Supporting Information

Neutral-Color Semitransparent Organic Solar Cells with All-Graphene Electrodes

*Zhike Liu, Peng You, Shenhua Liu and Feng Yan**

Department of Applied Physics and Materials Research Centre
The Hong Kong Polytechnic University, Hong Kong, China
E-mail: apafyan@polyu.edu.hk

1. Effect of plasma treatment on graphene films

For the surface modification of graphene with thin films, the wettability of corresponding solution on the graphene substrate is critical to the quality of the film, which was investigated by characterizing the contact angle of the solution on graphene. The poly-(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) aqueous solution often shows a large contact angle (~86.3^o) on a pristine graphene film due to its high surface tension (~72.8 mNm^{-1}),[22] indicating a hydrophobic property of graphene (Figure S1). Therefore, the PEDOT:PSS solution is readily dewetted and can't form a uniform film on a hydrophobic graphene surface (Figure S2a). In order to prepare high-quality PEDOT:PSS film on graphene, the wettability of graphene was improved by plasma treatment for a short time (10s).[23] The contact angle of PEDOT:PSS on plasma-treated graphene was decreased to 40.3^o (Figure S1). In order to further improve its wettability, Zonyl FS-300 was added into PEDOT:PSS solution for modification, which is an effective additive surfactant in capable of providing a remarkably low surface tension (~23 mNm^{-1}) in PEDOT:PSS aqueous solution to improve its wettability.[43] The contact angles of the surfactant introduced PEDOT:PSS solution on pristine graphene and plasma-treated graphene are 37.1^o and 16.6^o, respectively (Figure S1), and a uniform coating of PEDOT:PSS film was obtained on plasma treated graphene (Figure S2b).

The plasma treatment of graphene is better not only for spin-coating uniform PEDOT:PSS as hole transport layer but also for spin-coating high-quality ZnO sol-gel or ZnO-NPs film as electron transport layer. ZnO sol-gel (zinc acetate dehydrate in 2-methoxyethanol) is also readily dewetted and can’t form a uniform film on pristine graphene (Figure S2c), whereas a uniform coating without any obvious signs of dewetting was created on plasma treated graphene (Figure S2d). ZnO-NPs in chloroform is relatively wettable on pristine graphene for low surface tension of chloroform (27.5 mNm^{-1}).[22] However, the ZnO-NPs film coated on the pristine graphene is inhomogeneous (Figure S2e), which will lead to large leakages in OPVs, while a uniform ZnO-NPs film can be obtained on graphene after the plasma treatment (Figure S2f).

The influence of plasma treatment on the conductivity of 2L-G transferred on glass and PMMA substrates is also characterized (Figure S4). The R_s of 2L-G on the two substrates exhibits a constant period followed by a rapid increase. The degradation rate of conductivity for graphene on PMMA is lower than that of graphene on glass due to the good contact between the graphene and PMMA transfer layer when the treatment time is longer than 20s. After plasma treatment for 10s, No decrease of conductivity and detectable defect-related Raman D band (at ~1350 cm^{-1})[28] of graphene can be observed (Figure 1c). So 10s is the safety time to treat graphene for improving its wettability while keeping its conductivity. After plasma treatment, surfactant modified PEDOT:PSS, ZnO sol-gel and ZnO-NPs can be uniformly spin-coated on graphene films (Figure S2).

2. Figures:

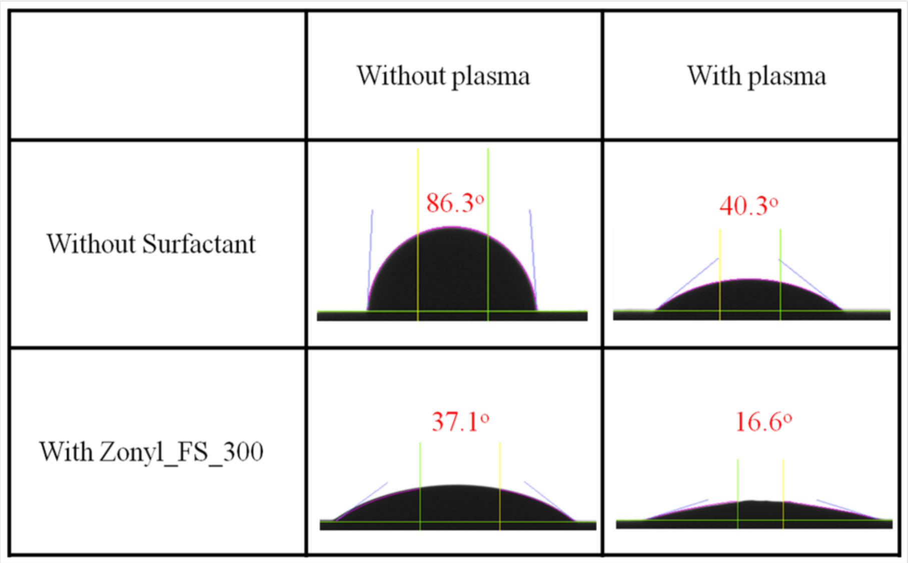

Figure S1. The contact angles of PEDOT:PSS aqueous solution without or with surfactant (Zonyl FS-300) on 2-layer CVD graphene films without or with plasma treatment on the surface for 10 seconds.

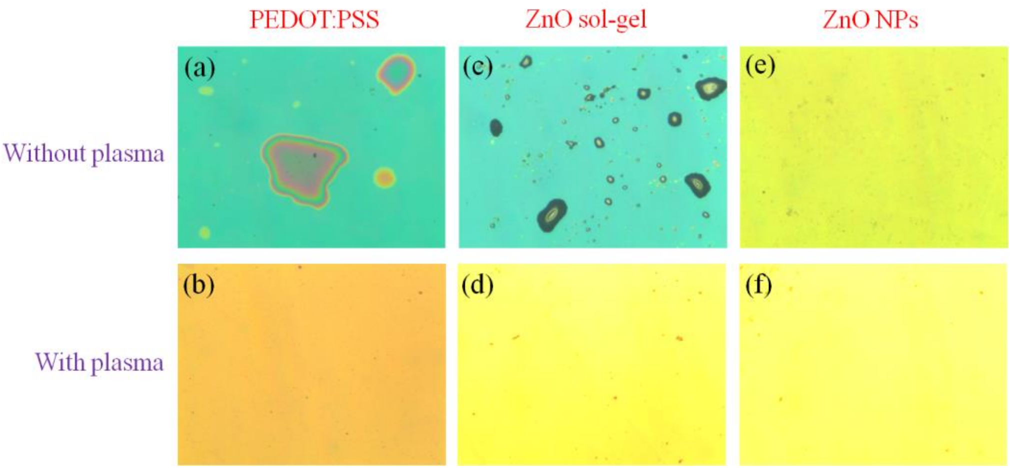

Figure S2. The photographs of (a), (b) PEDOT:PSS, (c), (d) ZnO sol-gel, and (e), (f) ZnO-NPs coated on graphene films without or with plasma treatment.

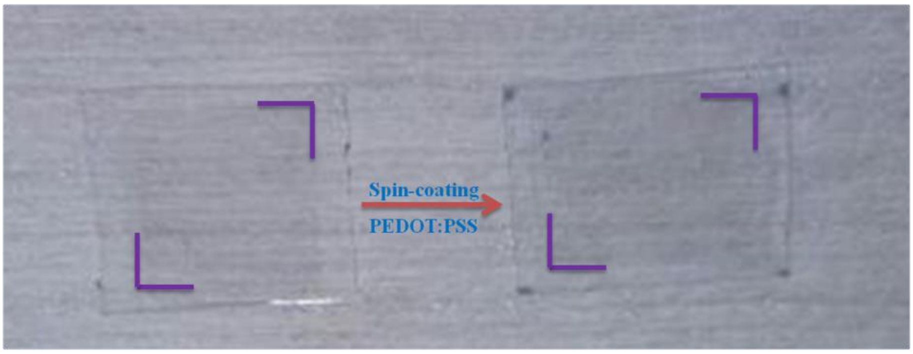

Figure S3. Graphene/PMMA/PDMS anode before and after spin-coating PEDOT:PSS film.

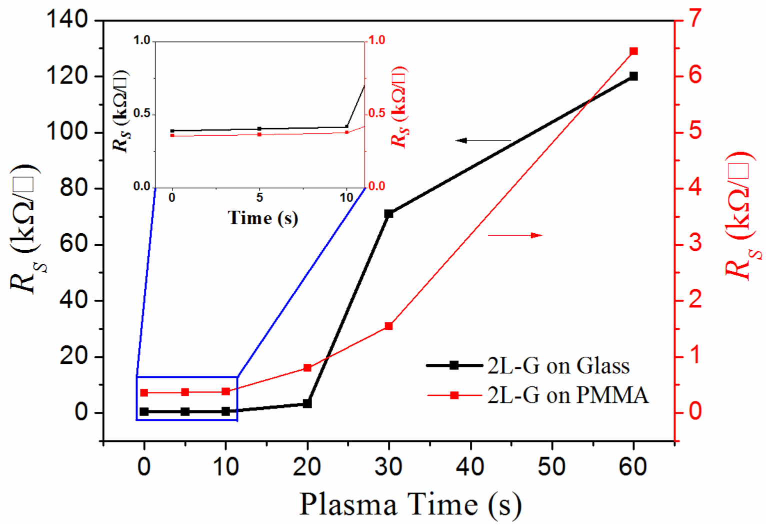

Figure S4. R_s of graphene on glass or PMMA after plasma treatment with different periods, inset: the enlarged area where the plasma time less 12s.

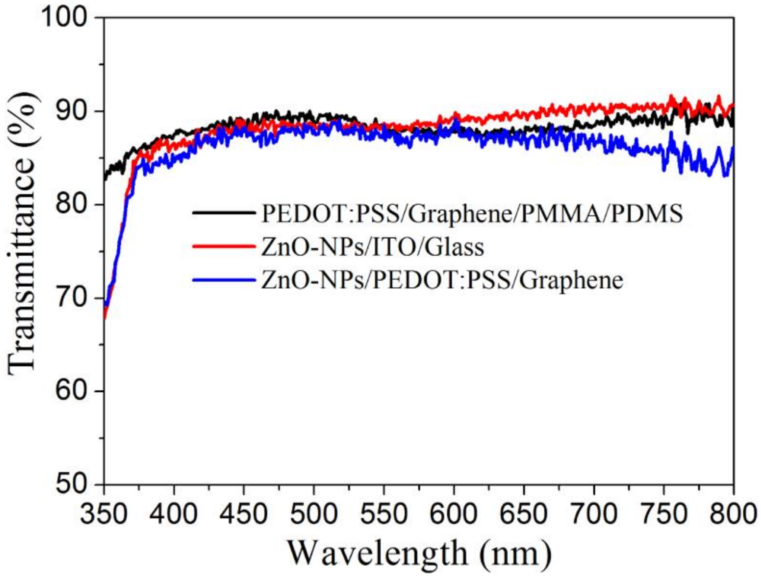

Figure S5. The transmittance spectra of different transparent electrodes, including graphene anode, graphene cathode and ZnO-NPs modified ITO electrodes.

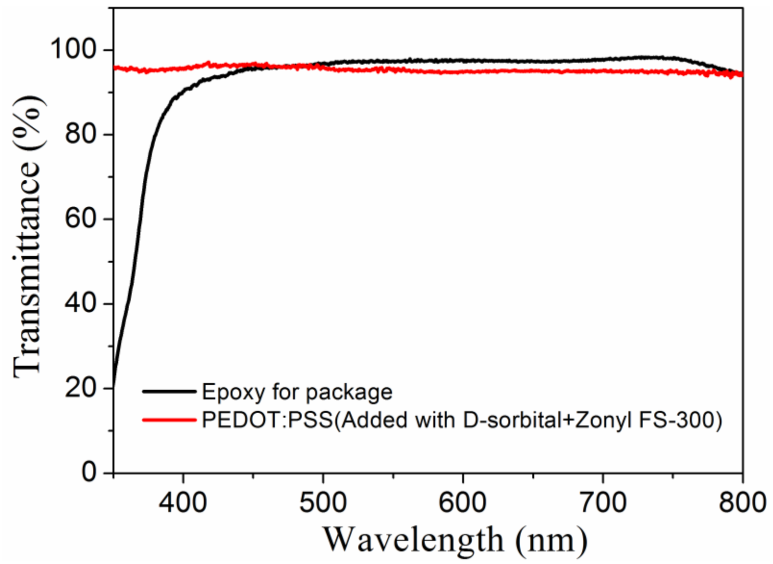

Figure S6. The transmittance spectra of the epoxy resin film and the PEDOT:PSS film added with D-Sorbital (5%) and Zonyl FS-300 (0.1%).

Table S1. PCEs of carbon-based soalr cells, including devices with all carbon active layers, devices with all carbon electrodes and all carbon solar cells

Device type	Device structure (*)	PCE (%)	Ref.
(I)All carbon active layers	ITO/PEDOT:PSS/TFB/SWCNT:$PC_{71}BM$:rGO/Al	1.30	[32]
	ITO/GO:SWCNT/SWCNT:C_{60}:GNR/$PC_{60}BM$/ZnO/Ca/Al	1.14	[33]
	ITO/PEDOT:PSS/rGO:SWCNT/C_{60}/Al	0.85	[34]
	ITO/CNT/C_{60}/BCP/Ag	0.6	[35]
	ITO/PEDOT/SWCNT/C_{60}/BCP/Ag	0.46	[36]
	ITO/PEDOT:PSS/SWCNT:C_{60}:rGO/C_{60}/Al	0.21	[37]
(II)All carbon electrodes	Carbon fiber/TiO_2/dye/electrolyte/Ink carbon	1.90	[38]
	CNT/TiO_X/TiO_2/dye/electrolyte/CNT	1.37	[39]
	Gra/PEDOT:PSS/ZnO/PTB7:$PC_{71}BM$/PEDOT:PSS/Gra	3.40	**This work**
(III)All carbon solar cells	rGO/SWCNT/C_{60}/SWCNT	0.0057	[36]

(*) TFB: poly[(9,9-dioctylfluorenyl-2,7-diyl)-co-(4,40-(N-(4-s butylphenyl))diphenylamine)]

SWCNT: single-walled carbon nanotubes

BCP: bathocuproine

rGO: reduced graphene oxide

Gra: graphene