## Supplementary Materials

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## 1. Determination of Carbamation Degree (DSTDI) using EDX

The elemental composition of the original CNCs (DSTDI=0%) was determined using EDX (C: 45.9%, O: 52.1%, N: 0.0%, S: 1.2%, Na: 0.8%) and based on it, the elemental composition of proposed carbamated samples of DSTDI up to 33% was calculated to construct **Figure S1**. As a result, the DSTDI for any carbamated CNC sample can be calculated using the equation in the figure after its N/C molar ratio is determined using EDX (**Figure S2**).

30 25 20 DS<sub>TDI</sub> (%) 10 5 0 0.080 0.000 0.020 0.040 0.060 0.100 0.120 0.140 0.160 N/C Molar Ratio

**Figure S1.** The estimation of DS<sub>TDI</sub> of carbamated CNCs using the N/C molar ratio obtained from EDX.

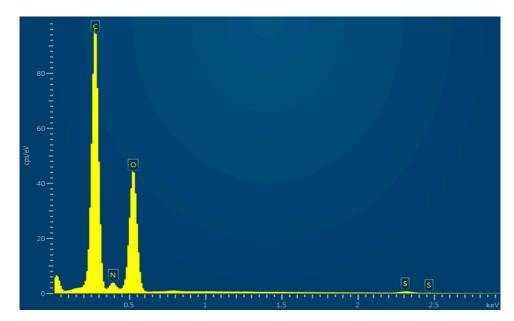


Figure S2. An example of an EDX measurement of a carbamated CNC sample.

**Table S1.** The estimation of DS<sub>TDI</sub> using EDX results based on N/C, N/O, and C/O Ratios.

T -(°C)	Using N/C		Using N/O		Using C/O	
	N/C Molar	$\mathbf{DS}_{\mathrm{TDI}}$	N/O Molar	$\mathbf{DS}_{TDI}$	C/O Molar	$\mathbf{DS}_{TDI}$
	Ratio	(%)	Ratio	(%)	Ratio	(%)
35	0.045±0.001	4.5±0.1	0.075±0.002	5.0±0.1	1.648±0.004	4.9±0.1
55	0.077±0.001	9.6±0.2	0.138±0.002	9.9±0.2	1.792±0.007	10.1±0.1
75	0.096±0.001	14.2±0.3	0.182±0.001	13.7±0.1	1.909±0.006	14.7±0.3

## 2. Determination of Carbamation Degree (DSTDI) using EDX, Elemental Analysis, and Mass Yield

The carbamation degree (DS $_{TDI}$ ) was determined using EDX and elemental analysis by monitoring the change in the N/C molar ratio. It was determined using mass yield (MY) following this equation:

$$DS_{TDI} = \frac{mmol\ TDI}{mmol\ CNC\ Hydroxyls} = \frac{(MY - 100)/17.4}{3*(mass\ CNCs(mg)/162)}*100\%$$

The estimated  $DS_{TDI}$  values from the three methods were comparable confirming the accuracy of all methods. However, the values obtained using mass yield were slightly lower due to mass losses during the carbamation process.

**Table S2.** The estimation of DS<sub>TDI</sub> using EDX, Elemental Analysis, and Mass Yield.

T	Using EDX		Using Elemental Analysis*		Using Mass Yield	
(°C)	N/C Molar	DSTDI	N/C Molar	DS <sub>TDI</sub> (%)	Mass Yield (%)	DS <sub>TDI</sub> (%)
	Ratio	(%)	Ratio			
35	0.045±0.001	4.5±0.1	0.059±0.003	4.8±0.3	112.4±0.6	3.8±0.2
55	0.077±0.001	9.6±0.2	0.093±0.003	9.4±0.5	128.4±2.1	8.8±0.7
75	0.096±0.001	14.2±0.3	0.115±0.000	13.7±0.1	143.5±1.8	13.5±0.5

\*Elemental Analysis was performed using Vario MICRO V3.1.1 CHNS-Modus (Elementar Analysensysteme GmbH, Germany)