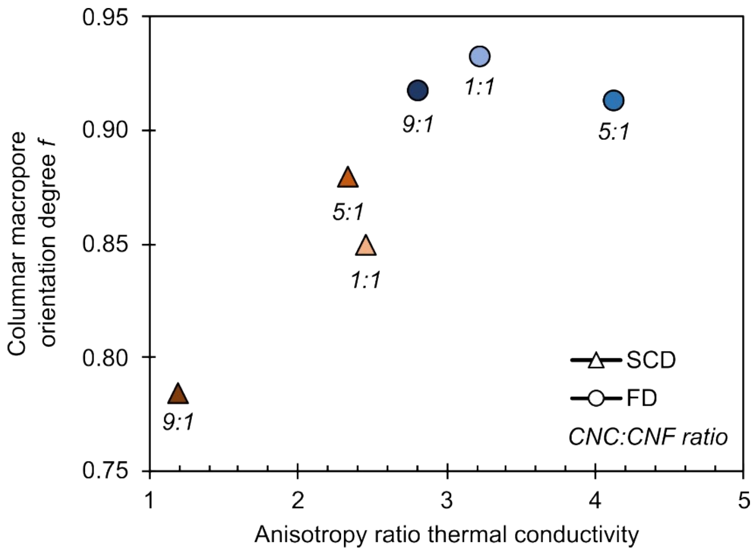

Figure S 8: Orientation degree *f* of the columnar macropores as a function of the anisotropy ratio of the thermal conductivity.