Flexible Polymer Solar Cells with Power Conversion Efficiency of 8.7%

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Polymer solar cells (PSCs) are being extensively investigated due to their perceived advantages as lightweight and low-cost energy source.[1,2] This new class of devices relies on the physical blend of conjugated semiconducting polymer (usually as electron donor) and fullerene derivative (as electron acceptor) (typically a n-type fullerene derivative), and can be processed from solution-based technology, such as ink-jet printing, roll-to-roll manufacturing, micro-contact printing, solution spraying. Owing to the rapid advancements of materials,[3-9] device structures[10-16] and processing techniques,[17,18] the power conversion efficiency of the state-of-art PSCs has increased dramatically in the past a few years, reaching 8% since 2011[10-12] and 9-10% more recently.[13,15,16,19-21]

Nowadays, most of the reported high-efficiency PSCs were fabricated on rigid glass substrate thus does not make full use of the processing advantages of organic semiconducting materials and as a result limit their potential applications. In order to fully exploit the potential of PSCs, it is desirable to develop PSCs on flexible plastic or metal foil substrates,[22] which are lightweight, mechanically flexible and conformable, and compatible with high-throughput roll-to-roll manufacturing process and may potentially reduce the cost of production and installation. The most straightforward strategy toward the realization of flexible PSCs is based on a conventional device structure, in which poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS)[23-26] with high conductivity or metal wires[27,28] were incorporated atop flexible substrate was usually used as anode for collecting photo-generated hole. In particular, Na *et al.* demonstrated that flexible PSCs with PCE of 2.8% from polyethylene terephthalate (PET) substrate that pre-coated with high conductivity.[29] Alternatively, flexible PSCs can be fabricated from an inverted

structure, in which n-type metal oxides (ZnO, TiO$_X$ etc) nanoparticles [30, 31] are usually used as ITO surface modification layer via solution process or sol–gel process to facilitate electron transport and extraction. Recently, Wang *et al*. demonstrated flexible inverted PSCs on poly(ethylene naphthalate) (PEN) substrate with a power conversion efficiency of 4.18% based on ZnO layer, which was be prepared by atomic layer deposition (ALD) at low temperatures (i.e, 40 °C and 80 °C).[32] More recently, a high performance flexible PSCs with a PCE of 6% on PEN substrate was developed, which fully takes advantage of low temperatures (below 150 °C) processing.[33]

Nevertheless, post treatments such as annealing of charge transporting metal oxide (over 200 °C) or photoactive layer (over 110 °C) are usually required to enhance device performance in many emergent flexible PSCs, making plastic substrate unsuitable for highly efficient PSCs. Therefore, the efficiency of flexible PSCs was inferior to that of the corresponding device on rigid ITO/glass substrate and PCE over 8% have not been reported in flexible PSCs. Before this kind of device can find practical applications as portable power sources, significant progress must be made in improving power conversion efficiency. Thus, further efforts should be devoted to the development of highly efficient PSCs on flexible substrates by using low temperature technology without big loss in efficiency.

We previously reported a technique to fabricate inverted type PSCs by incorporating an alcohol/water-soluble conjugated polymer, poly [(9,9-bis(3'-(N,N-dimethylamino) propyl)-2,7-fluorene)-*alt*-2,7-(9,9–dioctylfluorene)] (PFN) as surface modification layer atop of glass/indium tin oxide (ITO), resulting in a high-efficiency PSCs with a certified PCE of 9.214%.[13] This technique is solution-based and all of the film deposition processes are completed at room temperature, thus can offer easy-processibility over large area size. It is desirable to

extend our optimized device structure from glass substrate to a flexible plastic substrate. Usually, the efficiency of the reported flexible PSCs was usually limited by the inefficient utilization of photons and the non-ideal ohmic contact between the flexible substrate and the active layer. Here we report the fabrication of a low temperature processed high-efficiency flexible polymer solar cells on a PET substrate pre-coated with ITO via this recently developed inverted PSCs device structure. The PCE of the resulting flexible PSCs based on a blend of low bandgap semiconducting polymer thieno[3,4-b]thiophene/benzodithiophene (PTB7) [34] and [6,6]-phenyl C_{71}-butyric acid methyl ester (PC$_{71}$BM) is 8.7%, which is, to our best knowledge, the most efficient flexible PSCs and comparable to those on ITO/glass substrate (~9.2%). To further investigate the applicability of the PET/ITO/PFN substrate for flexible PSCs, we had also utilized other two typical polymers, each having different band gap, energy levels and processing conditions in the device fabrication, as photoactive layer. The flexible devices based on these two polymers also showed encouraging device performance, indicating that the method reported here is a promising approach towards the realization of high-efficiency, flexible PSCs, that can enable fast deposition on an industrial scale at room temperature.

Flexible PSCs in this study was fabricated on a commercial available polyethylene terephthalate (PET) substrate that pre-coated with ITO (PET/ITO). The transmission spectra of the PET/ITO in the UV, visible and infrared range was measured and showed in Fig. 1. PET/ITO substrate depicted high optical transmittance (with an optical transmittance of 86% @480 nm, 83%@550nm, and an average optical transmittance of 79.9%) in wide range between 350 nm and 1100 nm, approaching that of the control glass/ITO substrate (85.6%). This is an essential prerequisite for flexible electronics applications. Nevertheless, the high transmittance

of the PET/ITO substrate is somewhat compensated by its relatively higher sheet resistance of 40 ohm/square. The root-mean-square roughness of the PET/ITO substrate before and after PFN coating is 7.04 nm and 3.67 nm over a surface of 5 μm×5 μm, respective, while the RMS roughness of the glass/ITO substrate is 1.4 nm (Figure 2). Therefore we conclude that a more smooth surface morphology is formed after PFN coating and thus can effectively avoid electrical short in the resulted devices. Apart from the surface morphology, the electrical properties of the substrate change upon the incorporation of PFN. The work function of PET/ITO evaluated by Kelvin probe was decreased from 5.1 eV to 4.5-4.7eV after PFN coating, implying that the substrate can form ohmic contact with the lowest unoccupied molecular orbital (LUMO) of $PC_{71}BM$ for efficient electron extraction.
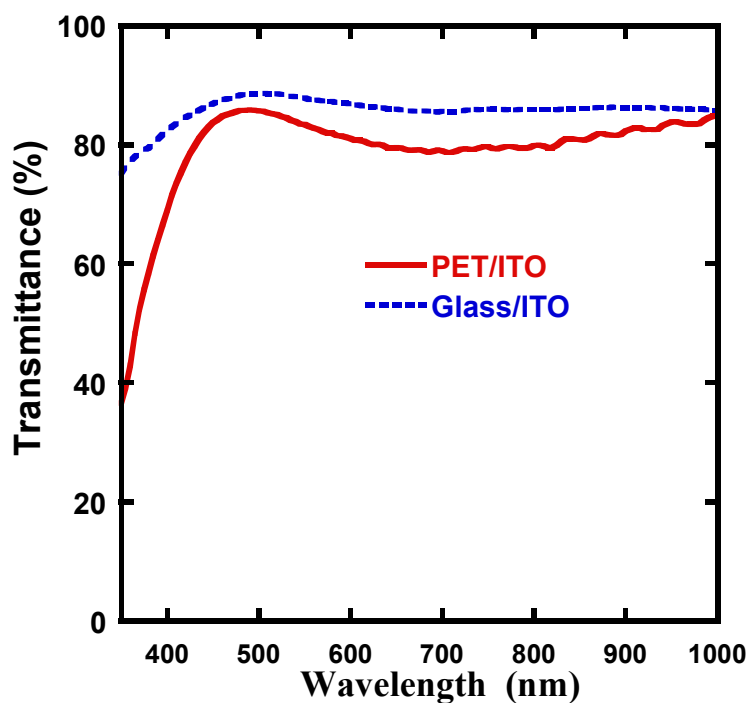

Figure 1 The optical transmittance of the PET (125 um)/ITO (100 nm) substrate (with a sheet resistance of 40 ohm/square), together with the transparency of glass/ITO (130 nm) substrate (with a sheet resistance of 15 ohm/square).
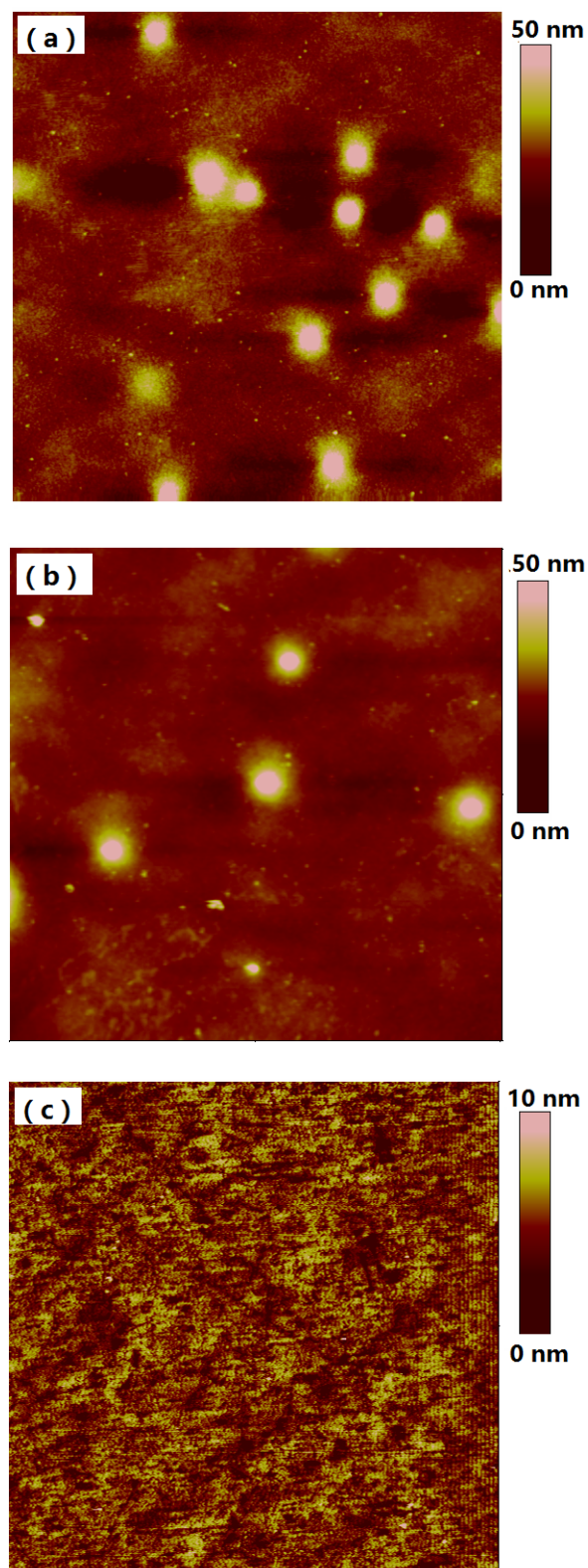
Figure 2 AFM images of PET/ITO substrate PET/ITO substrate before (a) and

after PFN coating (b), AFM images Glass/ITO substrate (c). The rms roughness of the PET/ITO susbtrate is 7.04 nm before PFN coating and 3.67 nm after PFN coating.

To demonstrate flexible, high-efficiency polymer solar cells, low bandgap semiconducting polymer thieno[3,4-b]thiophene/benzodithiophene (PTB7) was employed as electron donor material. The typical device structure of the flexible PSCs is PET/ITO/PFN/Photoactive Layer/MoO$_3$/Al (device 2). Figure 3a showed the current density versus voltage (J–V) characteristics of the best flexible device. For comparison, the best J–V characteristics of flexible device with the absence of any electron transporting layer (device 1) are also shown. The device performance parameters deduced from the J–V characteristics and that of flexible devices with ZnO film derived from sol gel as electron transporting layer (device 3) and a reference inverted PSCs on rigid glass (device 4) from previously reported literature are summarized in Table 1. The flexible device 1 from PTB7:PC$_{71}$BM showed a highest PCE of 8.60%, with an open circuit voltage (V_{OC}) of 0.74 V, a short-circuit current density (J_{SC}) of 17.4 mA cm^{-2}, and fill factor (FF) of 66.8%, when measured under 1000 W m^{-2} AM 1.5G illumination. On the other hand, the control flexible device 2 only showed a moderate efficiency of 5.60%, while the flexible PSCs with ZnO film derived from sol gel as electron transporting layer showed a PCE of 8.12% after annealing the obtained ZnO film at 120°C for 10 min (Table 1). These results clearly demonstrated that the proposed device structure is indeed suitable for PET substrate-based flexible polymer solar cells. After encapsulation, the device was sent to the National Center of Supervision & Inspection on Solar Photovoltaic Products Quality of China (CPVT) for certification. Fig. 3b shows the certified J-V curve of the device measured under AM1.5G illumination. A certified PCE of 8.709% was

obtained (Fig. 3b), which is 94% of the PCE of our best cells on glass substrate.[13] To

the best of our knowledge, the remarkable efficiency over 8% is among the highest

efficiency for flexible polymer solar cells reported to date. The device has a V_{OC} of

0.7359 V, a J_{SC} of 17.94 mA cm^{-2}, and fill factor of 65.9%. Comparison of the

parameters of the flexible device on PET/ITO substrate and the device on rigid

glass/ITO substrate indicates a difference in FF (65.9% vs. 69.99%, Table 1). On the

other side, the J_{SC} in both devices is very close despite PET/ITO substrate have a

slightly lower optical transmittance, which can be ascribed to interference effects in

the flexible devices. We expect that further optimization of the interface between the

photoactive layer will result in an improvement in the FF and the overall device

performance.[10] Thus we believe flexible polymer solar cells with efficiency above 9%

should be achievable with this device structure.

Fig. 3c compared the *J-V* characteristics of the devices fabricated in this study in

dark in semi-log axis. The device 2 showed obviously smaller leakage current in

reverse voltage and low forward voltage region (between 0 V and the turn-on voltage)

than that of the control devices (device 1 and the ZnO-based device 4), which is likely

due to a more smooth surface morphology in the PET/ITO/PFN substrate (Fig. 2).

To demonstrate the flexibility of the devices, sheet resistance measurement under

different bending condition and device performance test under different bending

cycles was performed. Figure S1 (a) shows the sheet resistance of the PET/ITO under

different bending conditions. It was found that with the bending angle change

between 0~90°or 0~ -90°, the measured sheet resistance remained nearly constant (at

around 40 ohm/square). Secondly, we investigated the device performance on convex

bending-recovery cycles at fixed angle (here is fixed at 30 °). We performed this test

using a device fabricated 150 days that have a PCE of 6.8% before bending.

Surprisingly, the device showed even better efficiency even after repetitive bending cycles up to 300 cycles (the overall efficiency, Voc, Jsc and FF as a function of bending cycles are shown in Figure S1 (b)~(e) and summarized in Table S1 for comparison). It is not very clear why enhanced performance was observed upon repetitive bending at this moment and more in-deep study will be carried out in the forthcoming research.

After simple encapsulation by epoxy, the device 2 retained 92% of its initial efficiency when stored in ambient air condition for 40 days, representing very good shelf storage stability. Device performance parameters of a typical Device 2 in longer shelf storage period up to 150 days are shown in Table S2, with corresponding *J*-V characteristics displayed in Figure S2. In short, after 120 and 150 days storage, the device retained 92%, 82 of its initial efficiency. It was observed that the PCE loss in the first four months is mainly due to the reduction in FF while in the consequent stage, losses in Jsc seemed to be dominant.
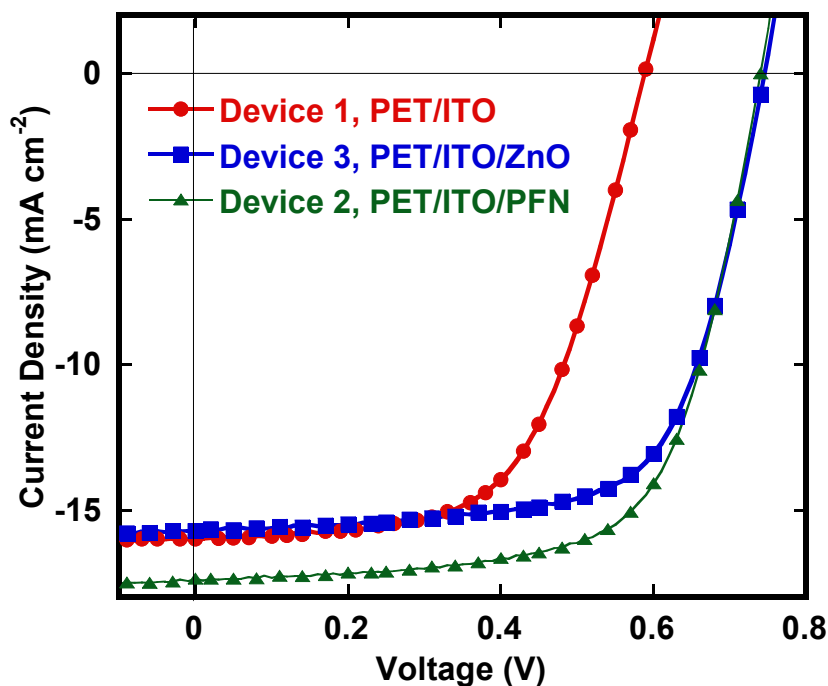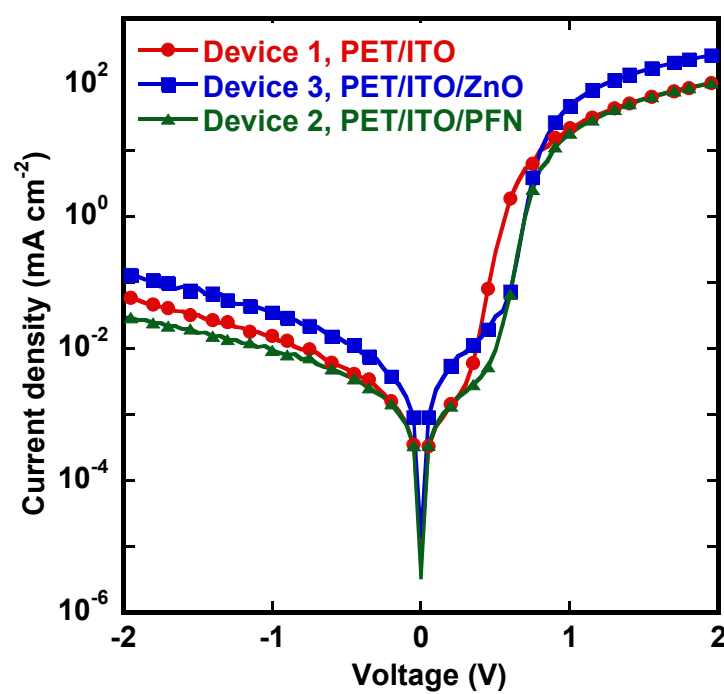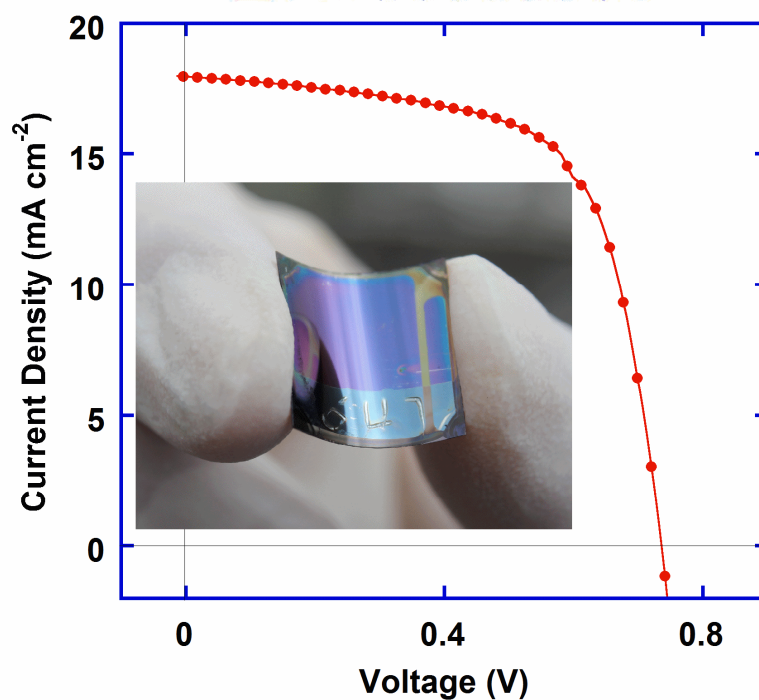
Figure 3. a) the *J–V* characteristics of the flexible PSCs with device structure of PET/ITO/PFN/PTB7:PC$_{71}$BM/MoO$_3$/Al device and the control flexible devices with device structure of PET/ITO/PTB7:PC$_{71}$BM/MoO$_3$/Al, tested under 1000 W m^{-2} air mass 1.5 global (AM 1.5 G) illumination at home laboratory. b) CPVT-certified *J–V* characteristics of a flexible PTB7:PC$_{71}$BM solar cells with a PCE of 8.709% (inset shows a typical photograph of a completed device 2). c) the *J-V* characteristics of device 1, 2, and 4 measured in dark.

Table 1. Best device performance/parameters from the flexible PTB7:PC$_{71}$BM solar cells with PFN and ZnO as electron transporting layer, respectively, measured under 1000 W m^{-2} AM 1.5G illumination. Our previously reported best data from rigid glass is also included for comparison.

Devices	V_{OC}	J_{SC}	FF	PCE
	(V)	(mA cm^{-2})	(%)	(%)
Device 1, PET/ITO/PTB7:PC$_{71}$BM/MoO$_3$/Al	0.59	16	59.4	5.61 [a]
Device 2, PET/ITO/PFN/PTB7:PC$_{71}$BM/MoO$_3$/Al	0.74	17.4	66.8	8.60 [a]
	0.7359	17.94	65.9	8.709 [b]
Device 3, PET/ITO/ZnO/PTB7:PC$_{71}$BM/MoO$_3$/Al	0.74	16	68.6	8.12
Device 4, Glass/ITO/PFN/PTB7:PC$_{71}$BM/MoO$_3$/Al	0.754	17.46	69.99	9.214 [c]

Notes: a) Test performed at home lab, under AM 1.5G illumination with light intensity of 1000 W m^{-2}, with an effective device area of 0.16 cm^2; b) Test performed at CPVT, under AM 1.5G illumination with light intensity of 1000 W m^{-2}, with a mask of 0.1377 cm^2; c) Data obtained from our previous paper.[13]

The PET/ITO substrate coated with PFN also work very well with the most commonly studied P3HT:PC$_{61}$BM system, resulting in a highest PCE of 3.81%, which is very close to that of Glass/ITO-based device (4.15%) by considering the

measurement errors.

The deformation of a series of flexible devices upon thermal treatment at different temperature either at 30, 80, 110, 140, 155 or 170℃ is shown in Fig. 4. We found that there is no appreciable deformation in the devices if the annealing temperature is below 140 ℃ (Fig. 4a-c) while annealing temperature at 155℃ and 170℃ caused a significant bend and a complete roll-up, respectively. The *J*-V characteristics of a typical device 2 (here we take a device stored for more than 150 days as an example) after 110, 140 and 155℃ annealing for 10 min was shown in Figure S3 and the device parameters was summarized in Table S3. It was found that with increasing annealing temperature, the PCE of the flexible device decreased from 6.17% to 5.42%, 5.20%, and 4.88%, respectively. Nevertheless, since all of the film-forming procedures for PFN, photoactive layer were processed at room-temperature, and the thermal deposition of metal electrode would not result in high temperature above 100 ℃ in the devices, and the PTB7:$PC_{71}BM$ system did not require additional thermal annealing, we therefore believe that the method reported here is indeed suitable toward flexible PSCs and the fabricated flexible devices may find practical applications in different occasions.
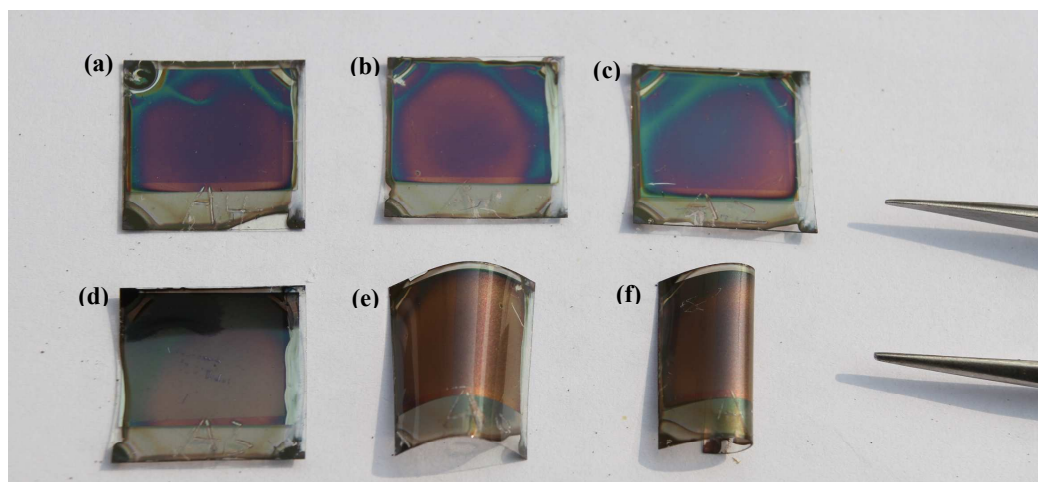
Fig 4. Photographs of the device 2 after thermal treatment at different temperatures. (a)-(f) corresponds to thermal treatment at 30, 80, 110, 140, 155 and 170℃ for 10 minutes atop a hotplate, respectively.

In addition to the high power conversion efficiency of 8.7%, the fabricated device also has a very high specific power (defined as the electrical power output per unit weight) of 400 W kg^{-1}, [35] which is comparable with that of commercially available space-rated high-efficiency silicon cells (~800 W kg^{-1}).[36] It is worthy to note that this metric for this flexible device has the potential of ~10000 W kg^{-1} if ultrathin (i.e 2 um) PET substrate is used.[36] Thus this kind of lightweight, flexible polymer solar cells can potentially outperform inorganic counterpart in some environments and open new opportunities for many novel terrestrial and space applications.

In summary, flexible polymer solar cells with a record high power conversion efficiency of 8.7% and a very high specific power of 400 W kg^{-1} have been demonstrated by depositing physical blend of a conjugated semiconducting polymer and a fullerene derivative on highly flexible polyethylene terephthalate (PET) substrate. The flexible device reported here perform basically as well as those based on rigid glass/ITO substrate in most of the device parameters, except a relative lower

fill factor, which can be further improved via interface engineering between the photoactive layer and electrodes or the development of novel flexible substrates. Thus there are potential for further improvement in device efficiency and efficiency above 9% should be achievable in follow up study. It is worthy to mention such flexible polymer solar cells potentially be suitable for novel terrestrial and space applications, such as bendable, portable power sources etc, owing to their very high specific power. The processing method for the flexible polymer solar cells is solution-based and all of the film deposition processes are completed at room temperature, thus this simplicity of preparing flexible polymer solar cells can offer easy-processibility over large area size with fast deposition speed on an industrial scale at room temperature.

Experimental

Materials and device fabrication: The polyethylene terephthalate (PET) substrate that pre-coated with ITO (PET/ITO, with a sheet resistance of 40 ohm/square, 125μm in thick) was provided by Kaivo Electronic Components Co., Ltd. (Zhuhai, China) and while the control device on ITO coated rigid glass (Glass/ITO, 15 ohm/square) was provided by CSG Holding Co., Ltd. (Shenzhen, China). Electron donor polymers thieno[3,4-b]thiophene/benzodithiophene (PTB7) was purchased from 1-material Chemscitech Inc. (St-Laurent, Quebec, Canada), while $PC_{71}BM$, P3HT was purchased from Luminescent Technology Inc., all were used as received. The interlayer PFN was synthesized in the lab following literature.[37] Device preparation and characterization were carried out in clean room conditions with protection against dust and moisture. The fabrication of PSCs followed the procedures described in our previous papers.[10, 13] The deposition of sol-gel ZnO film followed

the procedure described in a reported literature.[30]

Characterization and measurement: the values of power conversion efficiency were determined from J–V characteristics measured by a Keithley 2400 source-measurement unit under AM 1.5G spectrum from a solar simulator (Oriel model 91192). Masks made from laser beam cutting technology with well-defined area of 16.0 mm^2 or 13.77 mm^2 were attached to define the effective area for accurate measurement. Solar simulator illumination intensity was determined using a monocrystal silicon reference cell (Hamamatsu S1133, with KG-5 visible color filter) calibrated by the National Renewable Energy Laboratory (NREL). The incident photon-to-current efficiency spectra were measured by a solar cells–photodetector responsivity measurement system (Model DSR-100UV-B, Zolix, China). The film thickness of as-spun PFN film was determined by a surface profiler (Alfa Step-500, Tencor), in combination with extrapolation from an absorbance-thickness curve that assumes a linear dependence of absorbance at 380 nm on film thickness. Transmittance was obtained using a UV-VIS-NIR spectrophotometer (Shimadzu UV-3600). The film morphology was studied by atomic force microscopy (AFM, Veeco MultiMode V) operating in tapping mode.

Acknowledgements

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35 Our typical completed device is 1.5cm*1.5 cm in size, 40 mg in weight (unencapsulated), ~120 um in thick, with a active area ratio of 80% and a power conversion efficiency of 8.7%, thus the theoretical upper limitaion for the specific power is determined to be 400 W kg^{-1} while under 1000 W m^{-2} AM 1.5G illumination.

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