Nanocrystalline cellulose-based mixed ionic-electronic conductor for bioelectronics

This SI replaces the original version published on 27 September 2024

Katharina Matura,^a Rosarita D'Orsi,^b Laura Spagnuolo,^b Felix Mayr,^a Munise Cobet,^a Christoph Putz,^c Alessandra Operamolla,^b Serpil Tekoglu^a*

^aLinz Institute for Solar Cells (LIOS) and Institute of Physical Chemistry, Johannes Kepler University Linz, Altenberger Str. 69, A-4040, Linz, Austria

^bDepartment of Chemistry and Industrial Chemistry, University of Pisa, Via Giuseppe Moruzzi 13, I-56124, Pisa, Italy

^cDivision of Soft Matter Physics and Institute of Experimental Physics, Johannes Kepler University Linz, Altenberger Str. 69, A-4040, Austria

*Corresponding author: serpil.tekoglu@jku.at



Figure S1. X-ray diffraction (XRD) analysis of the sulphated cellulose nanocrystals (S-CNCs) and microcrystalline cellulose, MCC (Avicel). In red: S-CNCs, in blue: Avicel PH-101.

Table S1. Crystallinity Index and elemental analysis of pristine Avicel PH-101 and sulphated cellulose Nanocrystalsobtained from Avicel PH-101.

Entry	Sample	Crystallinity Index ¹ / %	Elemental Analysis				
			С	Н	Ν	S	O ²
1	Avicel	80	43.09	6.55	-	-	50.58
2	S-CNCs	75	40.11	6.55	0.00	0.60	52.73

¹Oxygen was calculated by subtracting from 100 C, H, N, and S.

²Crystallinity values are evaluated by Segal method CrI = $[(I_{200}-I_{am})]/I_{200} \times 100)$



Figure S2. XPS spectrum of PEDOT:S-CNCs. Peak deconvolution for the (a) O1s and (b) Fe2p signals

	Energy / eV	FWHM / 1	Amplitude / 1	Atom percentage / %
S2p	162.38	1.92	11046.98	5.68
O1s	531.07	2.65	91910.82	36.47
C1s	284.66	3.08	56945.2	57.85

 Table S2.
 Elemental analysis PEDOT:S-CNCs with the calculation of C, O, and S ratios.



Figure S3. (a) Raman spectra of pristine PEDOT and the PEDOT:S-CNC biocomposite, recorded using an excitation wavelength of 1064 nm and a resolution of 4 cm^{-1} . The shown spectra are the average of 1000 scans. (b) Enlarged picture of the peak shift of 4 cm^{-1} .



Figure S4. Optical appearance of S-CNCs aggregates on glass when imaged through polarization filter (recorded in reflective mode).

Table S3. Conductivity and synthesis conditions of PEDOT:S-CNCs in comparison to literature values and commercial PEDOT:PSS (Clevious PH 1000).

Sample Material	SO₃H / 100 glucose units	Mass ratio EDOT:CNC	Oxidant	EDOT: Oxidant molar ratio	Conductivity / S cm ⁻¹
PEDOT:S-CNC + A ^a	5	1:1	FeTos ¹	1:2.5	5 ± 1
PEDOT:S-CNC-SO ₃ H	3.50	1:2.5	APS ²	1:1	1.26 ± 0.13
+ A ^b					
PEDOT:S-CNC-	37.23	1:2.5	APS	1:1	25.6 ± 0.72
[high]-SO₃H + A ^b					
PEDOT:PSS + A ^a	-	-	-		280.0

^a Dispersion was prepared with the additives (A) 0.5% (v/v) DBSA, 10% (v/v) Glycerol and an additional 1% (v/v) of GOPS. The statistics for the conductivity values were performed on n = 11 thin film substrates (all in the range of approx. 140-270 nm), whereby the used value for each thin film is an average value calculated including three individual thickness and resistance measurements on each thin film.

^b Dispersion from literature reference ¹ was prepared with 5% (v/v) DMSO.

¹FeTos: Iron (III) tosylate, ²APS: Ammonium persulfate



Figure S5. Steady-state electrical characterization of PEDOT:PSS (Clevious PH 1000) based OECTs. (a) Transfer characteristics and the corresponding transconductance curve ($g_{max} = 27.3 \text{ mS}$ for $V_D = -0.7 \text{ V}$) and (b) the corresponding output characteristics for a single-channel OECT device (W = 2.11 mm, $L = 48.1 \mu \text{m}$, film thickness = 260 nm). (c) Transfer characteristics and transconductance ($g_{max} = 54.4 \text{ mS}$ for $V_D = -0.7 \text{ V}$) and (d) the corresponding output characteristics for an interdigitated OECT device (dimensions of one source-drain electrode pair: W = 0.54 \mu m, L = 44.9 \mu m, film thickness = 265 nm).

References

1 S. Atifi, M.-N. Mirvakili and W. Y. Hamad, ACS Appl. Polym. Mater., 2022, 4, 5626–5637.