Supplementary Information

All Annealing-Free Solution-Processed Highly Flexible Organic Solar Cells

Juanyong Wan,[‡,a,b] Rongjiang Wen,[‡,a,b] Yonggao Xia,[b,*] Mingzhi Dai,[b] Huihui Huang,[a,*] Lingwei Xue,[c,*] Zhiguo Zhang,[c] Junfeng Fang,[b,d] Kwun Nam Hui[e] and Xi Fan[b,*]

[a]School of Physics and Electronics, Hunan University, Changsha 410082, P.R. China. E-mail: huangh@hnu.edu.cn.

[b]Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, Ningbo, 315201, P.R. China. E-mail: xiayg@nimte.ac.cn; fanxi@nimte.ac.cn.

[c]State Key Laboratory of Organic/Inorganic Composites, Beijing Advanced Innovation Center for Soft Matter Science and Engineering, Beijing University of Chemical Technology, Beijing 100029, P.R. China. E-mail: xlingwei@mail.buct.edu.cn.

[d]School of Physics and Electronics Science, Engineering Research Center of Nanophotonics & Advanced Instrument, Ministry of Education, East China Normal University, Shanghai 200241, P.R. China.

[e]Joint Key Laboratory of the Ministry of Education, Institute of Applied Physics and Materials Engineering, University of Macau, Avenida da Universidade, Taipa, Macau SAR 999078, P.R. China.

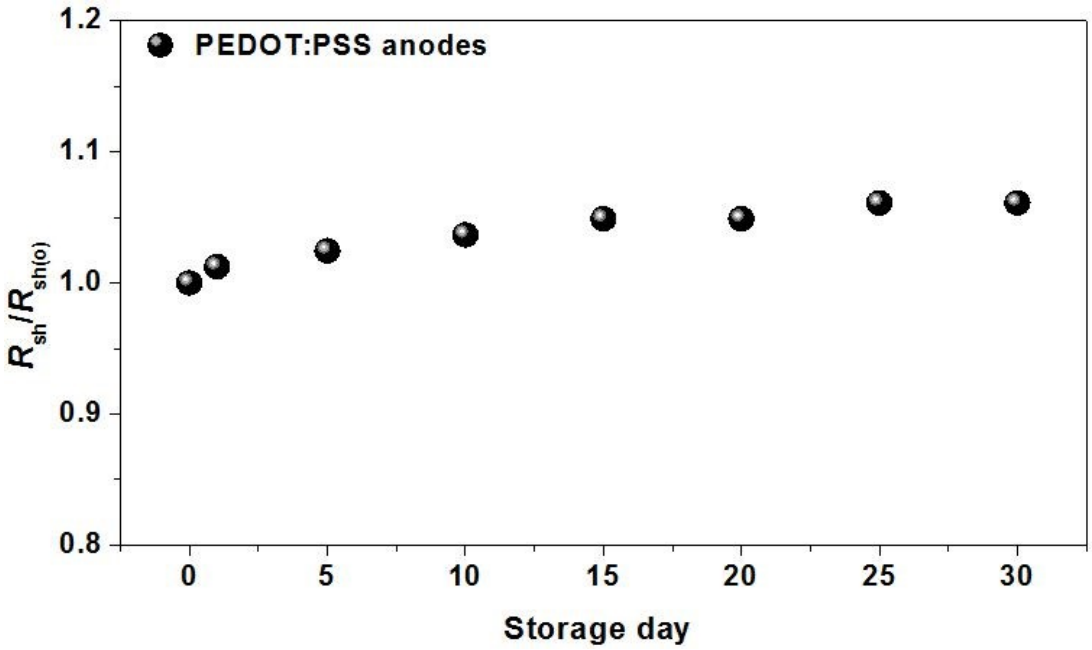


Fig. S1 $R_{sq}/R_{sq(o)}$ of the PEDOT:PSS anodes treated by CF_3SO_3H as a function of storage days in air atmosphere.

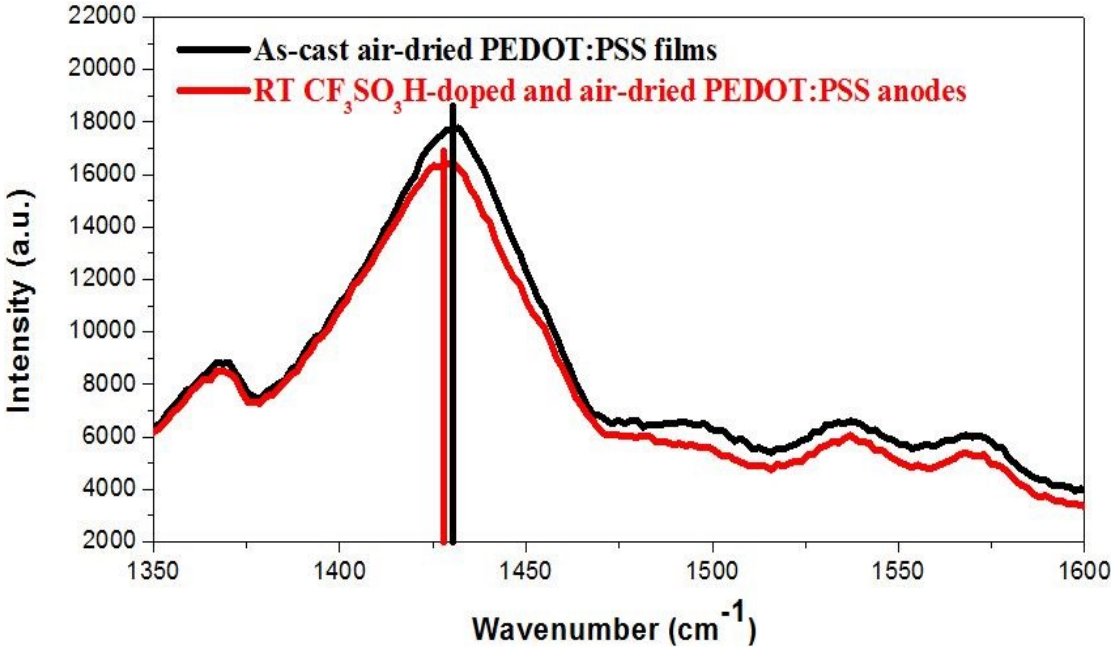


Fig. S2 Raman spectra of the as-cast air-dried PEDOT:PSS films and the room-temperature CF_3SO_3H-doped and air-dried PEDOT:PSS anodes.

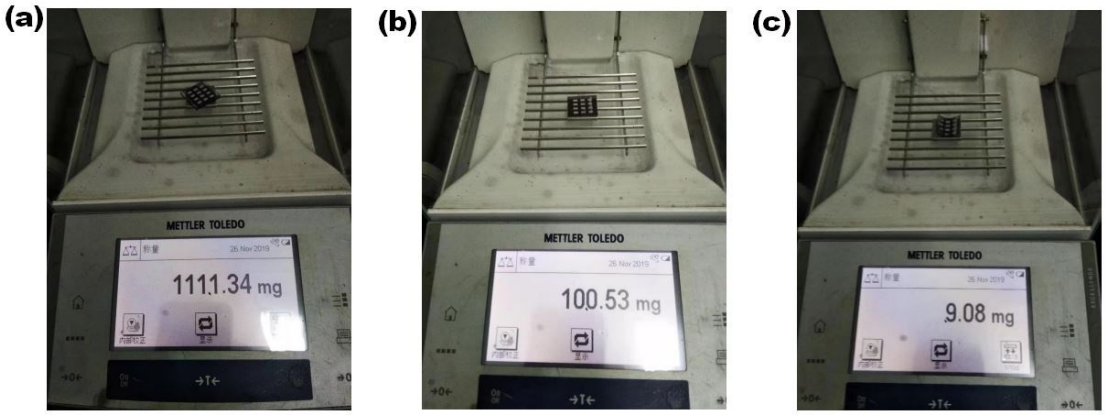


Fig. S3 Weights of the devices including the rigid ones on glass substrates (**a**), the flexible ones on 200-μm-thick PET substrates (**b**) and the ultrathin soft ones on 24-μm-thick PE substrates (**c**).

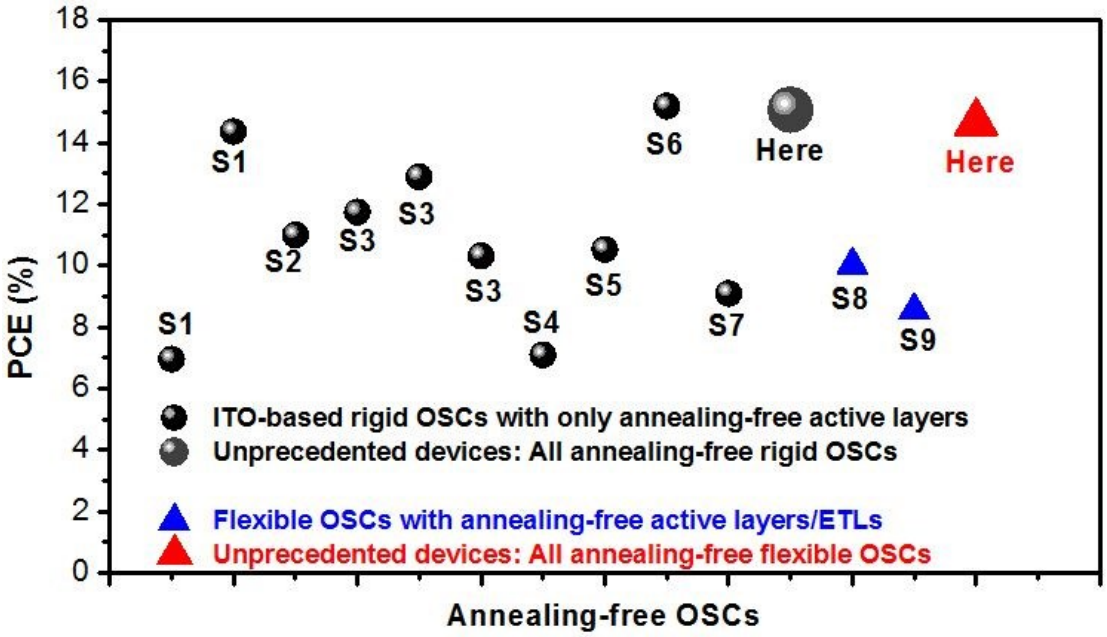
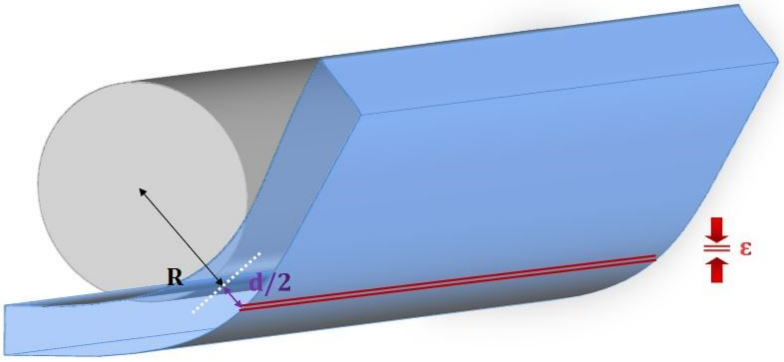


Fig. S4 Plotted values of PCEs of the rigid and flexible OSCs with annealing-free functional layers reported recently.



Sample	R	d/2	ε
PE: 24 μm, *r*: 0.25 mm	262 μm	12 μm	4.58%
PET: 200 μm, *r*: 0.25 mm	350 μm	100 μm	28.57%
PE: 24 μm, *r*: 0.50 mm	512 μm	12 μm	2.34%
PET: 200 μm, *r*: 0.50 mm	600 μm	100 μm	16.67%

Fig. S5 Schematic illustration of the cross-sectional dimensions. Here, ε is the normal strain on the vertical cross-sectional plane, R is the radius of curvature, and d is the thickness of the PE and PET substrates, respectively.

Fig. S6 Cracks of the PM6:Y6 active blends (see bottom regions) coated on the 200-µm-thick rigid PET substrates in a harsh flexing at *r* of 0.25 mm.

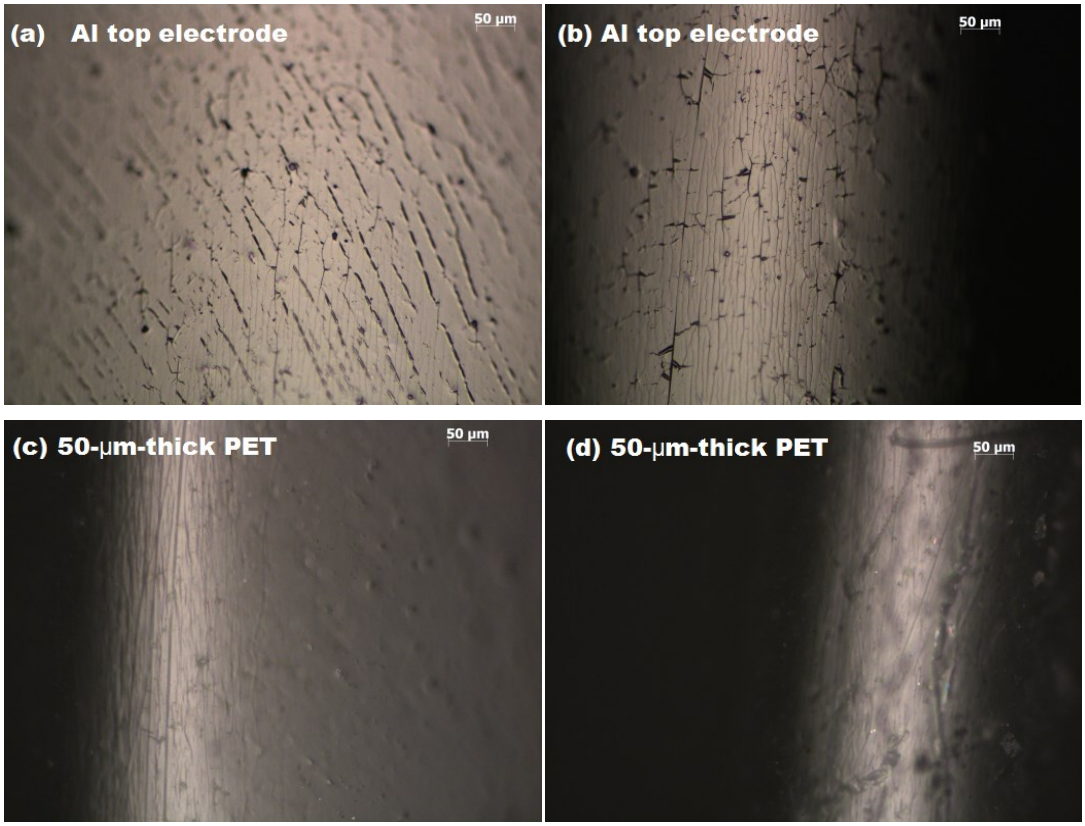

(a) Al top electrode
50 μm
(b) Al top electrode
50 μm
(c) 50-μm-thick PET
50 μm
(d) 50-μm-thick PET
50 μm

Fig. S7 Cracks of Al top electrodes on 50-μm-thick PET substrates and the 50-μm-thick PET substrates in 100 cyclic flexing tests. (**a,b**) Al top electrodes with flexing at *r* of 0.50 and 0.25 mm, respectively. (**c,d**) The PET in the flexing at *r* of 0.50 mm and 0.25 mm, respectively. Scale bar: 50 μm.

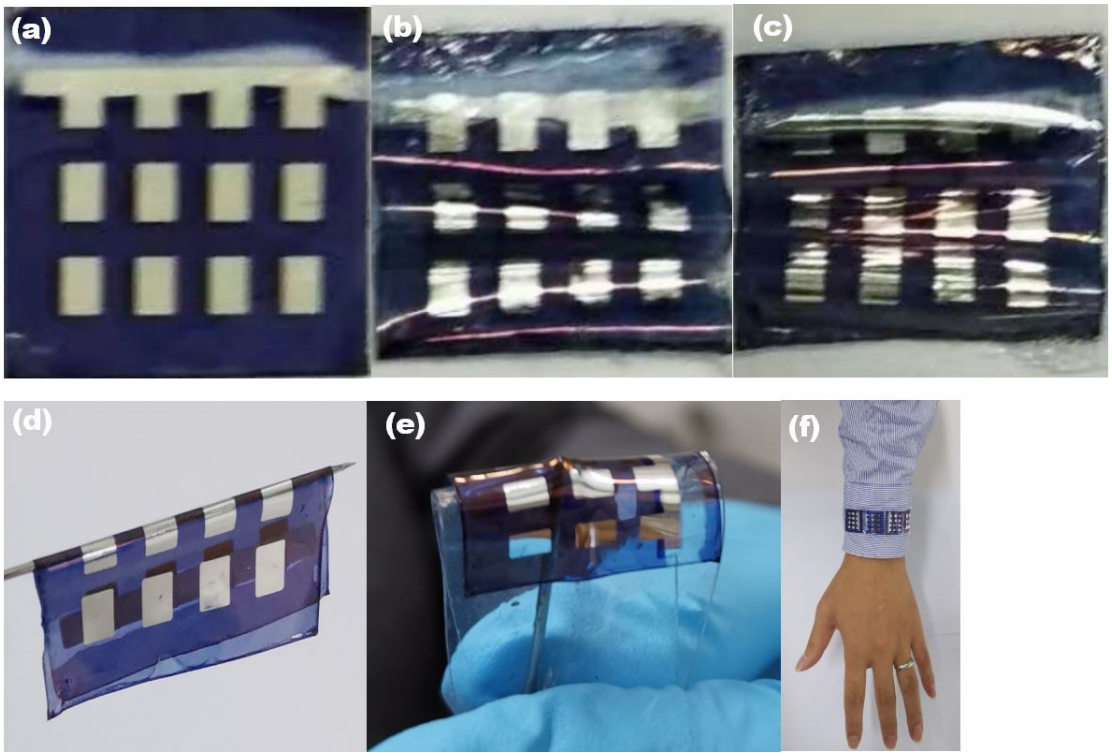


Fig. S8 (**a,b**) Pristine annealing-free solution-processed highly flexible organic solar cells. (**b,c**) Waved configuration and wrinkled configuration of the flexible devices through placing them on a pre-strained PDMS stamp (thickness: ca. 1 μm). (**d**) Due to the soft property, the device foils were hung on the metallic needles (radius: 0.25 mm). (**e**) The highly flexible device attached to PDMS elastomeric supports under three-dimensional deformation by pressure from a 0.2 mm-diameter metallic object. (**f**) The highly flexible devices attached to the sleeves of the shirt for a potential wearable integration in the future.

Tab. S1 Summaries of photovoltaic characteristics of the annealing-free OSCs

Device	TE	AL	Annealing-free components	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%) (Ave.)	Ref.
Rigid OSCs	ITO/ Glass	PM6:C4	AL,TME	0.689	15.74	67.01	7.28 (6.96)	[S1]
		PM6:C6	AL,TME	0.84	23.82	72.68	14.54 (14.37)	[S1]
		PBDS-T: ITIC	AL,TME	0.985	18.68	60.01	11.06	[S2]
		PBDB-T:IDTCN-C	AL,TME	0.84	20.33	69.6	11.92 (11.75)	[S3]
		PBDB-T:IDTCN-O	AL,TME	0.91	19.96	73.2	13.28 (12.91)	
		PBDB-T:IDTCN-S	AL,TME	0.85	19.04	65.7	10.60 (10.34)	
		DTBDT-Rho: $PC_{71}BM$	AL,TME	0.81	17.2	51.0	7.10	[S4]
		PTB7-TH: $PC_{71}BM$	AL,TME	0.080	18.74	71.27	10.53	[S5]

		PM6:Y6	AL,TME	0.86	24.3	73.2	15.2	[S6]
		PffBT-2TPF4-9/1: PCBM	AL,TME	0.769	17.18	70.8	9.4 (9.1)	[S7]
	PEDOT:PSS/ Glass	PM6:Y6	TE, HTL, AL, ETL,TME	0.840	24.55	73.1	15.08	Here
Flexible OSCs	**Ag NW/ PET**	**PTB7-Th: $PC_{71}BM$**	**ETL, AL,TME**	**0.79**	**18.51**	**68.68**	**10.04**	**[S8]**
	Ag NW/ PET	**PTB7-Th: $PC_{71}BM$**	**AL,TME**	**0.764**	**17.4**	**64.2**	**8.75 (8.56)**	**[S9]**
	PEDOT:PSS/ PE	**PM6:Y6**	**TE, HTL, AL, ETL,TME**	**0.84**	**23.92**	**0.729**	**14.66**	**Here**

TE: transparent electrode; **AL**: active layer; **HTL**: hole-transport layer; **ETL**: electron-transport layer; **TME**: top metal electrode.

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