

Supporting information



Figure S1. Image of a suspension of cellulose nanocrystals in DMF showing flow birefringence under crossed polars.

Table S1. Effect of polymer, EDAC and NHS to carboxyl groups molar ratios (N_P , N_{EDAC} and N_{NHS} , respectively) on the degree of substitution, DS, and peptidic coupling yield, $100 \times DS/DO$.

Sample	Reaction solvent	Grafted polymer	N_P	N_{EDAC}	N_{NHS}	DS ^a	$100 \times DS/DO^b$ (%)
S1000W	water	M1000	2	1.5	1.5	0.038	19.5
S1000W	water	M1000	4	4	4	0.059	30.1
S1000D	DMF	M1000	2	2	2	0.047	29
S1000D	DMF	M1000	4	4	4	0.1049	53.5
S2070D	DMF	M2070	2	2	2	0.041	26
S2070D	DMF	M2070	4	4	4	0.0973	49.6

Fourier-transform infrared spectroscopy in ATR mode.

To complement the results of infrared spectroscopy in transmission mode, some samples were also investigated in reflectance mode. For these measurements, CNC suspensions were first acidified to pH 2-3 to avoid the superposition of the carbonyl band with that of water and then freeze-dried. Dried samples were then immersed in D₂O and the resulting paste was deposited on the diamond sample support of an ATR equipment. Spectra were recorded in reflectance mode using the same apparatus as described in the experimental section. Under these conditions, the water band around 1600 cm⁻¹ was shifted to 2400 cm⁻¹ allowing the unambiguous observation of bands in the 1600 cm⁻¹ region.

Infrared spectra of oxidized and M2070-grafted samples recorded in ATR mode are presented in Figure S2. In ATR mode, the oxidized sample spectrum only shows the carboxyl group band at 1725 cm⁻¹ since the residual water absorption around 1650 cm⁻¹ has been shifted toward lower wave numbers (Figure S2a). After grafting, as seen in transmission mode, the carboxylic acid band is preserved but its intensity is lower since some carboxylic groups have reacted with the polymer (Figure S2b). In ATR mode, the grafting reaction between carboxylated CNCs and Jeffamine results in the appearance of two absorption bands at 1650 and 1550 cm⁻¹ that can unambiguously be attributed to amide I and amide II bands. FTIR measurements in ATR mode facilitate the observation

of the amide groups and thus confirm the covalent grafting of Jeffamine chains onto the CNC surface.

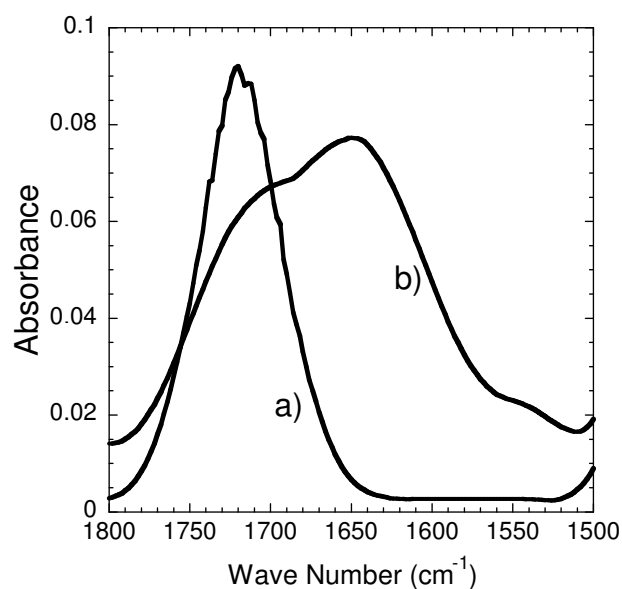


Figure S2. Infrared spectra in ATR mode of oxidized (a) and M2070-grafted (b) cellulose nanocrystals.

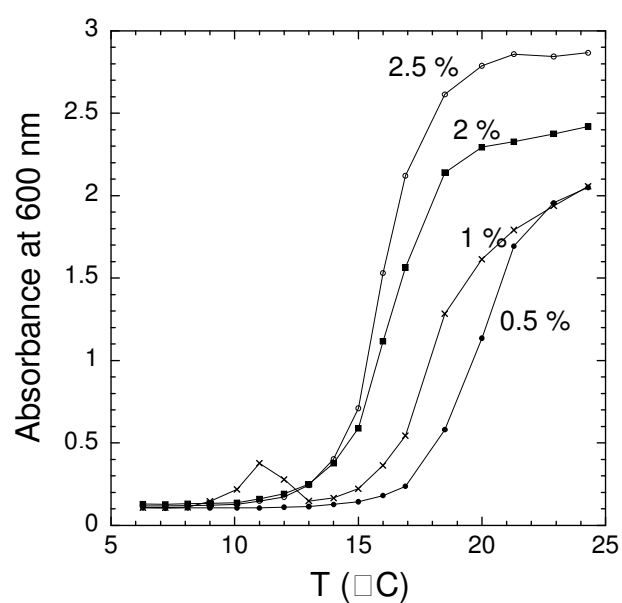


Figure S3. Absorbance at 600 nm as a function of temperature of a Jeffamine M2005 solution at different concentrations in wt%.