## Supporting information

**Table S1.** Parameters obtained from parallelepiped and large disk model 'fits'. The tabulated data correspond to the following plotted curves: (1) red line in Figure 1, (2) blue line in Figure 1, (3) green line in Figure 1, (1') red dashed line in Figure 2 and (2') green dashed line in Figure 2.

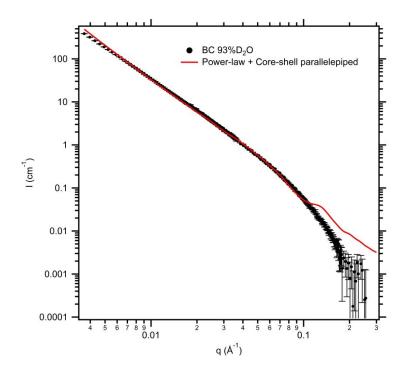
	Parallelepiped			Large disk	
	(1)	(2)	(3)	(1')	(2')
Scale factor	0.015	0.009	0.012	0.005	0.005
Ribbon width (nm)	1.0 (*)	7.0 (*)	4.9	8.0 (*)	8.0 (*)
Ribbon thickness (nm)	16.0 <sup>(*)</sup>	70.0 <sup>(*)</sup>	55.3	231.6 <sup>(*)</sup>	60.0
Ribbon length (nm)	500 <sup>(*)</sup>	500 <sup>(*)</sup>	500 (*)		
Polydispersity				0.95 (*)	0.34
SLD cellulose (10 <sup>10</sup> cm <sup>-2</sup> )	2.18	2.95	2.21	2.08	2.11
SLD solvent (10 <sup>10</sup> cm <sup>-2</sup> )	5.89 <sup>(*)</sup>	5.89 <sup>(*)</sup>	5.89 (*)	5.89 <sup>(*)</sup>	5.89 <sup>(*)</sup>

Parameters fixed during the fitting process are displayed with (\*).

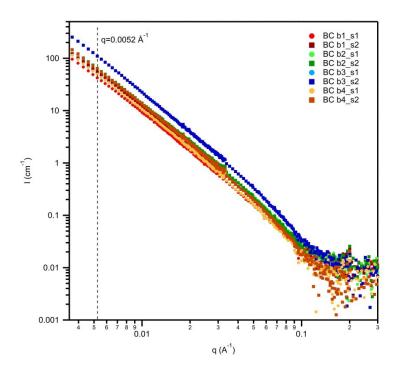
**Table S2.** Parameters obtained from the SAXS fits of the power-law plus core-shell cylinder with polydisperse radius model, adapted to account for the microfibril structural level, for the BC, BC-AX and BC-XG hydrogels. Standard deviations on the last digit are shown in parentheses.

	BC	BC-AX	BC-XG
Scale factor	0.017 (7)	0.06 (2)	0.05 (3)
Core radius (nm)	1.19 (6)	1.49 (4)	1.18 (2)
Polydispersity	0.60 (2)	0.53 (1)	0.600 (5)
Core length (nm) <sup>(*)</sup>	500.0	500.0	500.0
Radial shell thickness (nm)	1.31 (3)	0.78 (2)	0.63 (5)
Cellulose volume fraction (Core) (*)	1.0	1.0	1.0
Cellulose volume fraction (Shell)	0.65 (1)	0.58 (1)	0.28 (3)
SLD solvent (10 <sup>10</sup> cm <sup>-2</sup> ) <sup>(*)</sup>	11.84	11.84	11.84
SLD core cellulose (10 <sup>10</sup> cm <sup>-2</sup> ) (*)	14.46	14.46	14.46
SLD shell cellulose (10 <sup>10</sup> cm <sup>-2</sup> ) <sup>(*)</sup>	13.38	13.38	13.38
Power-law coefficient	2.4·10 <sup>-5</sup> (1)	7.5·10 <sup>-5</sup> (3)	6.6·10 <sup>-5</sup> (8)
Power-law exponent	3.12 (1)	3.19 (1)	3.02 (3)

Parameters fixed during the fitting process are displayed with (\*).



**Figure S1.** SANS experimental data for hydrated BC hydrogel (soaked in  $D_2O$ ) (dots) and corresponding model curve obtained using a power-law plus core-shell parallelepiped fitting function (red line). The following parameters were considered for the fitting procedure: The scale factor was allowed to vary between 0.001 and 0.03, the cylinder length was fixed to 500 nm, the core width and thickness, as well as the shell thickness were constrained between 6 and 60 nm, the SLD of the parallelepiped core and shell were allowed to vary between  $-0.7 \cdot 10^{10}$  cm<sup>-2</sup> (SLD<sub>bound H2O</sub>) and 7.97  $\cdot 10^{10}$  cm<sup>-2</sup> (SLD<sub>bound D2O</sub>), and the SLD of the solvent was fixed to 5.89  $\cdot 10^{10}$  cm<sup>-2</sup>



**Figure S2.** SANS patterns (background subtracted) corresponding to different pellicle samples (s1 and s2) from four BC batches (b1, b2, b3 and b4).