Electronic Supplemental Information for

High-Efficiency ITO-Free Polymer Solar Cells Using Highly Conductive PEDOT:PSS/Surfactant Bilayer Transparent Anodes

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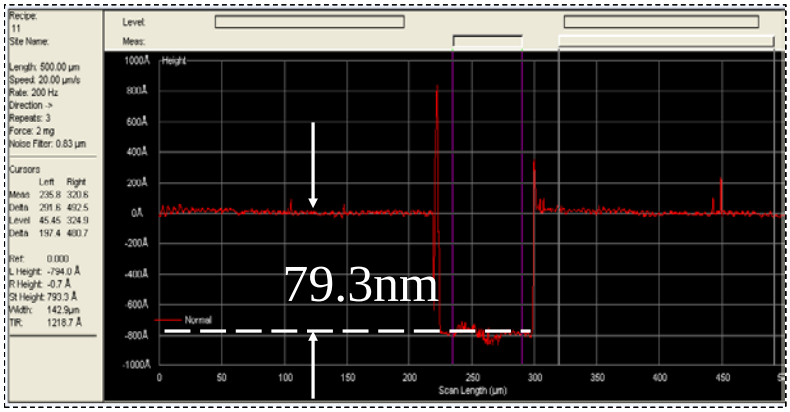
Contents

S1. Electrical properties of PEDOT:PSS/GMS bilayer films

Table S1. Electrical properties of PEDOT:PSS/GMS bilayer films prepared by spin-coating GMS solution in methanol at different spin-coating speeds.

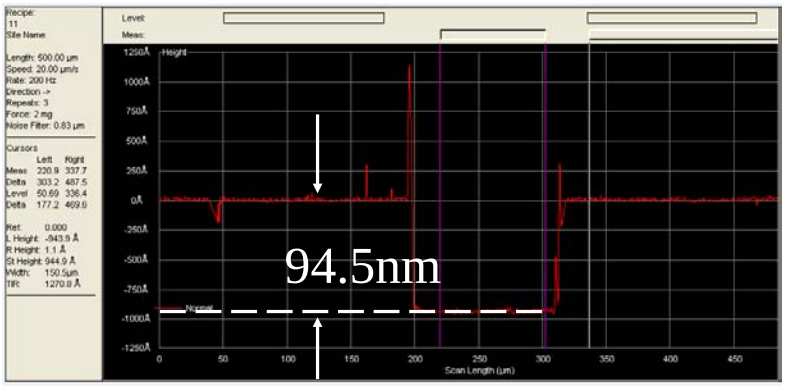
Electrode	Spin-coating speed of GMS (rpm)	Sheet resistance (Ω sq^{-1})	Conductivity (S cm^{-1})
Clevios P	-	2.46×10^{5}	0.41
Clevios P/GMS	800	275	364
	1000	285	351
	2500	352	284
	4000	606	165
Clevios PH 1000	-	9.15×10^{4}	1.09
Clevios PH 1000/GMS	800	100	997
	1000	98	1019
	2500	143	701
	4000	168	594

S2. Determination of the thickness of the GMS layers by surface profilometer



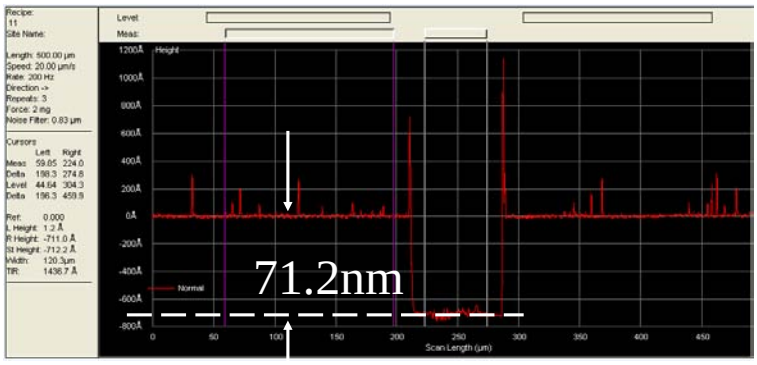
Clevios P spin-coated at 1000 rpm

(thickness of Clevios P=79.3 nm)



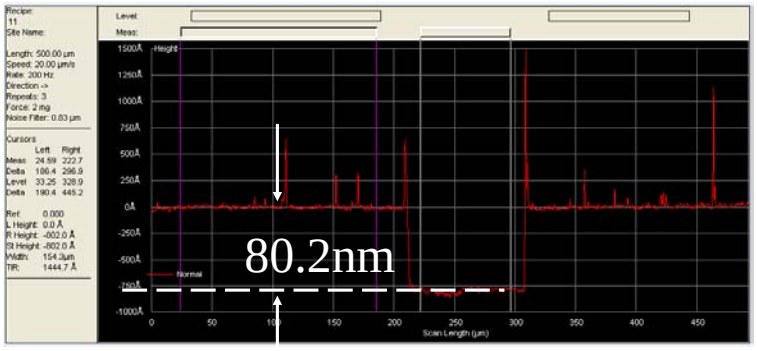
Clevios P/GMS

(thickness of Clevios P /GMS 800 =94.5 nm)



Clevios PH 1000 spin-coated at 1000 rpm

(thickness of Clevios PH 1000=71.2 nm)



Clevios PH 1000/GMS

(thickness of Clevios PH 1000/GMS=80.2 nm)

Figure S1. Surface profiles of PEDOT:PSS and PEDOT:PSS/GMS bilayer films. The thicknesses of GMS on PEDOT:PSS films are directly calculated by subtracting the sum of the thicknesses of PEDOT:PSS/GMS bilayers with that of the PEDOT:PSS single-layer.

S3. Determination of the PEDOT-to-PSS ratios by S(2p) XPS results and C(1s) XPS spectra

Table S2. The PEDOT-to-PSS ratios of PEDOT:PSS and PEDOT:PSS/GMS bilayer films determined from S(2p) XPS spectra.

film	PEDOT-to-PSS ratio
Clevios PH 1000	0.41
Clevios PH 1000/GMS	0.42
Clevios P	0.41
Clevios P/GMS	0.43

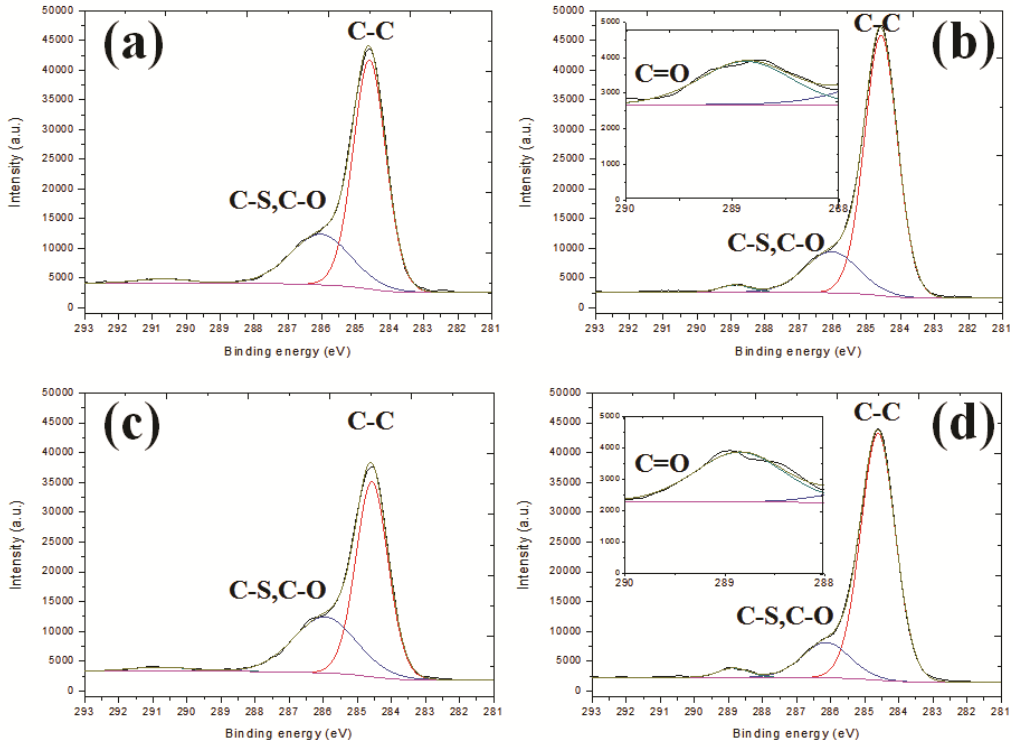


Figure S2. The C(1s) XPS spectra of Clevios P (a), Clevios P/GMS (b) , Clevios PH 1000 (c) and Clevios PH 1000/GMS (d) films.

S4. SEM images of PEDOT:PSS and PEDOT:PSS/GMS bilayer films

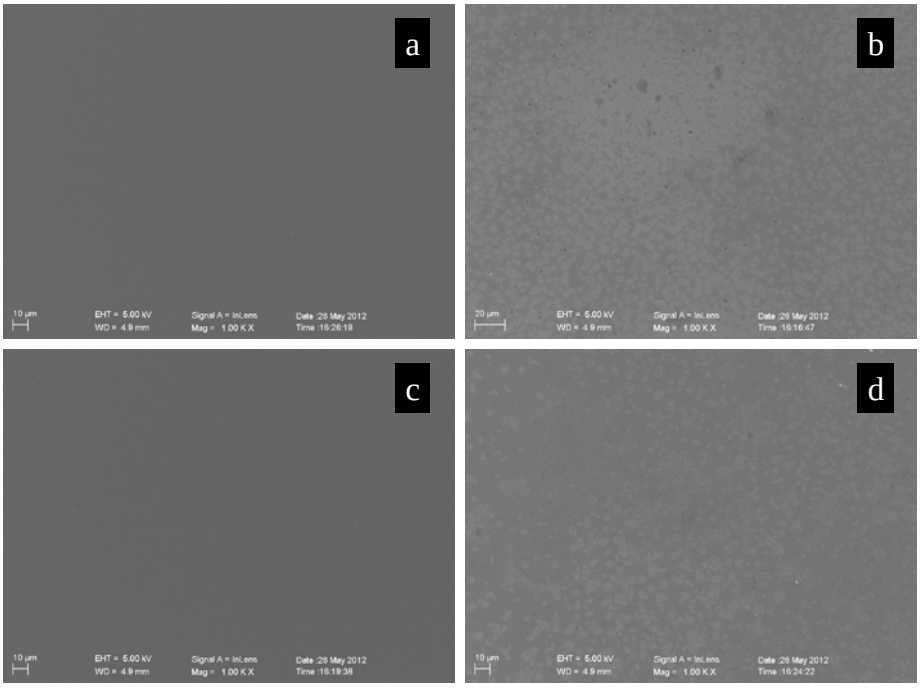


Figure S3. SEM images of Clevios P (a), Clevios P/GMS (b), Clevios PH 1000 (c) and Clevios PH 1000/GMS (d) films.

S5. Performance of different BHJ-PSC devices using PEDOT:PSS/GMS bilayer films as transparent anodes

Table S3. Device performances of P3HT:$PC_{61}BM$ BHJ-PSC devices with PEDOT:PSS/GMS bilayer films or ITO as the transparent anodes. For each condition, 10 devices were measured in order to obtain an average value.

Electrode	Active layer	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
Glass/Clevios PH 1000	P3HT:$PC_{61}BM$	0.62±0.003	0.69±0.17	25±1	0.11±0.05
ITO/Clevios PH 1000	P3HT:$PC_{61}BM$	0.61±0.001	11.42±0.51	54±2	3.75±0.09
Glass/Clevios PH 1000/GMS	P3HT:$PC_{61}BM$	0.61±0.005	12.16±0.46	53±1	3.90±0.24
ITO/Clevios P	P3HT:$PC_{61}BM$	0.61±0.006	11.27±0.24	50±1	3.40±0.15
Glass/Clevios P/GMS	P3HT:$PC_{61}BM$	0.61±0.003	11.49±0.35	47±2	3.25±0.20
Glass/Clevios P	P3HT:$PC_{61}BM$	0.61±0.005	0.46±0.038	25±1	0.07±0.02

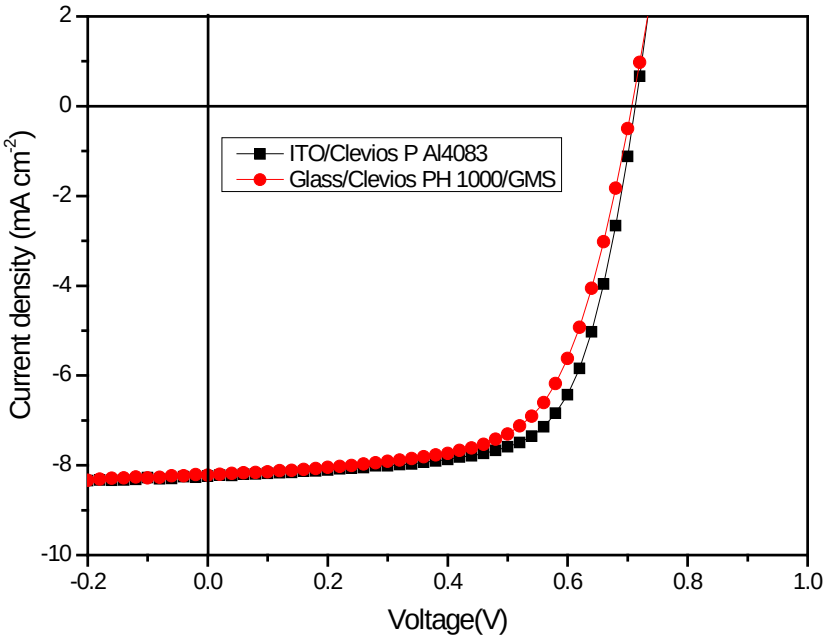


Figure S4. J–V curves of PTB7:$PC_{71}BM$ BHJ-PSC devices with PEDOT:PSS/GMS bilayer films or ITO as the transparent anodes. The measurements were carried out under AM 1.5G illumination at an irradiation intensity of 51±2 $mW{\cdot}cm^{-2}$.

Table S4. Device performances of PCDTBT:$PC_{71}BM$ and PTB7:$PC_{71}BM$ BHJ-PSC devices with PEDOT:PSS/GMS bilayer films or ITO as the transparent anodes. For each condition, 10 devices were measured in order to obtain an average value.

Electrode	Active layer	V_{oc} (V)	J_{sc} (mA cm^{-2})	FF (%)	PCE (%)
ITO/Clevios P	PCDTBT:$PC_{71}BM$	0.88±0.009	5.91±0.19	60.7±1.1	6.16±0.37
ITO/Clevios PH 1000	PCDTBT:$PC_{71}BM$	0.88±0.006	6.12±0.16	60.1±1.0	6.33±0.18
Glass/Clevios PH 1000/GMS	PCDTBT:$PC_{71}BM$	0.84±0.005	6.09±0.27	59.0±1.3	5.90±0.34
Glass/Clevios P/GMS	PCDTBT:$PC_{71}BM$	0.80±0.008	5.57±0.15	58.0±0.9	5.04±0.23
ITO/Clevios P Al4083	PTB7:$PC_{71}BM$	0.72±0.005	8.24±0.32	67.4±1.0	7.77±0.31
Glass/Clevios PH 1000/GMS	PTB7: $PC_{71}BM$	0.70±0.004	8.22±0.31	64.8±1.5	7.06±0.36