ADVANCED MATERIALS

Supporting Information

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Repurposing Poly(3-hexylthiophene) as a Conductivity-Reducing Additive for Polyethylene-Based High-Voltage Insulation

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Table S1. Effect of different types of conductivity-reducing additives on the DC conductivity of LDPE expressed in terms of the DC conductivity σ_{DC} of the additive-containing resin relative to the DC conductivity of the neat LDPE resin σ_{DC}^{PE}, and as the efficiency $\eta = (\sigma_{DC}^{PE}/\sigma_{DC})/\phi$, where ϕ is the additive content in wt%. We limit our survey to DC conductivity measurements where the electric field was applied for at least 1 h, alhough values approaching the steady-state are only obtained for much longer measurements. [a] The matrix is crosslinked LDPE (XLPE); [b]Values of the reported charging current in A.

	type of additive	ϕ (wt%)	σ_{DC}^{PE} (S m^{-1})	σ_{DC} (S m^{-1})	$\sigma_{DC}^{PE}/\sigma_{DC}$	η (wt%$^{-1}$)	Ref.
inorganic nanoparticles	ZnO	3	3×10^{-14}	1×10^{-16}	300	100	[1]
	MgO	3	3×10^{-14}	5.5×10^{-16}	54	18	[2]
	Al_2O_3	3	3×10^{-14}	1×10^{-15}	30	10	[3]
	SiO_2	2	[b] 6.5×10^{-10}	[b] 8×10^{-11}	8	4	[4]
polyolefins	HDPE [a]	1	1×10^{-14}	8×10^{-16}	13	13	[5]
hybrid	HDPE + Al_2O_3	4 + 3	1×10^{-14}	3×10^{-17}	333	48	[6]
carbon allotropes	graphene	0.1	1×10^{-14}	1.1×10^{-15}	9	90	[7]
	graphene	0.1	2.5×10^{-13}	1.2×10^{-13}	2	20	[7]
	graphene oxide [a]	0.01	5.5×10^{-14}	8×10^{-15}	7	700	[8]
voltage stabilizers	anthracene	0.5	2×10^{-11}	4×10^{-12}	5	10	[9]
	3- aminobenzoic acid [a]	1	9×10^{-13}	5×10^{-14}	18	18	[10]
	4,4'-dihydroxy benzophenone	0.5	7×10^{-15}	1.5×10^{-15}	5	10	[11]
conjugated polymers	P3HT	0.0005	7×10^{-14}	2.6×10^{-14}	3	6'000	this work

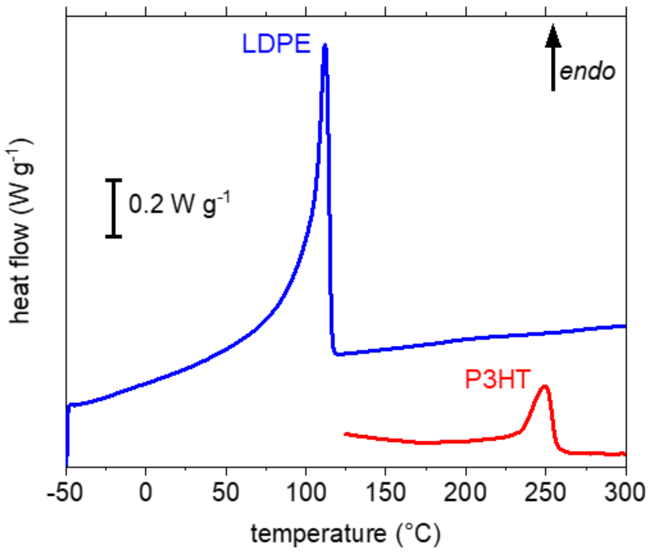

Figure S1. Second heating DSC thermograms of neat LDPE and P3HT.

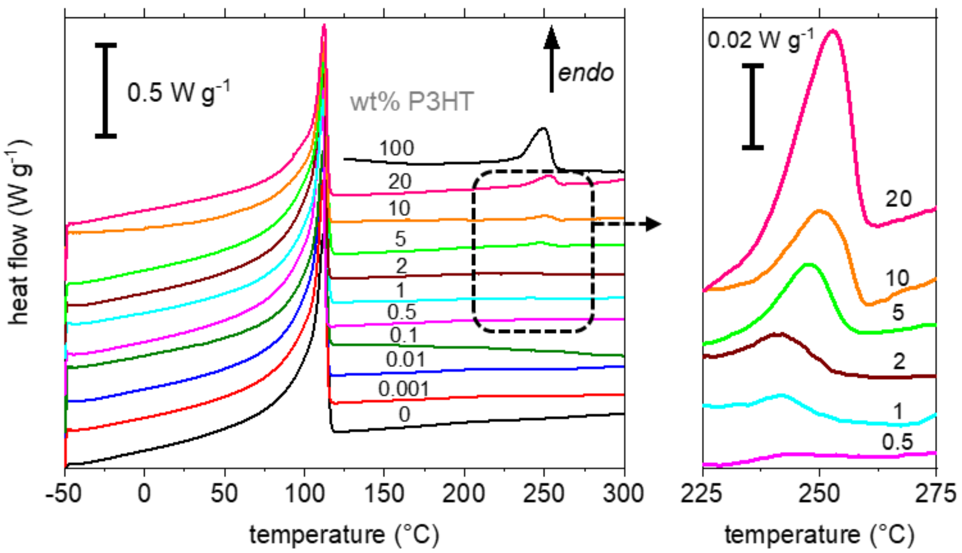

Figure S2. Second heating DSC thermograms of P3HT:LDPE blends.

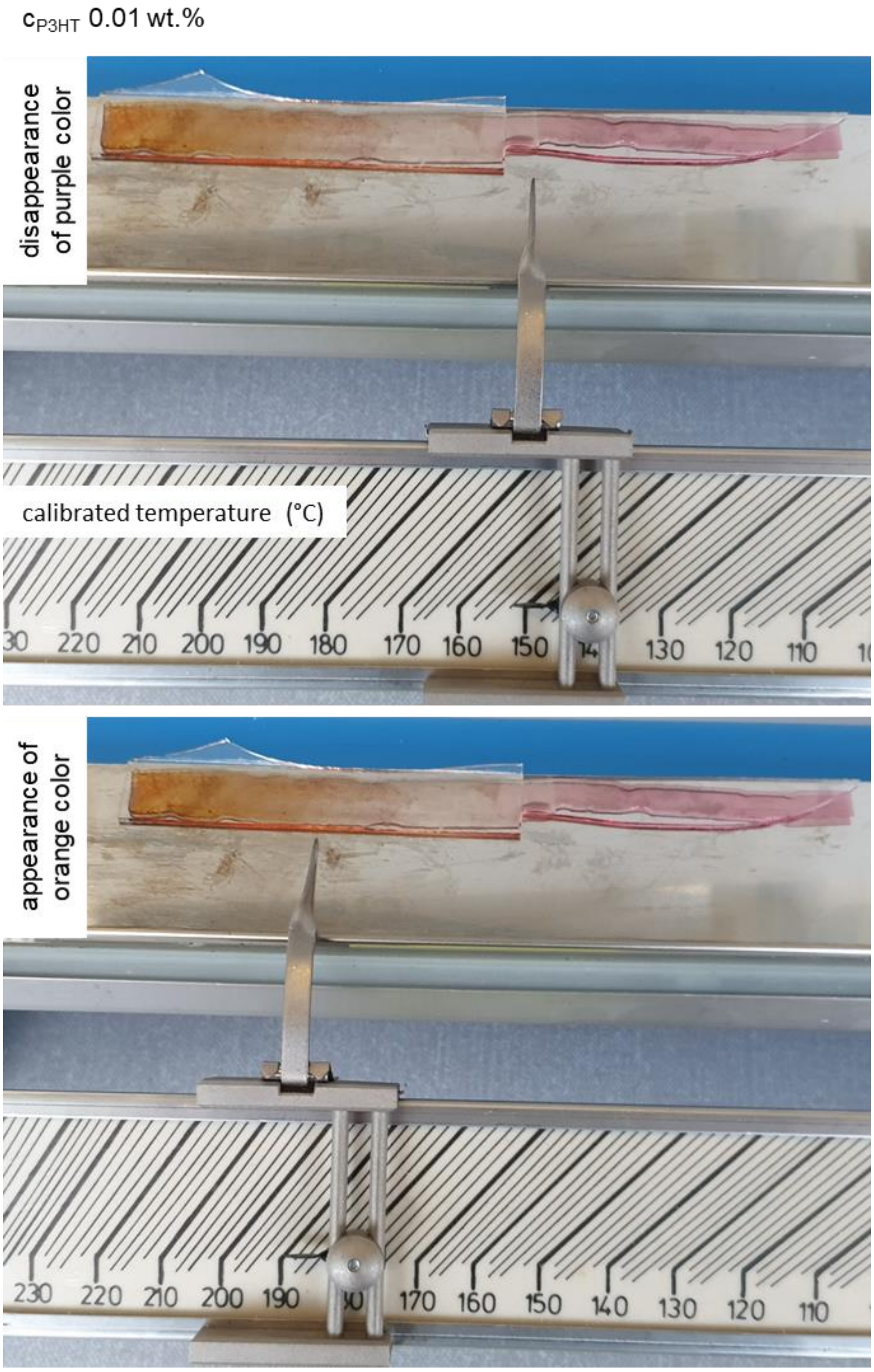

Figure S3. Visual detection with a Kofler bench of melting of P3HT in a P3HT:LDPE blend c_{P3HT} = 0.01 wt%; sample thickness ~ 0.3 mm.

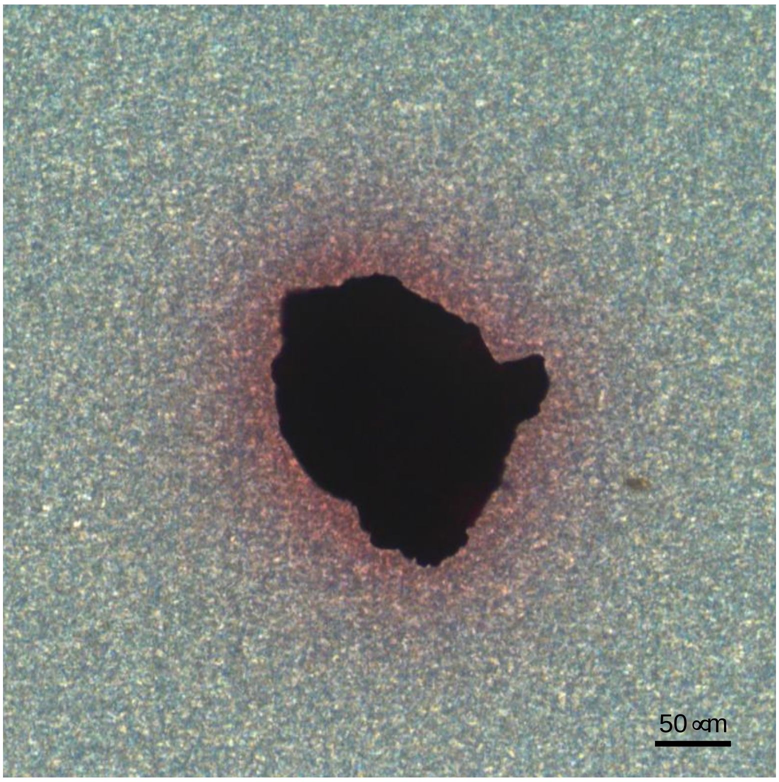

Figure S4. Polarized optical micrograph of a P3HT grain placed on top of a 30 μm thick sheet of polyethylene, sandwiched between glass slides and heated for 2 min at 250 °C.

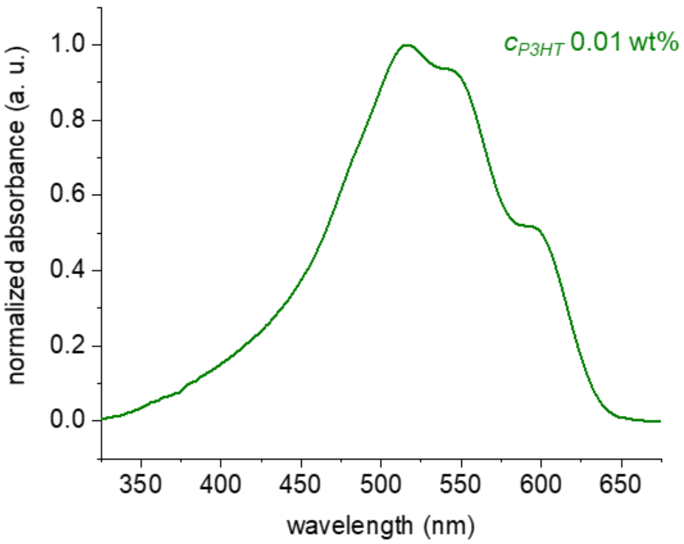

Figure S5. UV-Vis absorbance spectrum of a melt-pressed plaque of a blend with c_{P3HT} = 0.01 wt%; sample thickness ~ 1 mm.

According to standard Franck-Condon standard progression, the relative intensity of the vibronic replica is given by[12]:

$$I_{0\to m} \propto (\hbar\omega)^3 n_f^3 \frac{S^m e^{-S}}{m!} \qquad \text{(Eqn. 1)}$$

where n_f is the refractive index at the given photon energy of $\hbar\omega$ at optical frequency of ω, m is the Franck−Condon vibronic index, and S is the Huang−Rhys factor. PL spectrum can be modeled as a modified Franck-Condon progression with a variable 0–0 amplitude[12]:

$$I(\omega) \propto (\hbar\omega)^3 n_f^3 e^{-S} \times [(\alpha\Gamma(\hbar\omega - E_0) + \sum_{m=1} \frac{S^m}{m!} (\hbar\omega - (E_0 - mE_P))] \qquad \text{(Eqn. 2)}$$

where E_0 is the 0–0 transition energy, E_P is the phonon energy of the C=C symmetric stretch, Γ is the line-shape function (simplified to be purely Gaussian with a constant width), and α is a constant, known as relative intensity of 0–0 band. In above fitting, the parameters of S, E_P are kept constant and they are respectively, 1 and 0.18 eV. The fitted α and E_0 are obtained respetively, 0.52 ± 0.01 and 1.916 ± 0.002, for the PL spectra of P3HT:LDPE blends with c_{P3HT} = 0.1 wt%.

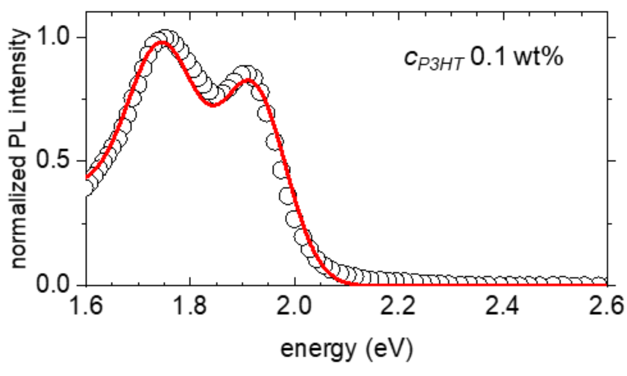

Figure S6. PL spectrum of a melt-pressed plaque of a blend with c_{P3HT} = 0.1 wt% as well as a fit using the modified Franck-Condon model proposed by Spano et al.[12-15] (red solid line); sample thickness ~ 0.1 mm.

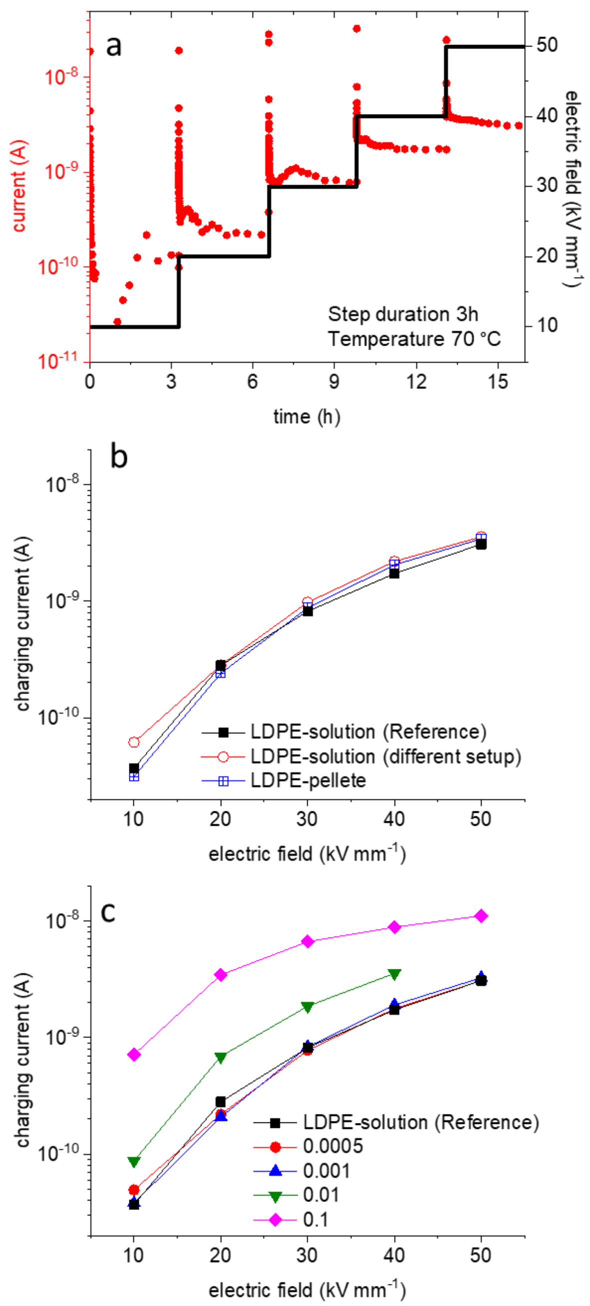

Figure S7. High-voltage charging current of P3HT:LDPE blends at 70 °C. (a) the applied step-wise electric field and resulting current as a function of time, (b) charging current at the end of each 3 h step as a function of the applied electric field for samples measured with two different setups (with the same arrangments and electrode dimensions) as well as different processing protocols (melt-pressing of precipitated material vs. as-received pellets), (c) quasi-steady-state charging current at the end of each 3 h step as a function of the applied electric field for blends with c_{P3HT} = 0.0005-0.1 wt%.

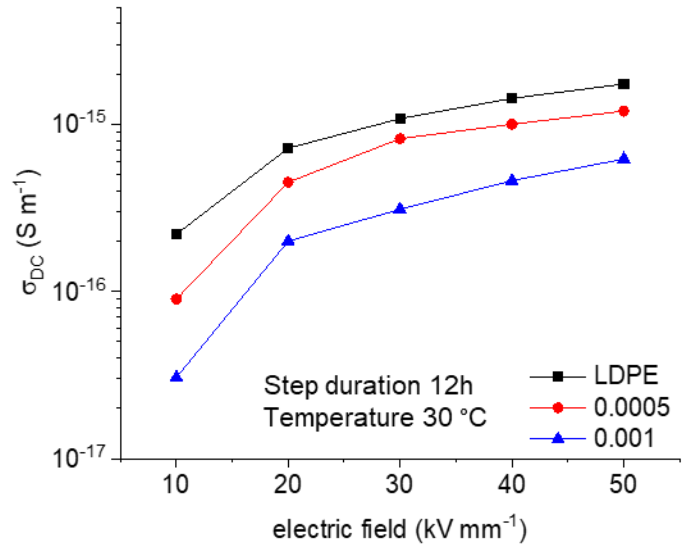

Figure S8. High-voltage DC conductivities of P3HT:LDPE blends at 30 °C.

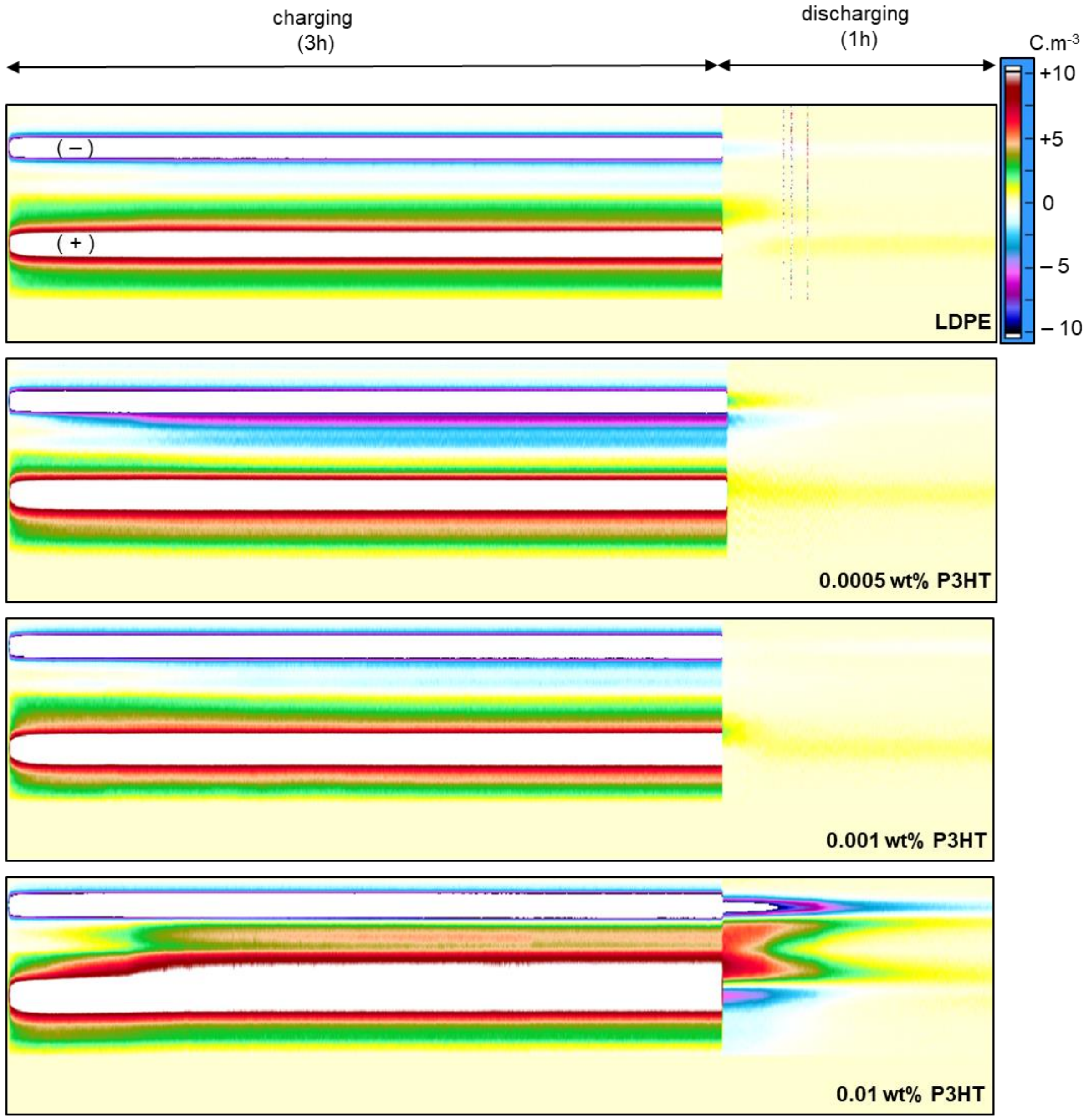

Figure S9. Space charge distribution in melt-pressed plaques of LDPE and P3HT:LDPE blends at 70 °C; PEA space charge distribution of film samples during charging (3 h, applied electric field of 50 kV mm^{-1}) and depolarization (1 h, removal of the electric field). The position of electrodes is shown as (-) and (+) for LDPE.

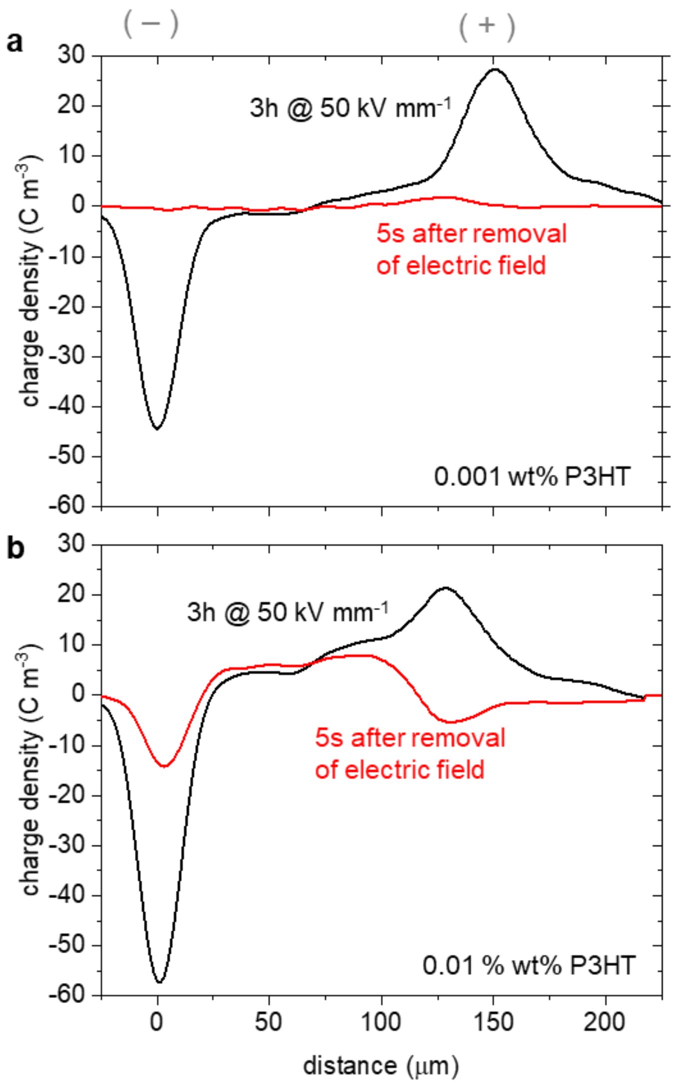

Figure S10. Space charge distribution of P3HT:LDPE blends with (a) c_{P3HT} = 0.001 wt% and (b) c_{P3HT} = 0.01 wt.% at 70 °C.

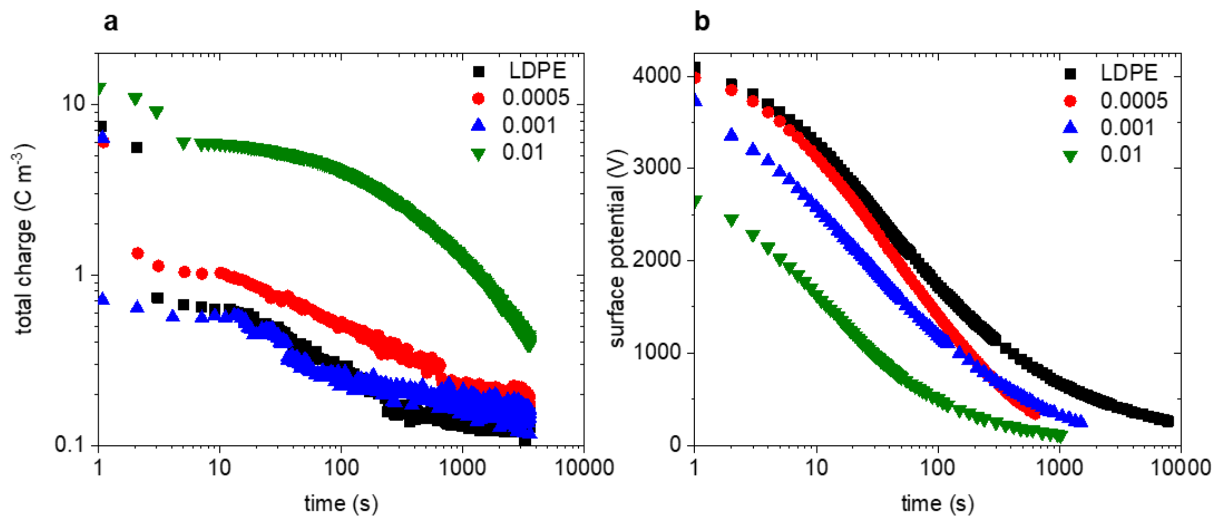

Figure S11. (a) PEA total charge (not net-charge) decay during 1 h discharging, (d) isothermal surface potential decay (ISPD) after charging of 0.1 mm thick melt-pressed plaques at 8 kV.

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