Supplementary Information (SI) for Journal of Materials Chemistry C.


# Nanocrystalline cellulose-based mixed ionic-electronic conductor for bioelectronics

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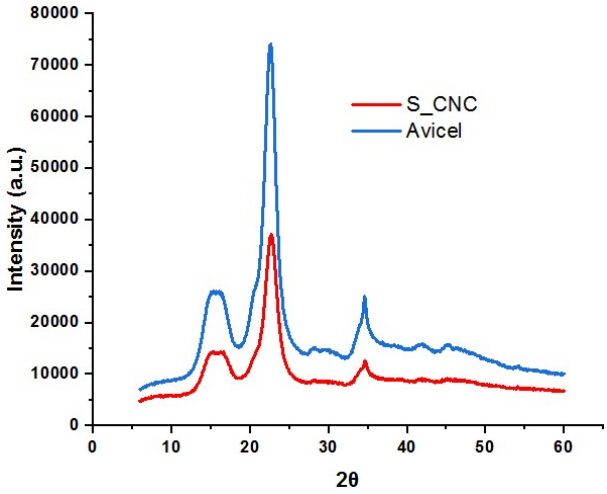


**Figure S1.** X-ray diffraction (XRD) analysis of the sulphated cellulose nanocrystals (S-CNCs) and microcrystalline cellulose, MCC (Avicel). In red: S-CNCs, in blue: Avicel PH-101.

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

| Entry | Sample | Crystallinity Index[1] / % | Elemental Analysis | | | | |
|---|---|---|---|---|---|---|---|
| | | | C | H | N | 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 |

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

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

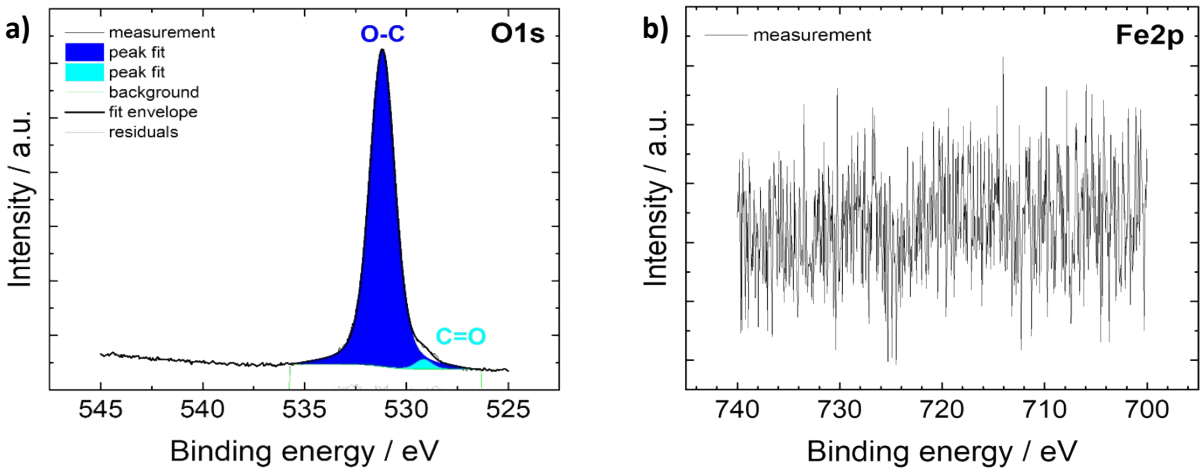


**Figure S2.** XPS spectrum of PEDOT:S-CNCs. Peak deconvolution for the (a) O1s and (b) Fe2p signals

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

| | 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 |

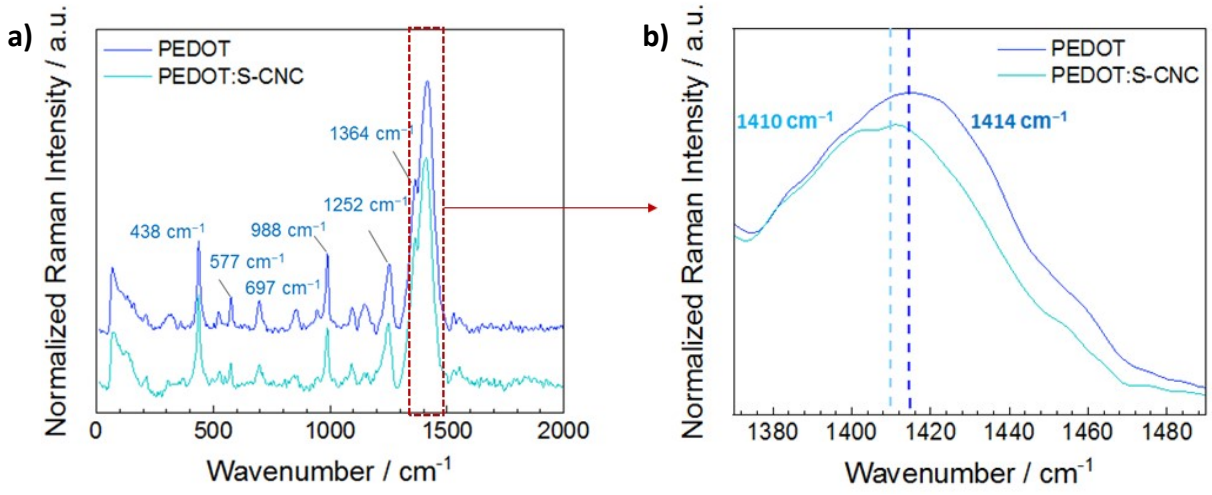


**Figure S3.** (a) Raman spectra of pristine PEDOT and the PEDOT:S-CNC biocomposite, recorded using an excitation wavelength of 1064 nm and a resolution of 4 $cm^{-1}$. The shown spectra are the average of 1000 scans. (b) Enlarged picture of the peak shift of 4 $cm^{-1}$.

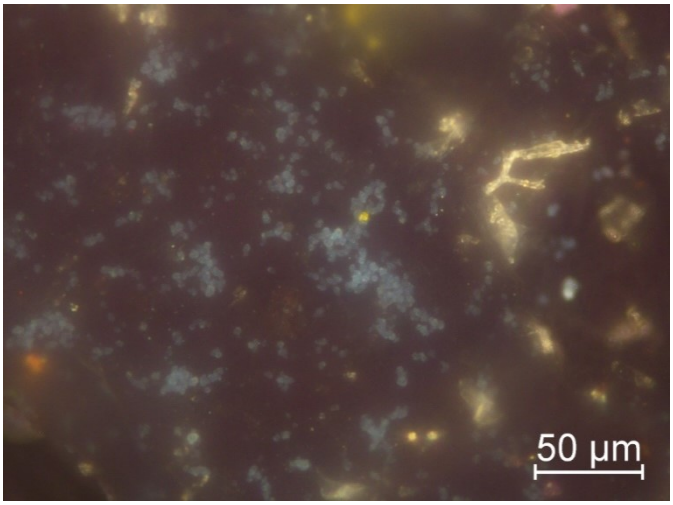


**Figure S4.** Optical appearance of S-CNCs aggregates on glass when imaged through polarization filter (recorded in reflective mode).

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

| Sample Material | $SO_3H$ / 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-$SO_3H$ + A[b] | 3.50 | 1:2.5 | APS[2] | 1:1 | 1.26 ± 0.13 |
| PEDOT:S-CNC-[high]-$SO_3H$ + A[b] | 37.23 | 1:2.5 | APS | 1:1 | 25.6 ± 0.72 |
| PEDOT:PSS + A[a] | - | - | - | | 2800 |

[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.

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

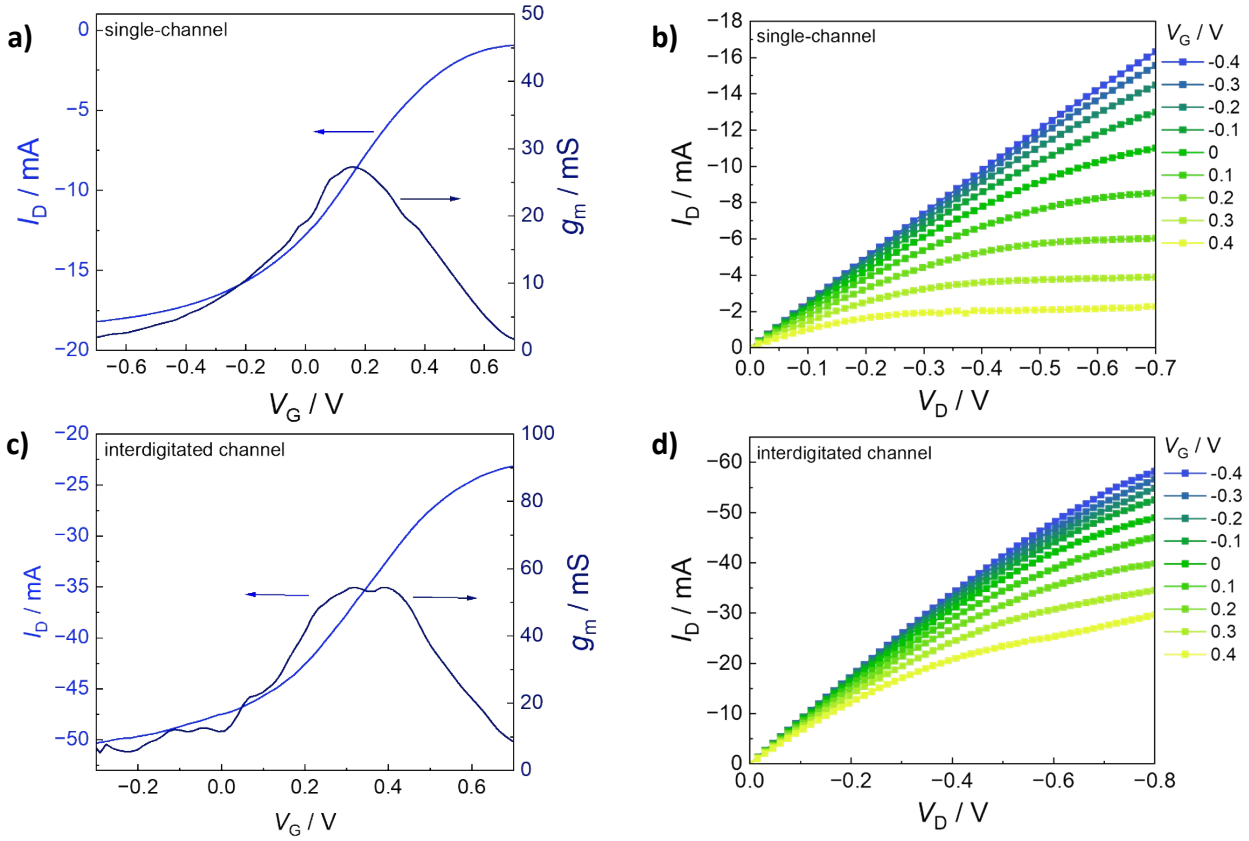


**Figure S5.** Steady-state electrical characterization of PEDOT:PSS (Clevious PH 1000) based OECTs. (a) Transfer characteristics and the corresponding transconductance curve ($g_{max}$ = 27.3 mS for $V_D$ = -0.7 V) and (b) the corresponding output characteristics for a single-channel OECT device ($W$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

## References

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