Electronic Supplementary Information

Voltage Losses in Indoor Light Harvesting Organic Photovoltaic Devices: Case Study of Green Solvent Processed PM6/IT-4Cl Devices

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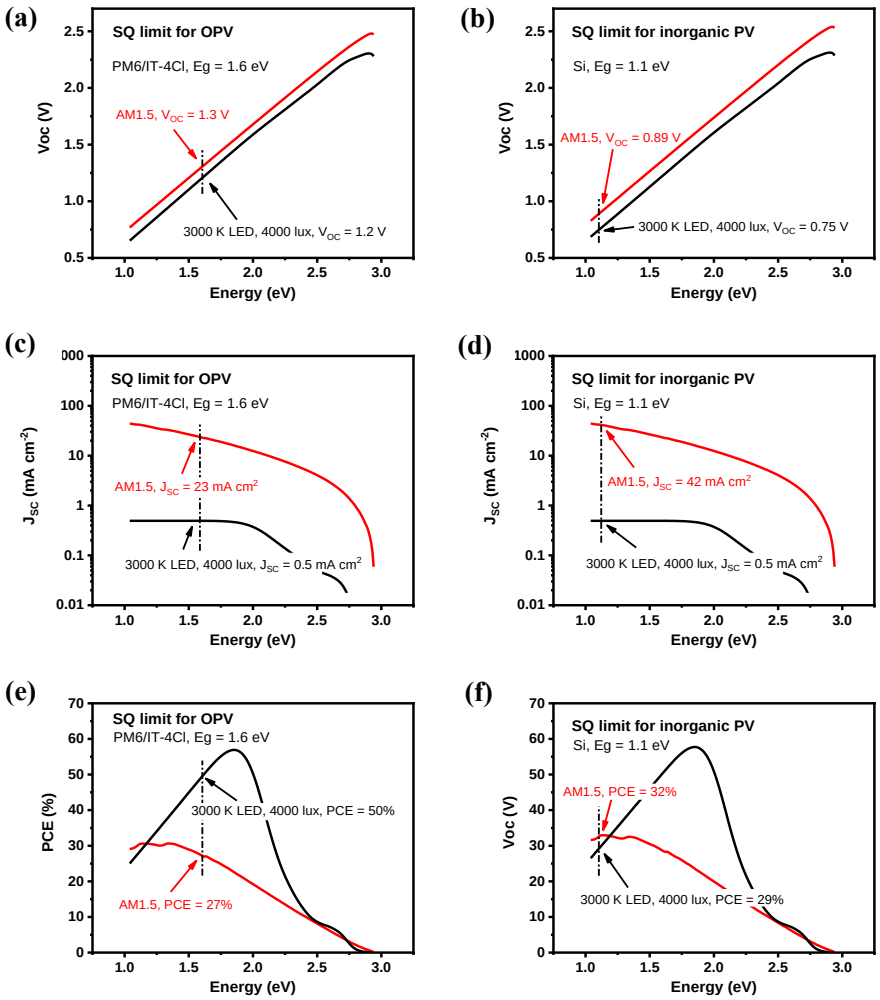

Figure S1. Theoretical upper limits as a function of E_g for organic PV devices, compared to those for inorganic PV devices, derived using the Shockley-Queisser method. For the calculation of the upper limit for inorganic devices, the expression used for the dark saturation current density (J_0) is the same as that used in the original paper by Shockley and Queisser (*J. Appl. Phys.* 1963, 32(3), 510). For OPV devices, the expression for the J_0 used is (*Phys. Rev. B* 2010, 81(12), 125204),

$$J_0 = \frac{2\pi q f E_g}{h^3 c^2} \exp(-\frac{E_g}{kT})$$

where q is elementary charge, h is the Planck constant, c is light speed, k is the Boltzmann constant, T is temperature, f is a pre-factor used to normalize the value of J_0 to that predicted by the Shockley-Queisser theory.

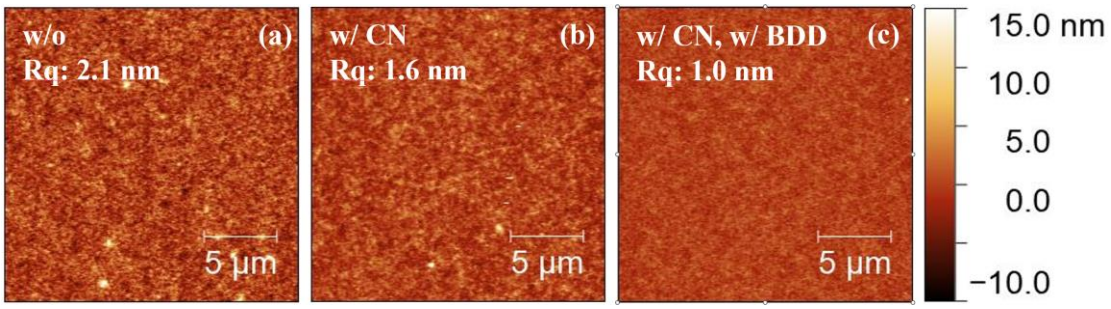

Figure S2. AFM images for the thin films of **a)** PM6/IT-4Cl without additive, **b)** PM6/IT-4Cl with CN (0.5 vol%) and **c)** PM6/BDD/IT-4Cl, with BDD content of 20% and with CN (0.5 vol%). The AFM images indicate that for the active layers processed without additives, the root-mean-square (RMS) surface roughness is high, over 2 nm, and pin-holes represented by the dark spots are observed. For the active layers PM6/IT-4Cl processed with CN, the RMS is reduced. Although the topographic feature is not distinctly different from that of the active layer without additives, the dark spots representing pin-holes are no long observed. Finally, for the active layers processed with CN and BDD, the surface is much smoother, indicating an altered morphological property, compared to the active layers processed without additive or with only CN.

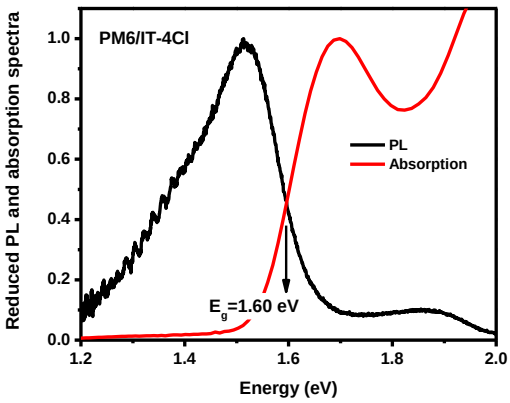

Figure S3. Normalized and reduced PL and absorption spectra for the determination of E_g for the thin film of PM6/IT-4Cl, using the method described in the literature (*Sustain. Energy Fuels* 2018, 2(3), 538).

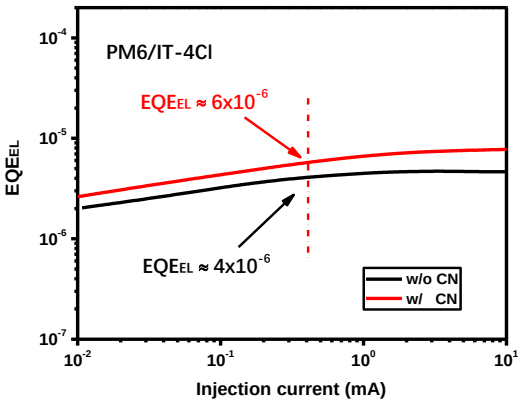

Figure S4. *EQE*$_{EL}$ of the OPV devices based on PM6/IT-4Cl processed with and without CN.

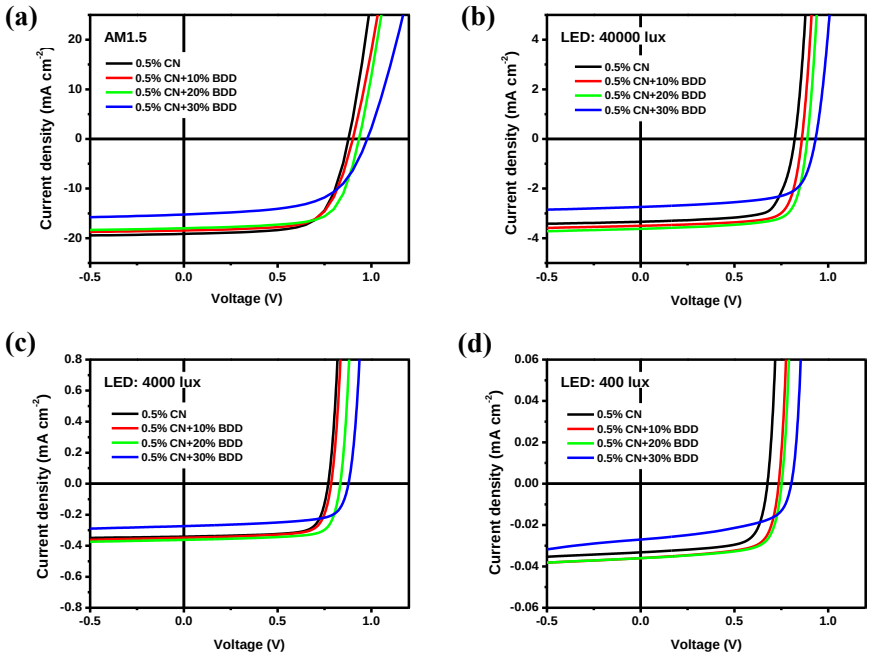

Figure S5. *J-V* curves of the OPV devices based on PM6/BDD/IT-4Cl with different BDD content, measured under different light sources: **a)** AM1.5, **b)** 40000 lux LED (3000 K), **c)** 4000 lux LED (3000 K), and **d)** 400 lux LED (3000 K).

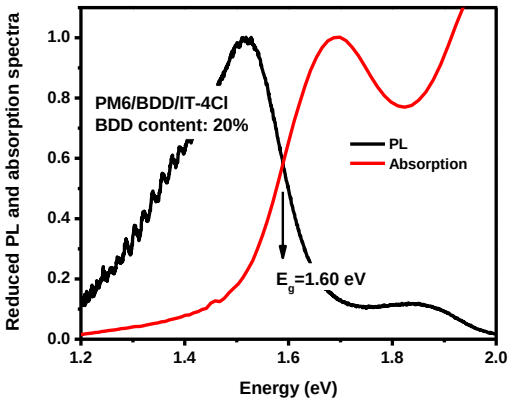

Figure S6. Normalized and reduced PL and absorption spectra for the determination of E_g for the thin film of PM6/BDD/IT-4Cl, with BDD content of 20%.

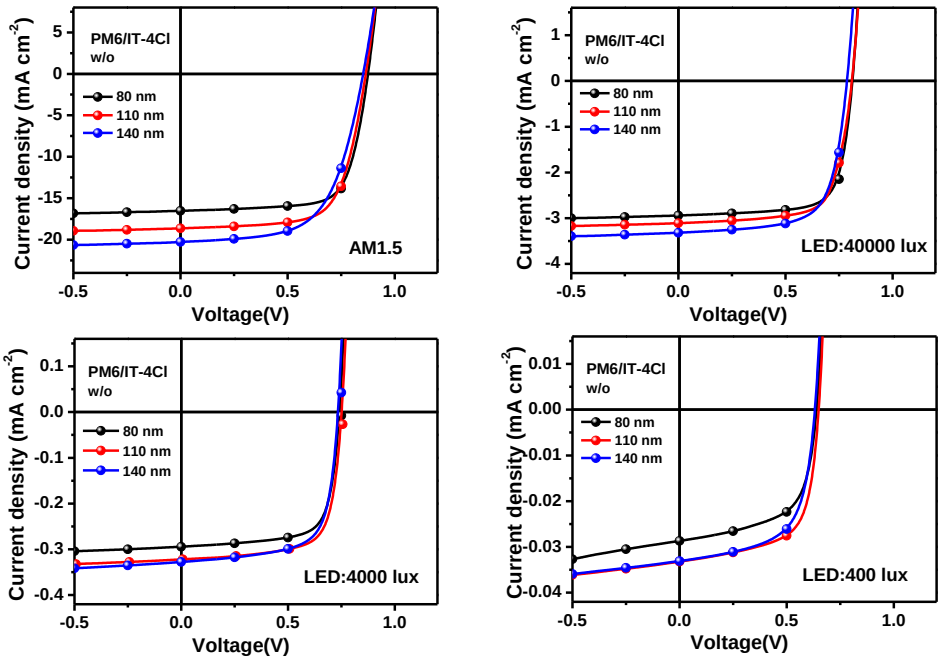

Figure S7. *J-V* curves of the OPV devices based on the active layers with different thicknesses processed without additives under different illumination condition.

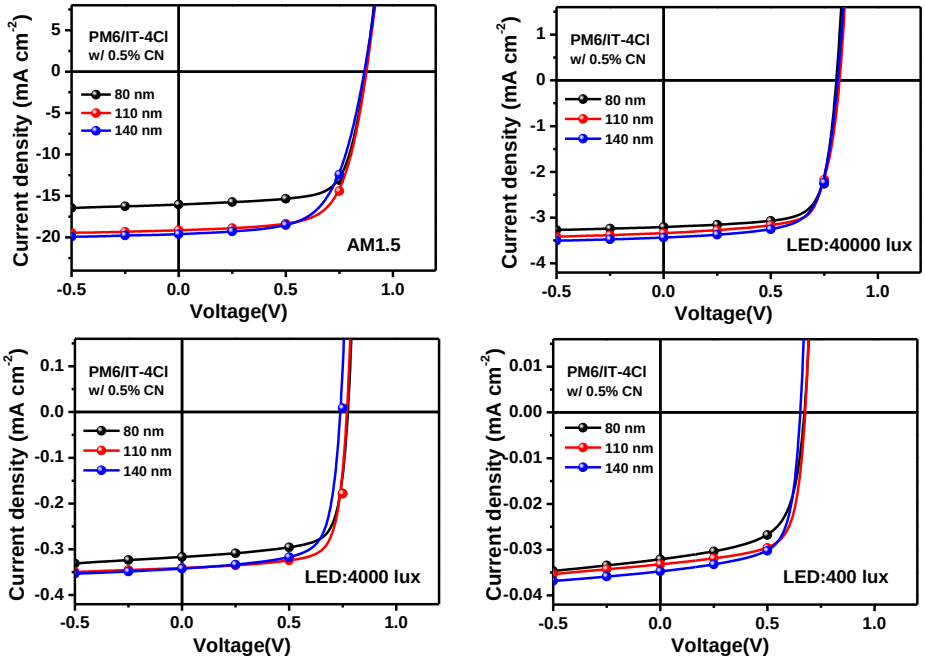

Figure S8. *J-V* curves of the OPV devices based on the active layers with different thicknesses processed with CN (0.5 vol%) under different illumination condition.

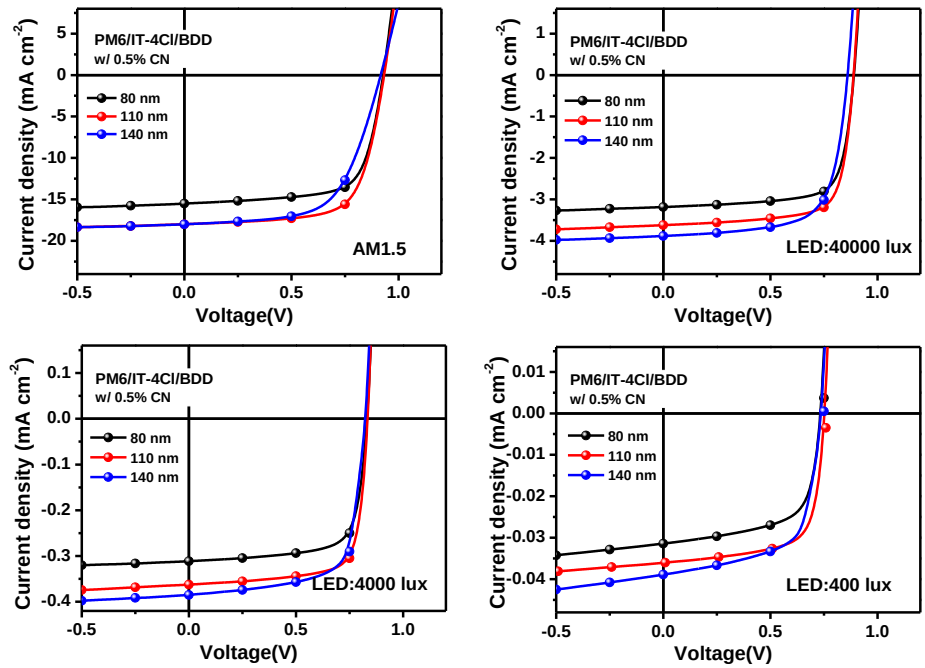

Figure S9. *J-V* curves of the OPV devices based on the active layers with different thicknesses processed with CN (0.5 vol%) and with BDD content of 20% under different illumination condition.

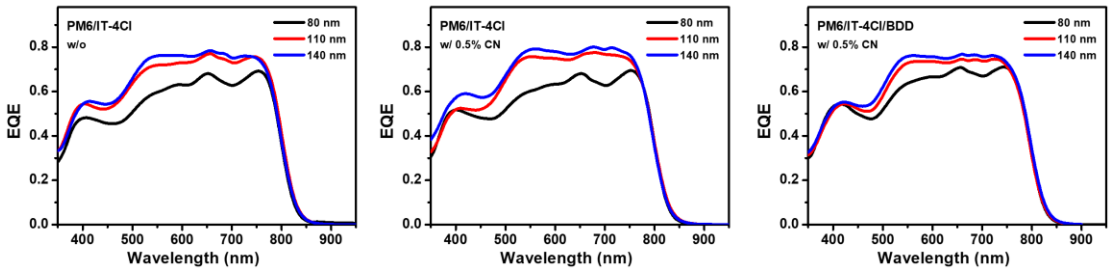

Figure S10. EQE Spectra of the OPV devices based on the active layers with different thicknesses processed without additives, with CN (0.5 vol%), and with CN (0.5 vol%) and with BDD content of 20%.

Table S1. Representative photovoltaic performance parameters for the OPV devices based on the active layers with different thicknesses, processed without additives. The measurements were conducted under different illumination conditions.

Light source	Intensity (lux)	Pin (mW cm^{-2})	Thickness (nm)	J_{SC} (mA cm^{-2})	V_{OC} (V)	FF (%)	PCE (%)
AM1.5	213000	100	80±5	15.63±0.42	0.86±0.02	71.7±0.9	9.85±0.40 (10.25)
			110±5	18.40±0.62	0.85±0.02	66.0±2.1	10.38±0.72 (11.10)
			140±5	18.46±0.53	0.84±0.02	60.9±0.8	9.72±0.36 (10.08)
LED	40000	10.8	80±5	2.90±0.04	0.81±0.02	72.9±0.9	15.91±0.36 (16.27)
			110±5	3.13±0.06	0.81±0.01	70.3±1.0	16.53±0.65 (17.18)
			140±5	3.14±0.04	0.78±0.01	68.1±1.2	15.46±0.61 (16.07)
LED	4000	1.08	80±5	0.292±0.002	0.74±0.01	71.0±0.9	14.37±0.30 (14.67)
			110±5	0.319±0.003	0.75±0.005	70.8±0.9	15.68±0.33 (16.01)
			140±5	0.327±0.004	0.73±0.01	68.4±1.0	15.07±0.50 (15.57)
LED	400	0.108	80±5	0.0285±0.0002	0.63±0.01	59.0±1.0	9.97±0.23 (10.20)
			110±5	0.0330±0.0002	0.66±0.01	64.1±0.9	12.95±0.39 (13.14)
			140±5	0.0329±0.0002	0.63±0.01	61.1±0.8	11.98±0.16 (12.14)

Table S2. Representative photovoltaic performance parameters for the OPV devices based on the active layers with different thicknesses, processed with CN (0.5 vol%). The measurements were conducted under different illumination conditions.

Light source	Intensity (lux)	Pin (mW cm^{-2})	Thickness (nm)	J_{SC} (mA cm^{-2})	V_{OC} (V)	FF (%)	PCE (%)
AM1.5	213000	100	80±5	15.66±0.52	0.86±0.02	70.2±1.2	10.38±0.72 (11.10)
			110±5	18.92±0.61	0.88±0.02	68.0±1.3	10.98±0.43 (11.41)
			140±5	18.96±0.73	0.86±0.01	62.1±1.5	10.23±0.61 (10.84)
LED	40000	10.8	80±5	2.98±0.06	0.80±0.01	72.6±1.2	16.12±0.59 (16.71)
			110±5	3.32±0.04	0.82±0.01	72.4±0.9	18.34±0.24 (18.59)
			140±5	3.38±0.07	0.80±0.01	69.6±0.8	17.68±0.43 (18.11)
LED	4000	1.08	80±5	0.315±0.004	0.76±0.01	72.4±1.0	16.36±0.42 (16.78)
			110±5	0.340±0.001	0.77±0.005	74.3±1.2	18.06±0.30 (18.36)
			140±5	0.341±0.003	0.75±0.01	70.2±0.9	16.68±0.48 (17.06)
LED	400	0.108	80±5	0.0320±0.0002	0.67±0.01	62.7±1.0	12.62±0.21 (12.83)
			110±5	0.0331±0.0001	0.68±0.01	68.7±0.8	14.27±0.21 (14.48)
			140±5	0.0345±0.0003	0.67±0.02	68.0±0.8	13.91±0.40 (14.31)

Table S3. Representative photovoltaic performance parameters for the OPV devices based on the active layers with different thicknesses, processed with CN (0.5 vol%) and with BDD content of 20%. The measurements were conducted under different illumination conditions.

Light source	Intensity (lux)	Pin (mW cm^{-2})	Thickness (nm)	J_{SC} (mA cm^{-2})	V_{OC} (V)	FF (%)	PCE (%)
AM1.5	213000	100	80±5	15.08±0.38	0.92±0.01	69.8±0.9	10.58±0.28 (10.86)
			110±5	17.86±0.43	0.93±0.02	69.1±0.8	11.42±0.36 (11.78)
			140±5	17.66±0.43	0.92±0.02	61.1±0.9	9.90±0.36 (10.26)
LED	40000	10.8	80±5	3.14±0.06	0.86±0.02	72.6±1.3	18.86±0.50 (19.36)
			110±5	3.60±0.03	0.88±0.01	73.0±1.2	21.65±0.43 (22.08)
			140±5	3.86±0.03	0.86±0.01	68.6±0.9	21.25±0.41 (21.66)
LED	4000	1.08	80±5	0.292±0.002	0.82±0.02	72.1±0.9	16.26±0.43 (16.69)
			110±5	0.363±0.003	0.83±0.02	74.2±0.9	21.03±0.37 (21.40)
			140±5	0.380±0.004	0.81±0.02	69.9±0.8	20.15±0.48 (20.63)
LED	400	0.108	80±5	0.0287±0.0002	0.73±0.01	63.7±0.8	12.48±0.21 (12.69)
			110±5	0.0356±0.0002	0.75±0.01	69.0±0.9	17.18±0.22 (17.40)
			140±5	0.0365±0.0003	0.74±0.01	62.6±0.8	15.89±0.27 (16.16)