

26.75 cm² organic solar modules demonstrate a certified efficiency of 14.34%

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Organic solar cells (OSCs) have been developed rapidly in past years, due to the fast evolution of wide-bandgap copolymer donors and low-bandgap non-fullerene acceptors^[1–9]. At present, the highest power conversion efficiencies (PCEs) for single-junction OSCs and tandem OSCs exceed 19% and 20%, respectively^[10, 11]. These OSCs are typically fabricated by using low-boiling-point solvent chloroform (CF) with an effective area <0.1 cm². The doctor-blading deposition is the most advantageous technique to fabricate OSCs with low-boiling-point solvent for upscaling lab cells to industrial-scale modules^[12], exhibiting simple operation, low cost, and high material utilization^[13–15]. Herein, a typical OSC material system PM6:Y6 (Fig. 1(a)) was used to fabricate OSCs modules *via* doctor-blading deposition in ambient condition, and the influence of the ambient temperature and substrate temperature on the film quality was investigated. Furthermore, PC₆₁BM was incorporated into PM6:Y6 blend, and a certified record efficiency of 14.34% was achieved for OSC modules with a designated illumination area of 26.75 cm².

Some works indicated that the performance parameters for doctor-bladed OSCs from low-boiling-point solvent CF were normally dependent on the temperatures (25–45 °C)^[12, 16–18]. However, controlling the ambient temperature and substrate temperature at the same value, for example, from 17 to 30 °C, interesting results were achieved, *i.e.*, doctor-bladed film quality and device performance are almost independent on temperature. Atomic force microscopy (AFM) images of doctor-bladed PM6:Y6 films at different temperatures (17, 24 and 30 °C) are almost the same (Fig. S1). Meanwhile, the root-mean-square roughnesses (R_{rms}) are around 1.50 nm. In addition, grazing incidence wide-angle X-ray scattering (GIWAXS) patterns for doctor-bladed PM6:Y6 thin films at different temperatures also exhibit the similar line-cut profiles (Fig. 1(b) and Fig. S2). The (010) peaks at

1.73 Å⁻¹ in the out-of-plane (OOP) direction and the (100) peaks at 0.29 Å⁻¹ in the in-plane (IP) direction indicate the co-existence of face-on and edge-on orientation in three cases. The diffraction peaks of PM6:Y6 films locate at the same q values, indicating the same lamellar stacking distance and π - π stacking distance.

Doctor-bladed cells at three temperatures were fabricated with a structure of ITO/ZnO:PEI/PM6:Y6/MoO₃/Ag^[19–21]. The current density–voltage (J – V) curves and external quantum efficiency (EQE) spectra are shown in Figs. 1(c) and 1(d), and the performance data are listed in Table S1. The cells processed at 17, 24 and 30 °C exhibit PCEs of 14.57%, 14.78% and 14.64%, respectively, and the average PCEs are 14.35%, 14.58% and 14.46%, respectively. The results suggest that the performance parameters for three cases are almost the same, which are consistent with the results of morphology and GIWAXS. The dependence of J – V characteristics on light intensity (P_{light}) was used to investigate the recombination kinetics in cells. As shown in Fig. S3(a), three cells show an almost equal a value of 0.975, indicating that the bimolecular recombination is not remarkable in PM6:Y6 cells made at three temperatures. The V_{oc} as a function of P_{light} is plotted in Fig. S3(b). When the slope of V_{oc} versus $\ln(P_{\text{light}})$ is equal to 1 or 2 kT/q , the dominant recombination mechanism is bimolecular recombination or Shockley-Read-Hall recombination, respectively. The calculated slopes for the devices at 17, 24 and 30 °C are 1.398, 1.387 and 1.414 kT/q , respectively. The results indicate that PM6:Y6 cells processed at three temperatures have relatively strong trap-assisted recombination, instead of bimolecular recombination.

Furthermore, doctor-bladed large-area OSC modules were made at 24 °C in ambient condition. The J – V curves of the modules with a designated illumination area of 26.75 cm² are shown Fig. 1(e), and the photovoltaic parameters are listed in Table 1. The modules gave a PCE of 13.71%, with a V_{oc} of 10.73 V, a J_{sc} of 1.792 mA/cm², and an FF of 71.29%. The certified PCE is 13.56% by the Chinese National PV Industry Measurement and Testing Center (NPVM) (Figs. S4–S10). Using fullerene derivatives is a simple and effective way to enhance

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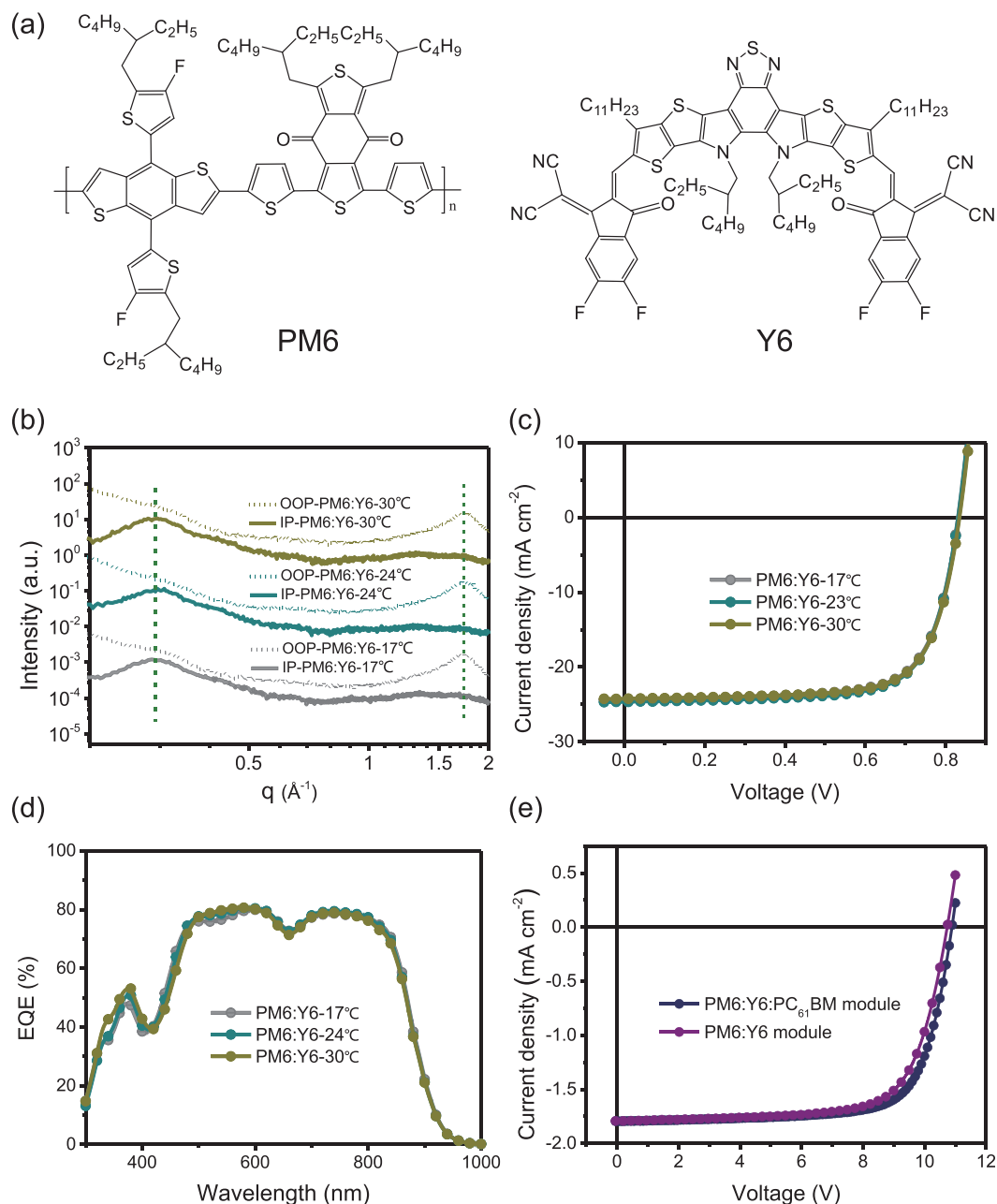


Fig. 1. (Color online) (a) Chemical structures of PM6 and Y6. (b) 1D GIWAXS line curves along the out-of-plane (OOP, dotted line) and in-plane (IP, solid line) directions. (c) J - V curves for PM6:Y6 cells made at different temperatures. (d) EQE spectra for PM6:Y6 cells made at different temperatures. (e) J - V curves for PM6:Y6 and PM6:Y6:PC₆₁BM modules with a designated illumination area of 26.75 cm².

Table 1. Performance data for doctor-bladed modules with a designated illumination area of 26.75 cm².

Active layer	V_{oc} (V)	J_{sc} (mA/cm ²)	FF (%)	PCE (average) (%)
PM6:Y6	10.73	1.792	71.29	13.71 (13.51)
PM6:Y6 ^a	10.64	1.815	70.22	13.56
PM6:Y6:PC ₆₁ BM	10.89	1.795	73.42	14.35 (14.01)
PM6:Y6:PC ₆₁ BM ^a	10.85	1.814	72.89	14.34

^aCertified results in the Chinese National PV Industry Measurement and Testing Center.

ce PCE^[22–24]. Thus, it is possible to further improve the module efficiency *via* introducing PC₆₁BM into PM6:Y6 blend. The modules based on PM6:Y6:PC₆₁BM (1 : 1 : 0.2) offer a record PCE of 14.35%, with a V_{oc} of 10.89 V, a J_{sc} of 1.814 mA/cm², and an FF of 73.42% (Table 1). NPVM demonstrates a PCE of 14.34% (Figs. S11–S17). This certified PCE is the highest value reported to date for large-area OSC modules.

In short, we found that the morphology and device performance for doctor-bladed PM6:Y6 thin films from low-boiling-point solvent are almost independent on the temperatures if controlling the ambient temperature and substrate temperature at the same value. With a designated illumination area of 26.75 cm², the large-area PM6:Y6 modules demonstrate a certified PCE of 13.56%, and the PM6:Y6:PC₆₁BM mod-

ules demonstrate a certified record PCE of 14.34%.

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Appendix A. Supplementary materials

Supplementary materials to this article can be found online at <https://doi.org/10.1088/1674-4926/43/10/100501>.

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