Supporting Information

Surface-induced Frustration in Solid State Polymorphic Transition of Native Cellulose Nanocrystals

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This Supporting Information contains:

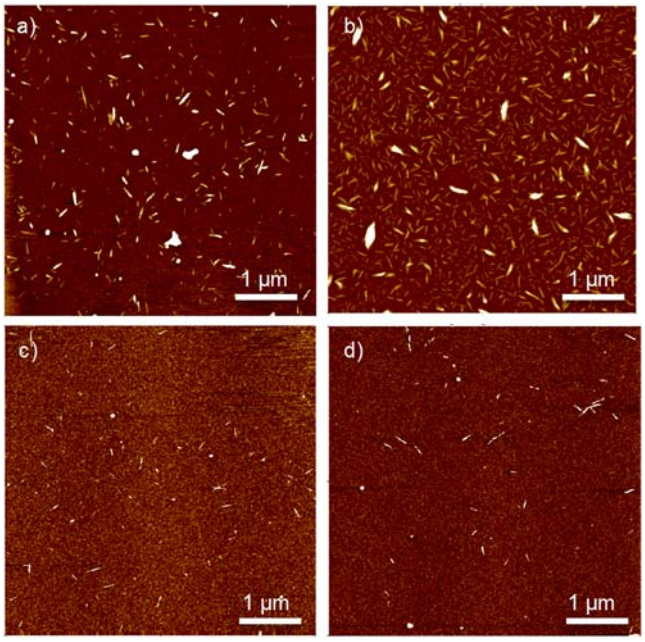

Figure S1. *AFM-image samples (5 x 5 µm^2) of a) cellulose I nanocrystals, b) cellulose III nanocrystals, c) surface immobilized EDA-treated CNC rinsed with methanol, and d) surface immobilized EDA-treated CNC rinsed with water.*

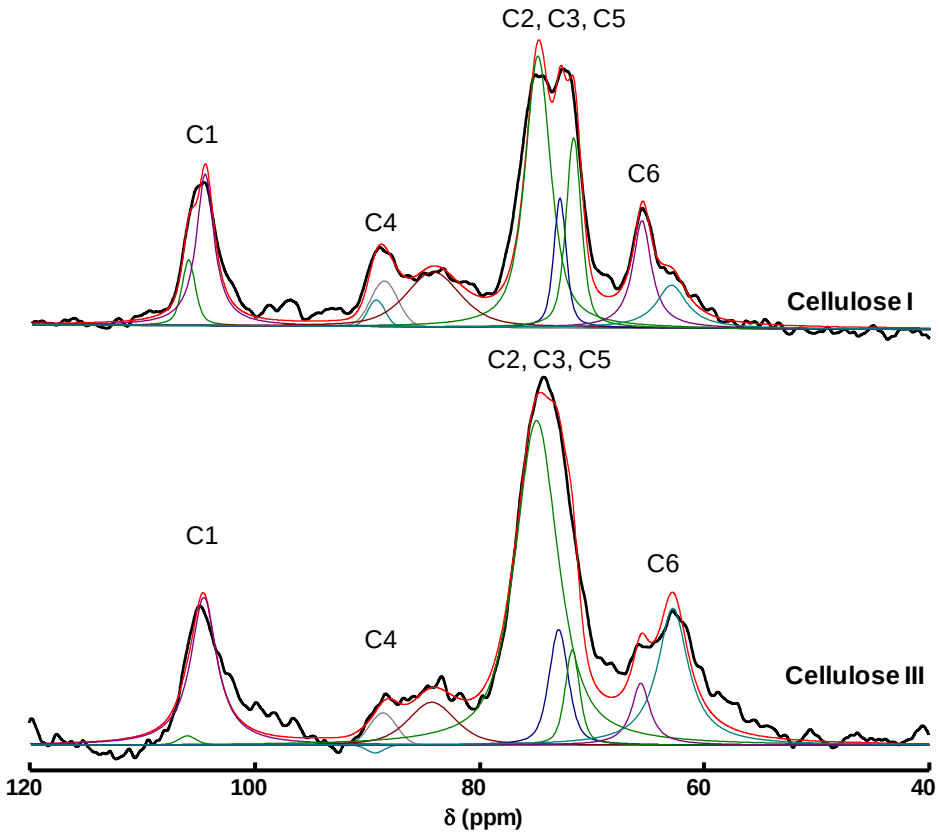

Figure S2. *SP-MAS 13C-NMR spectra with peak fitting. Red curve presents the sum curve of the peak fits.*

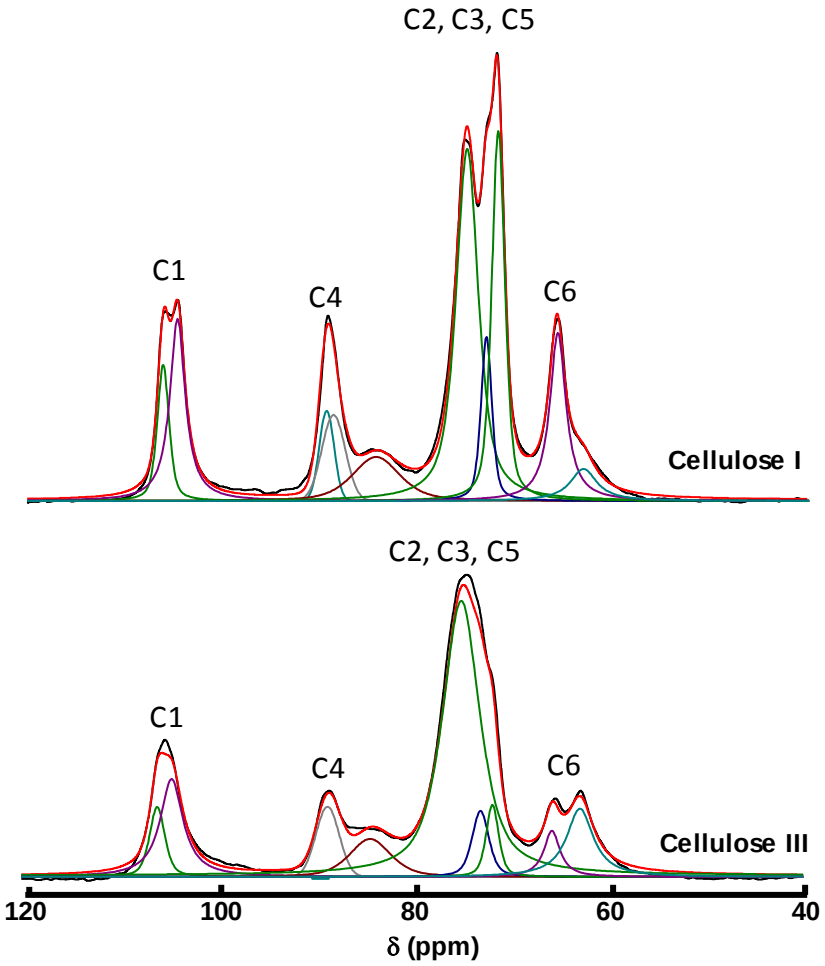

Figure S3. *CP(cross polarization)-MAS 13C-NMR spectra with peak fitting. Red curve presents the sum curve of the peak fits.*

Table S1. *The chemical shifts and intensity integrals of the NMR-spectra presented in Figure S2 and S3.*

attribution	**δ, ppm**	**cellulose I**		**cellulose III**	
		SP	CP	SP	CP
C1	106.01	2397	56296	357	39030
	104.54	8703	119173	9527	98402
C4	89.30	991	37536	-217	-11
	88.60	2663	57388	1456	48398
disordered	84.25	7938	70234	4104	55016
C2, C3, C5	74.93	20171	297821	30312	401345
	72.96	4365	62734	4561	40614
	71.75	7762	172029	2812	32935
C6	65.66	6432	114708	2822	32091
	63.04	4188	35012	10313	79168

Amount of Cellulose III after phase change: $100*C6_{65.66,\ III} / (C6_{63.04,\ I} + C6_{65.66,\ III})$

CP: 100*79168/(79168+32091)= 71%

SP: 100*10313/(10313+2822)= 79%

Average: 75%

Table S2. *The average and median widths and lengths determined with AFM and the particle counts of the samplings.*

	Aver. width, nm	**Aver. length, nm**	**Median width, nm**	**Median length, nm**	**Particle count**
Cellulose I CNC	4.5 ±1.4	114.0 ±61.8	4.5	97.8	2694
Cellulose III CNC	5.2±2.2	138.0 ±77.8	4.8	113.9	2337
Surface immobilized CNC	2.1 ±0.6	117.0 ±55.2	2.1	106.4	726
Surface immobilized, water rinsing	1.8 ±0.3	109.9 ±40.8	1.8	102.3	23

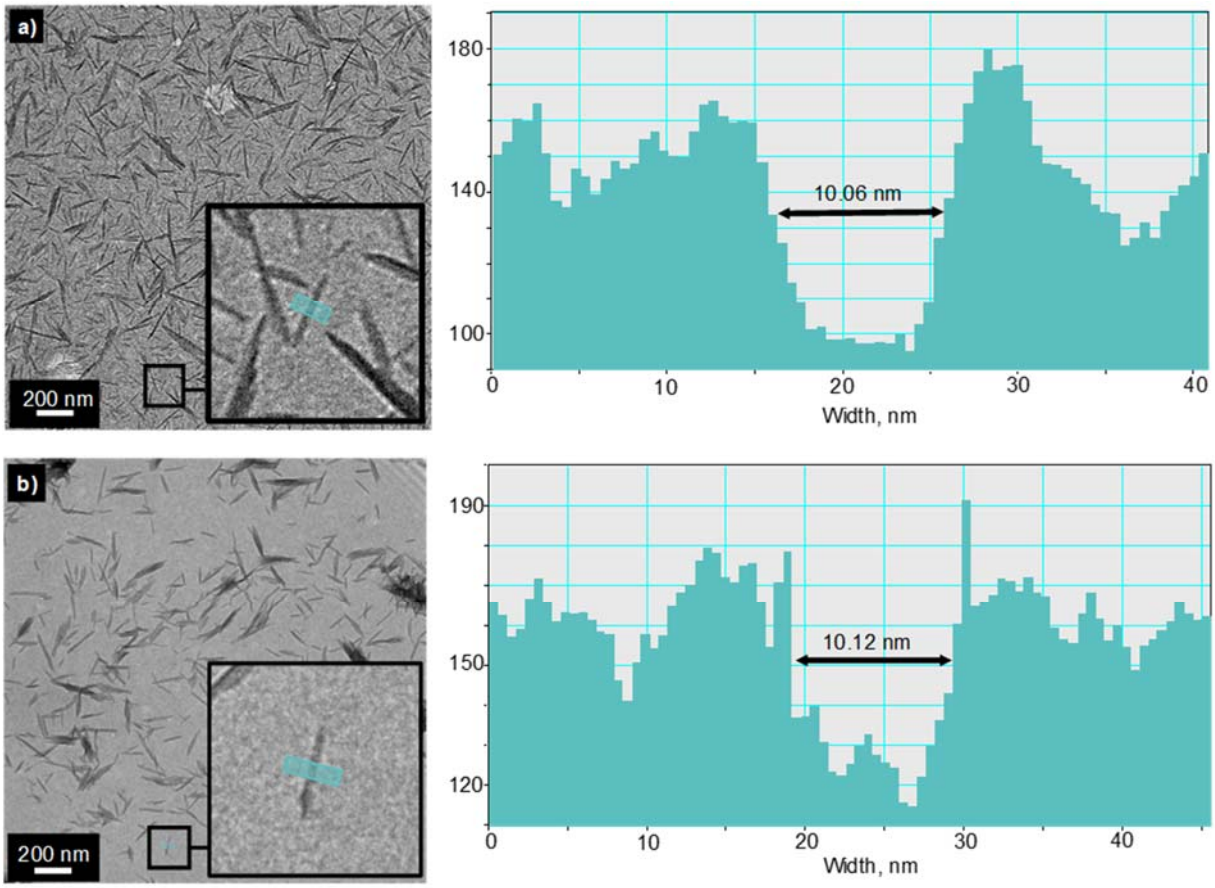

Figure S4. *TEM images of a) cellulose I nanocrystals and b) cellulose III nanocrystals with example widths of the crystals.*

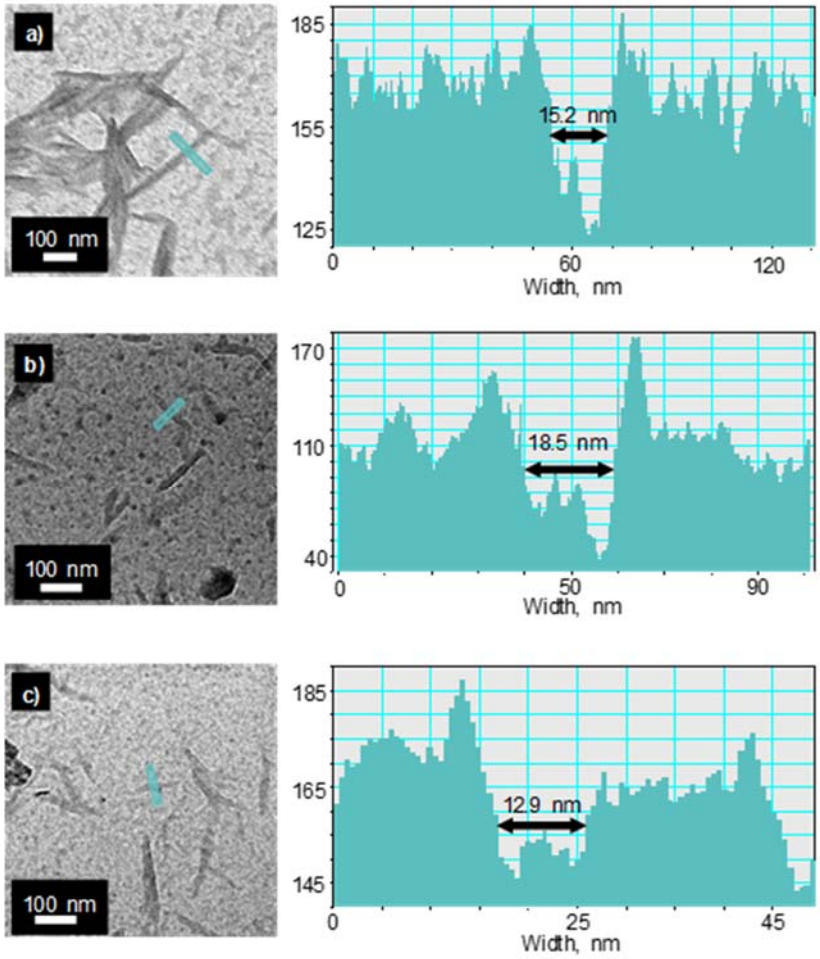

Figure S5. *TEM images and grayscale image analysis of sample particles of a) CNC from cellulose I, b) CNCs of cellulose III prepared in dispersion, c) CNCs of cellulose III from immobilized cellulose I CNCs.*

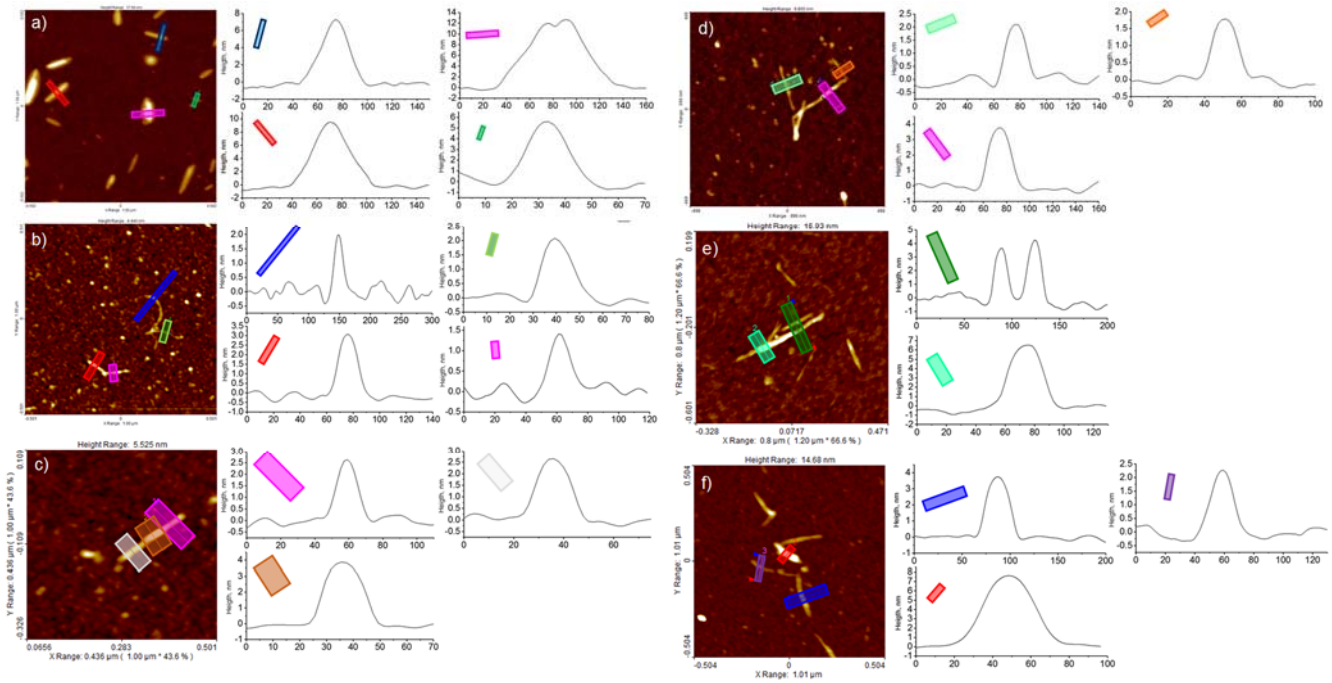

Figure S6. *AFM images and height profiles for a) native CNC, and for b-f) surface immobilized EDA-treated CNCs presented in Figure 8: b) Figure 8a and 8b; c) Figure 8c; d) Figure 8d; e) Figure 8e; f) Figure 8f*