Supplemental Information

Green bionanocomposites from high-elasticity "soft" polyurethane and high-crystallinity "rigid" chitin nanocrystals with controlled surface acetylation

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Amount of Surface Hydroxyl Groups (n_{surface-OH}) on Chitin Nanocrystals



$$n_{surface-OH} = \frac{N_1 N_2}{N_A} \tag{S-1}$$

$$N_1 = \left(\frac{w}{\rho_{chitin}}\right) / V_{ChN} = \left(\frac{w}{\rho_{chitin}}\right) / \left(\frac{\pi}{4}D^2L\right)$$
(S-2)

$$N_2 = 4 \left(\frac{S_{ChN}}{S}\right) = 4 \frac{\pi DL}{ab} \tag{S-3}$$

in here, $N_A = 6.02 \times 10^{23}$ mol⁻¹ (Avogadro's number), $\rho_{chitin} = 1.425$ g/cm⁻³, w = 1 g

The data of size of the unit cell of chitin (a, b) and density of chitin crystallites (ρ_{chitin}) were from following references:

- 1 G.L. Clark and A.F. Smith, J. Phys. Chem. 1936, 40, 863.
- J. Li, J.-F. Revol, E. Naranjo and R.H. Marchessault, *Int. J. Biol. Macromol.* 1996, 18, 177.



Figure S1. Size statistics for length (*L*, image A) and diameter (*D*, image B) of AChN from TEM images; dashed lines are Gaussian distribution fitting line according to the statistical data.



Figure S2. DSC thermograms of PU-based nanocomposites filled with various contents of AChN.



Figure S3. Amplified FTIR spectra of PU/AChN nanocomposites and neat PU material at the regions of 3600–3100 cm⁻¹ and 1800–1650 cm⁻¹. (a) PU, (b) PU/AChN-2, (c) PU/AChN-4, (d) PU/AChN-6, (e) PU/AChN-8, (f) PU/AChN-10.