Electronic Supplementary Material (ESI) for Energy Advances.

Supporting information

In situ polymerization of EDOT onto sulfonated onion-like carbon for efficient pseudocapacitor electrodes

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Polymerization of EDOT to PEDOT@SPOLC, PEDOT@SPOLC/OLC and PEDOT@OLC

EDOT was distilled in vacuo prior to use. To produce **PEDOT@SPOLC**, EDOT (288 mg) was dispersed in aqueous **SPOLC** suspension (4 mL, 576 mg, 14 wt%) after the addition of ethanol (3 mL). For **PEDOT@OLC**, pristine OLC (1 g) and EDOT (570 mg) was dispersed in a mixture of water (4 mL) and ethanol (3 mL). For **PEDOT@SPOLC/OLC**, EDOT (575 mg) and pristine **OLC** (570 mg) was dispersed in aqueous **SPOLC** (4 mL, 576 mg, 14 wt%) with the addition of ethanol (3 mL).
To polymerize EDOT, an aqueous solution of sodium persulfate $Na_2S_2O_8$ (2.4 M) was added dropwise to a solution with an EDOT : $Na_2S_2O_8$ ratio of 1 : 2. After 24 hours of vigorous stirring using a hand-held coiled wire stirrer (i.e. a cappucino creamer). The resulting mixture was centrifuged (6000 rpm, 20 minutes) and the supernatant subsequently removed. The particles were redispersed and centrifuged six more times, alternating between ethanol and water as solvents.

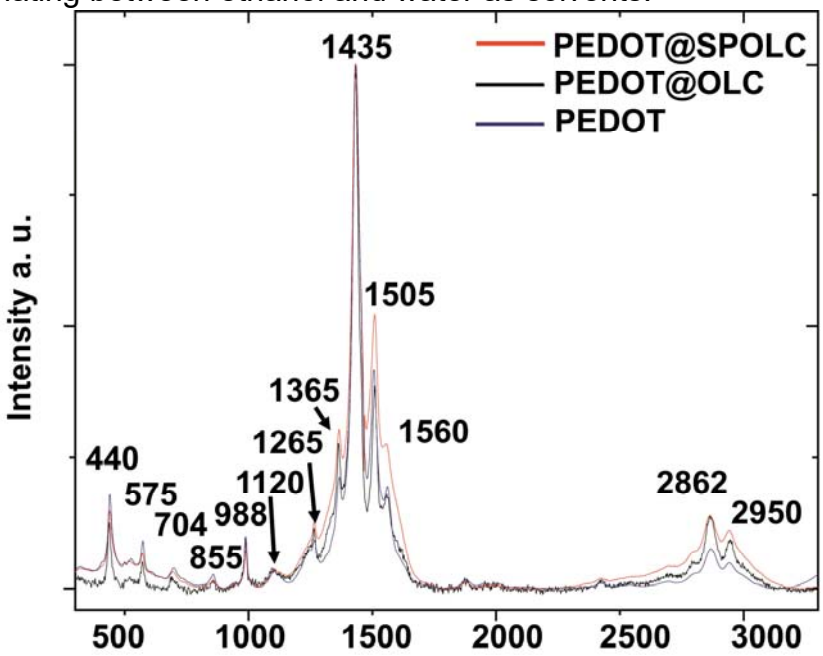

Fig. S1: Raman spectra of **PEDOT@SPOLC**, **PEDOT@OLC** and PEDOT (excitation wavelength: 532 nm).

Tab. S1: Assignment of Raman modes for functional groups of PEDOT

Raman shift (cm^{-1})	Assignment
440, 575, 988	oxy- ethylene ring CO deformation
1128	C-O-C deformation
1265	CαCα stretching and CH bending
1435, 1405, 1560	symmetric and asymmetric C=C stretching
1365	thiophene ring CβCβ stretching
2870, 2965	OH and CH stretching

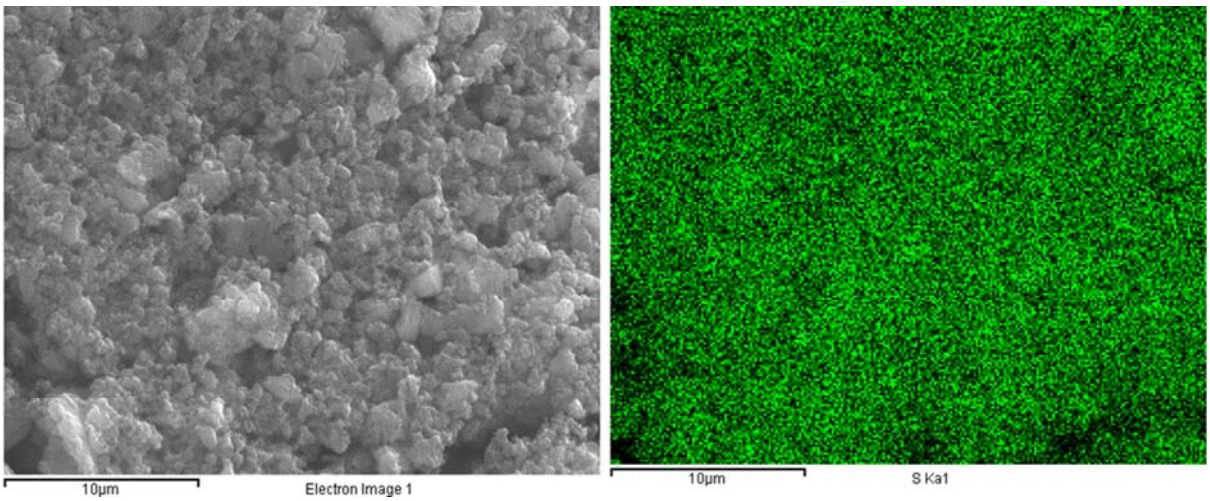

Fig. S2: SEM picture (left) and its corresponding EDX measurement (tracking sulfur) for the **PEDOT@SPOLC** composite.

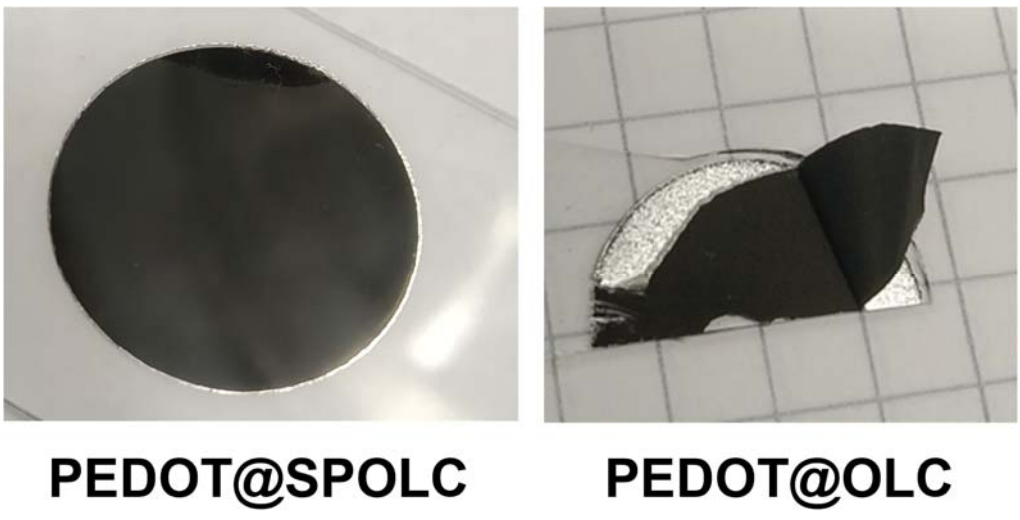

Fig. S3: Differences in electrode manufacturing of **PEDOT@SPOLC** and **PEDOT@OLC**, where **PEDOT@OLC** shows delamination despite the identical electrode manufacturing procedure. Variations also did not lead to improved electrode characteristics.

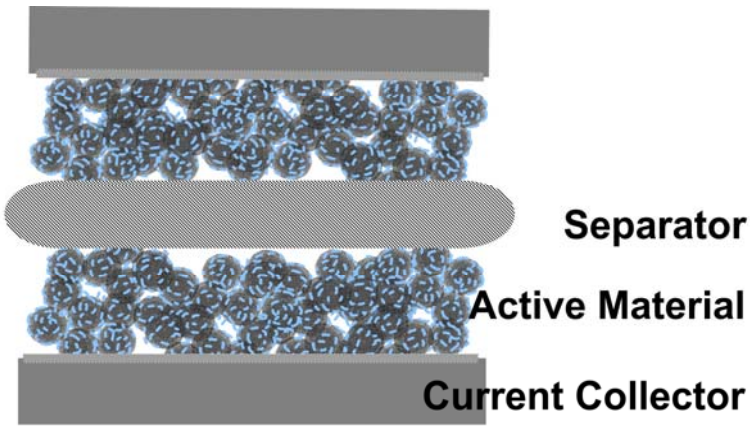

Fig. S4: Schematic overview of a symmetric supercapacitor setup with separator, active material and current collector.

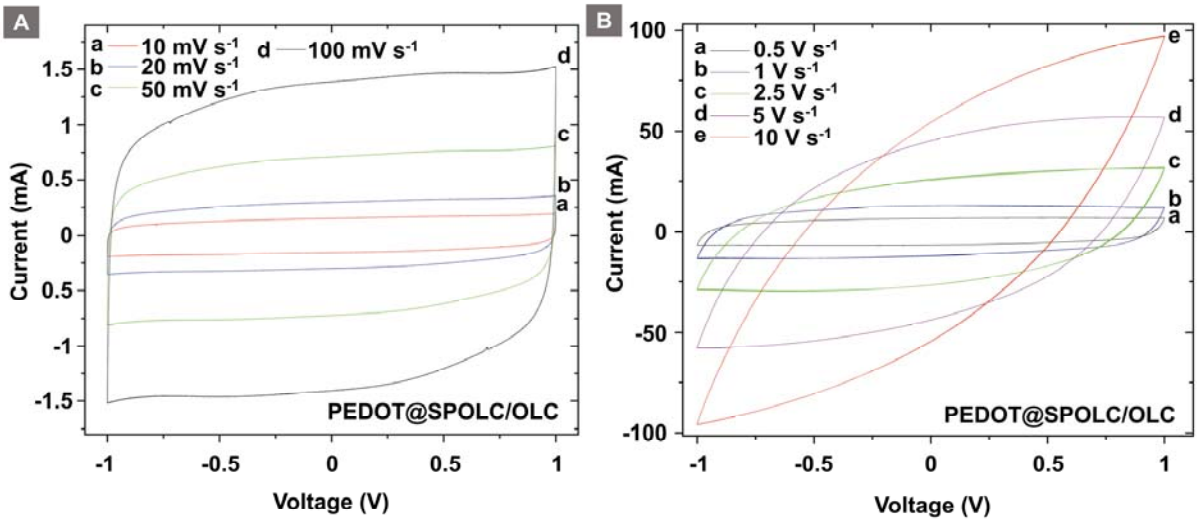

Fig. S5: Cyclic voltammogram with 1 V potential window of **PEDOT@SPOLC/OLC** at low (A) and high (B) scan rates.

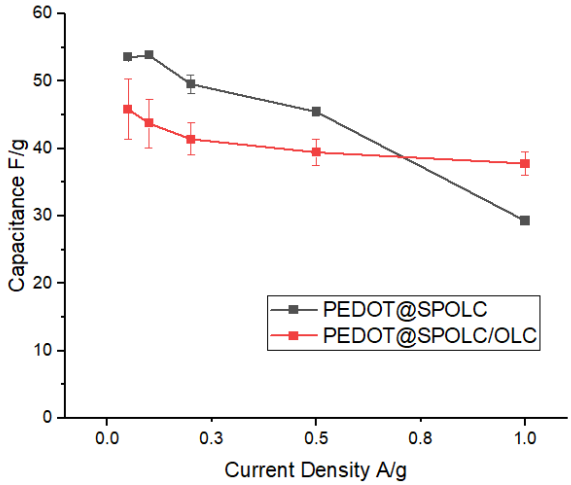

Fig. S6: Discharge capacitance of PEDOT@SPOLC and PEDOT@SPOLC/OLC

Tab. S2: Additional information on capacitances and conductivities of polymer-carbon composite materials

Material composition	**conductivity**	**capacitance**	**reference**
optimized PEDOT film	6529 S cm^{-1}	0.99 F cm^{-2} (PEDOT:PSS electrode)	[S1]
PEDOT on textile carbon fibres	790 S cm^{-1}	184 F g^{-1}	[S2]
polyvinyl alcohol-graphene oxide fibre coated with PEDOT	n.r.	224 F g^{-1}	[S3]
Carbon nanoonions + resorcinol-formaldehyde resin	n.r.	160 F g^{-1}	[S4]
Carbon nanoonions / PEDOT:PSS 1:1	n.r.	95 Fg^{-1}	[S5]
PEDOT:PSS with EDOT:PSS 1:11	1000 S cm^{-1}		[S6]
PEDOT:PSS film + Triton X-100	up to 1880 S cm^{-1}		[S7]

n.r.: not reported

Several benchmark numbers for the conductivity of different PEDOT containing devices have been reported in the literature. Ranging from 1000 S/cm [15] to 6200 S/cm [1]. This changes when the PEDOT is incorporated into nanomaterial composites. As an example, PEDOT on textile carbon fibers showed a conductivity of 790 S/cm [5]. While not directly measured, we assume reduced electrical conductivity for our system as well.

Another CNO/PEDOT:PSS composite, consisting of a 1:1 ratio of carbon material and polymer, was reported to achieve capacitances of up to 95 F/g [27], compared to our **PEDOT@OLC** with a capacitance of 77 F/g.

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