Supplementary Materials: In-Situ Approaches for the Preparation of Polythiophene-Derivative Cellulose Composites with High Flexibility and Conductivity

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^{1}H-NMR spectrum of PDBProDOT

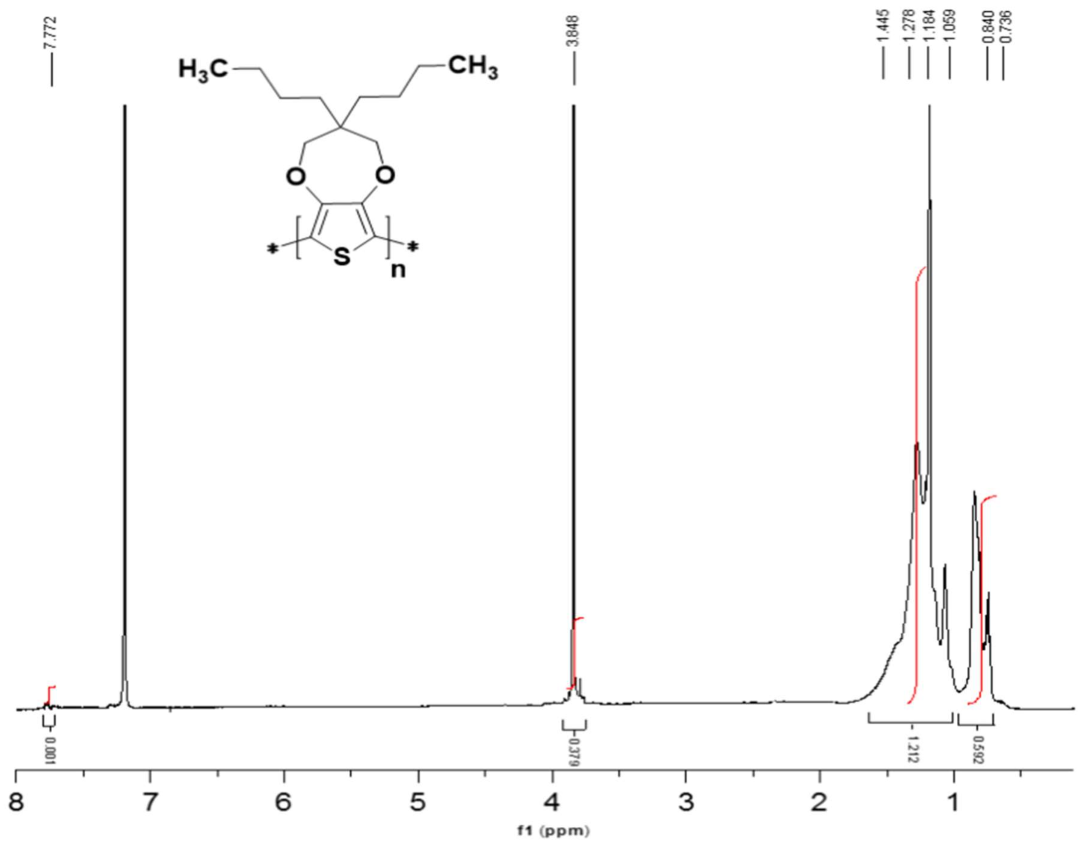

Figure S1. ^{1}H-NMR spectra of PDBProDOT.

Gel Permeation Chromatography (GPC)

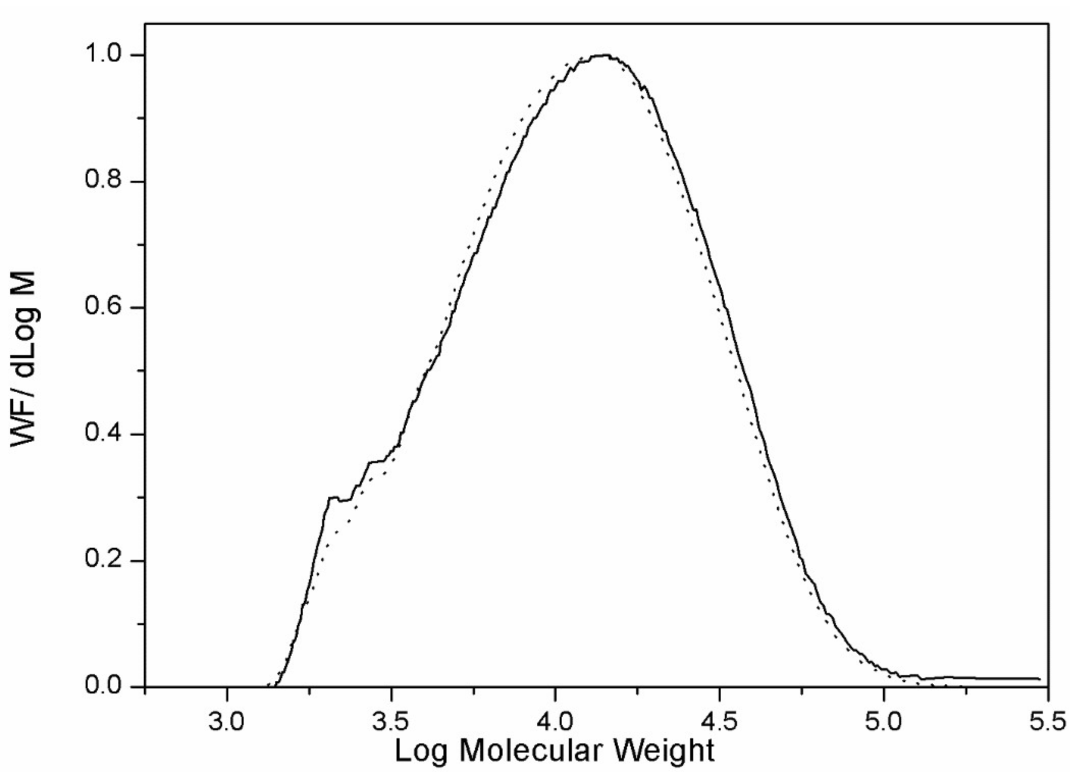

Figure S2. GPC traces of PDBProDOT synthesized using $Fe(Tos)_3$ ($0.6\ mol \cdot L^{-1}$) in ethanol. Measurements carried out using THF as the eluent and calibrated vs. polystyrene standards. (—) PDBProDOT-1; (--) PDBProDOT-2.

Thermogravimetric Analysis (TGA)

Table S1. T_{50}, T_5 and % residue for cellulose substrates and composite materials.

Pure polymer samples	***T50 (°C)***	***T5 (°C)***	***Residue (% wt.)***
PEDOT:Tos	343	186	1.3
PEDOT:Trif	360	265	2.1
PEDOT:PSS	418	114	6.3
PDBProDOT	410	252	1.0
Cellulosic substrates			
W	339	276	0.0
A	211	189	0.1
C	333	285	0.2
In-situ polymerization			
PTsWCt	319	151	1.0
PTsACt	290	173	0.8
PTsWSp	335	190	1.2
PTsASp	367	172	0.5
PTfWCt	303	100	0.3
PTfACt	358	100	5.2
PTfWSp	313	202	1.2
PTfASp	340	135	1.2
CP dispersions			
PPsACt	337	155	5.3
PPsCCt	353	151	4.1
PPDACt	223	190	0.1
PPDASp	198	191	0.3
PPDCSp	309	239	1.1

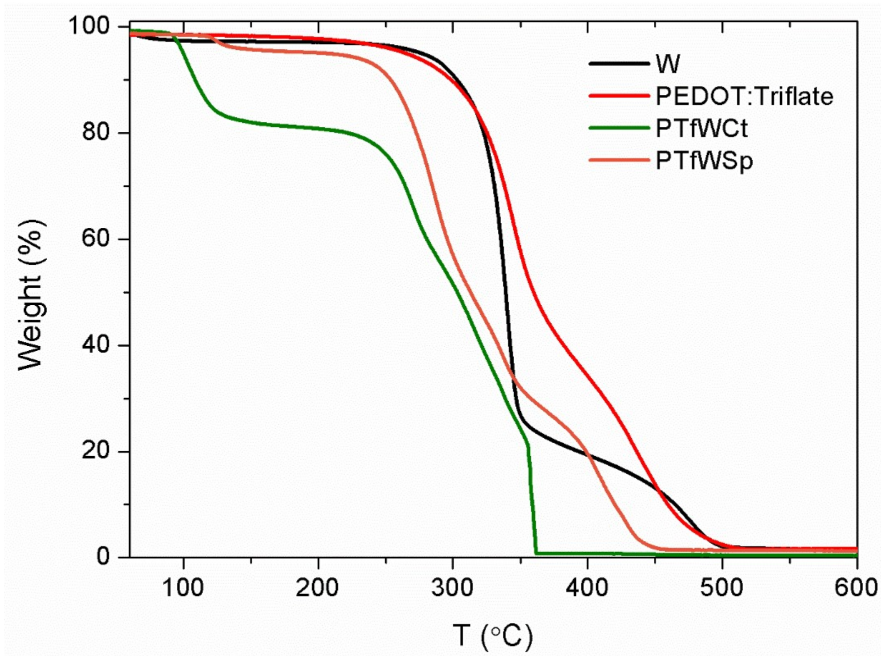

Figure S3. TGA curves of PEDOT:Triflate composite materials onto W substrates.

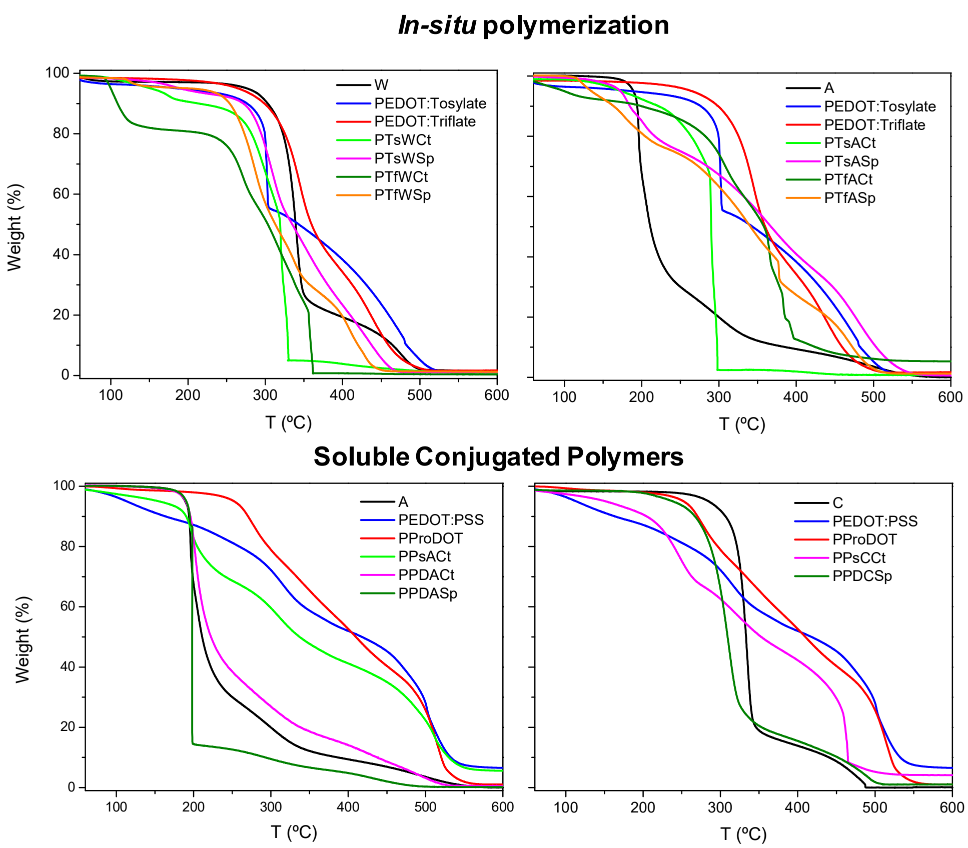

Figure S4. TGA curves in air atmosphere of polythiophene-derived composite materials.

Infrared Spectroscopy (ATR-FTIR)

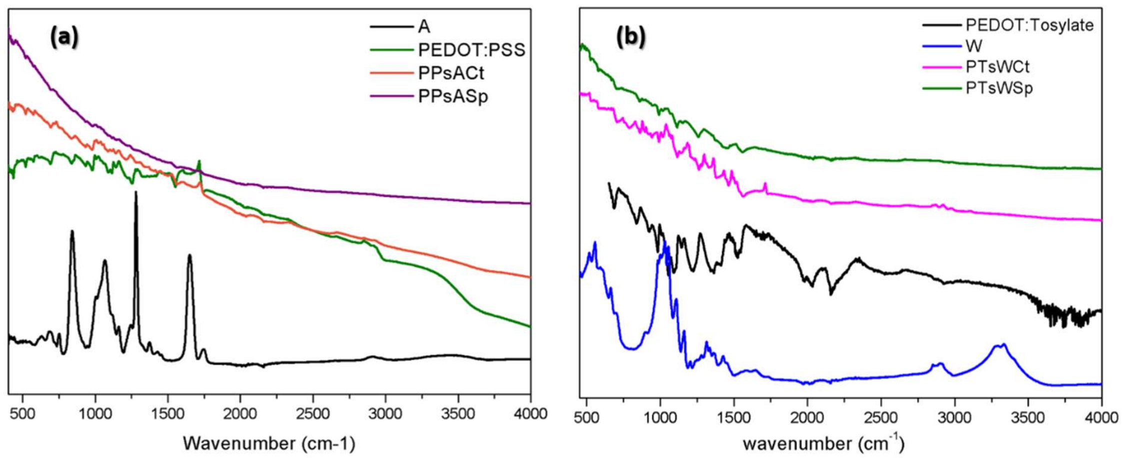

Figure S5. FTIR-ATR spectra of polythiophene-derivatives composite materials. (a) PEDOT:PSS/acetate cellulose composite materials; (b) PEDOT:Tosylate/Whatman paper composite materials.

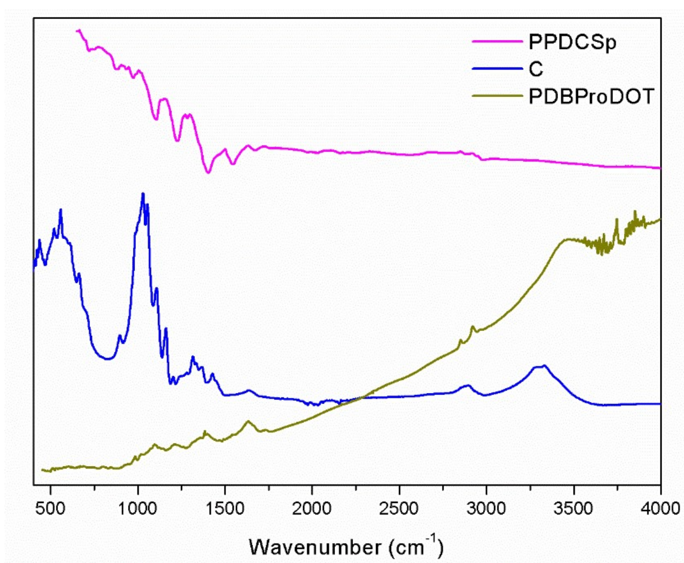

Figure S6. FTIR-ATR spectra of PDBProDOT/cellulose microfibers composite material.

Dielectric spectra of CPs and composite materials

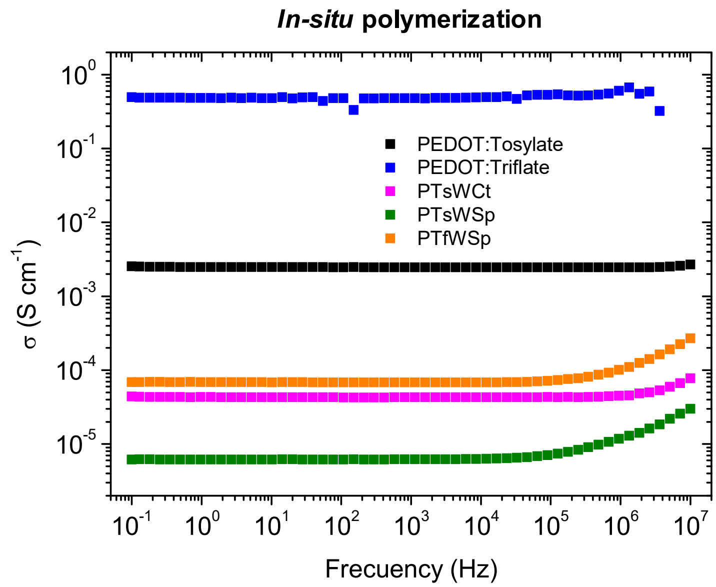

Figure S7. Bulk conductivity dependence with frequency for selected cellulose composite materials obtained by *in-situ* polymerization.

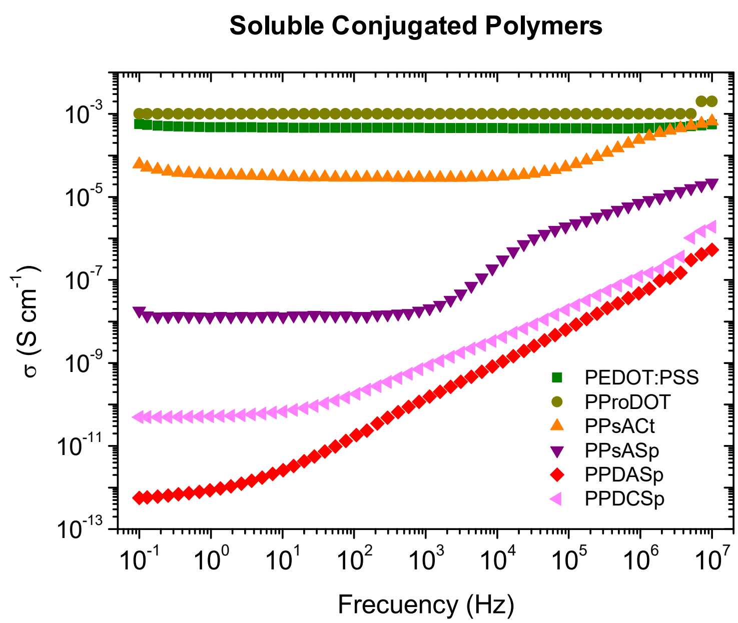

Figure S8. Bulk conductivity dependence with frequency for selected cellulose composite materials obtained by CPs solutions.

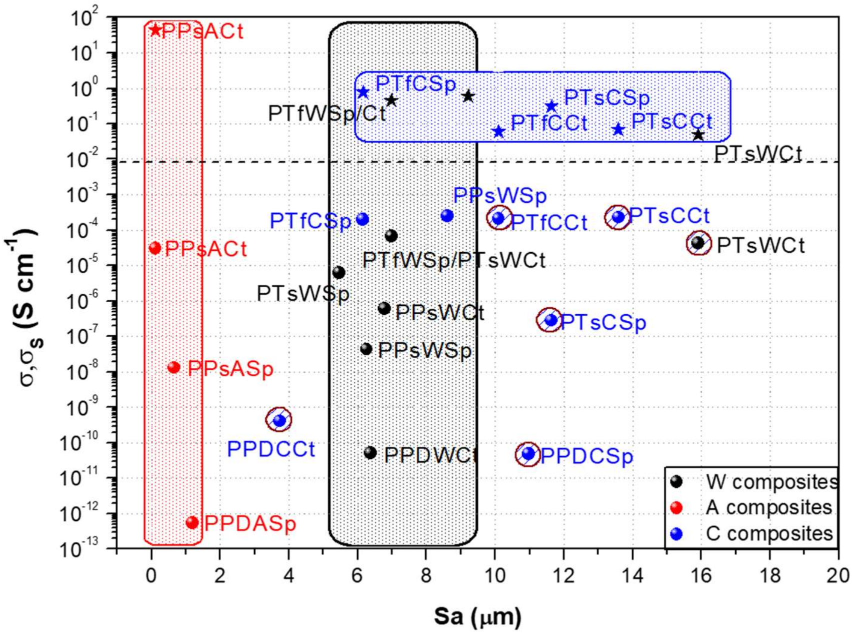

Figure S9. σ_s (stars) and σ (circles) trends on Sa roughness: (a) for composite materials prepared on different substrates: Whatman® paper (black); acetate cellulose (red) and cellulose microfibers (blue).

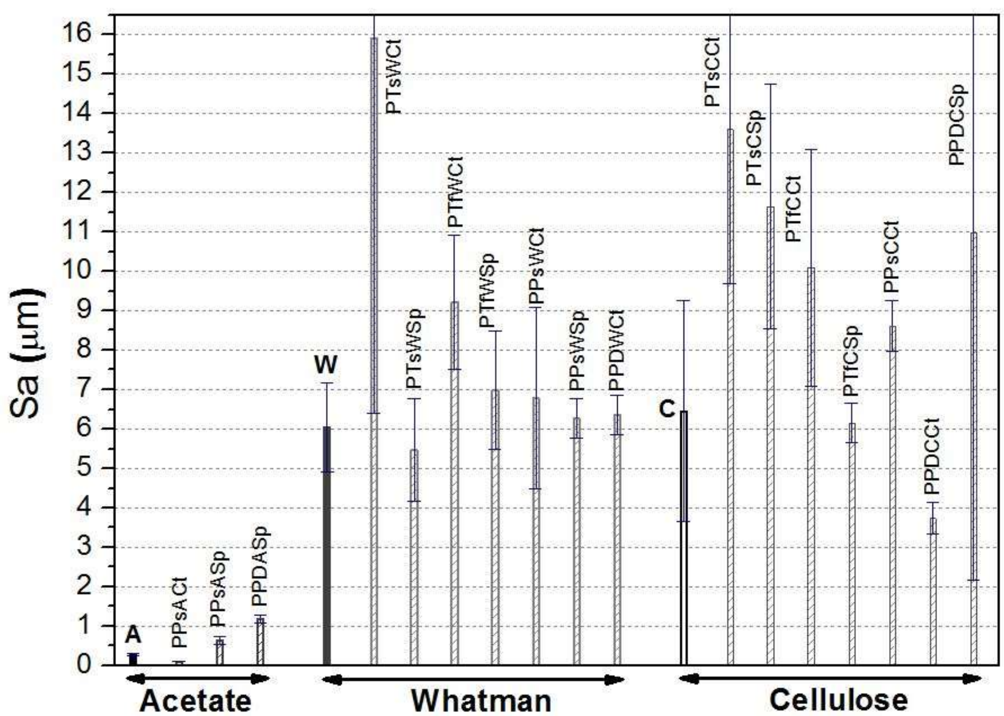

Figure S10. Sa values of raw substrates (solid bars) and selected composite materials (patterned bars).

Table 2. σ, $σ_s$ and Sa of composite materials.

In-situ polymerization	***σ (S·cm⁻¹)***	***σ$_s$ (S·cm⁻¹)***	***Sa (μm)***
PTsWCt	4.2×10^{-5}	0.05	15.9 ± 9.5
PTsCCt	2.3×10^{-4}	0.07	13.6 ± 3.9
PTsWSp	6.3×10^{-6}	*	5.5 ± 1.3
PTsCSp	2.8×10^{-7}	0.31	11.6 ± 3.1
PTfWCt	3.6×10^{-4}	0.60	9.2 ± 1.7
PTfCCt	2.1×10^{-4}	0.06	10.1 ± 3.0
PTfWSp	6.7×10^{-5}	0.46	7.0 ± 1.5
PTfCSp	2.0×10^{-4}	0.77	6.2 ± 0.5
Soluble CPs			
PPsWCt	6.0×10^{-7}	*	6.8 ± 2.3
PPsACt	3.0×10^{-5}	43.67	0.1 ± 0.002
PPsWSp	4.3×10^{-8}	*	6.3 ± 0.5
PPsASp	1.3×10^{-8}	*	0.6 ± 0.1
PPDWCt	5.1×10^{-11}	*	6.4 ± 0.5
PPDCCt	4.2×10^{-10}	*	3.7 ± 0.4
PPDASp	5.6×10^{-13}	*	1.2 ± 0.1
PPDCSp	4.9×10^{-11}	*	11.0 ± 8.8