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**Rachel Jakubiak
Manfred Eich
Jean-Michel Nunzi**
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Plastic Solar Cells: Breaking the 10% Commercialization Barrier

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ABSTRACT

Tandem solar cells provide an effective way to harvest a broader spectrum of solar radiation by combining two or more solar cells with different absorption ranges. However, for polymer solar cells (PSCs), the performance of tandem devices lags behind single-layer cells mainly due to the lack of a high-performance low-bandgap polymer with appropriate spectral response range. Here, we demonstrate a novel low bandgap conjugated polymer (~1.44 eV) specifically suitable for tandem structure. In the single-layer device, power conversion efficiency (PCE) of 6.5% was achieved. When the polymer was applied to tandem solar cells, we demonstrated a NREL certified PCE of 8.62%^[1]. Further optimization on materials and devices of this system has led to record breaking efficiency of 10.6%. Furthermore, the tandem devices show excellent stability due both to the intrinsic stability of the polymer and the advanced device structure.

Keywords: Low bandgap polymer, polymer solar cell, tandem polymer solar cell

1. INTRODUCTION

Organic photovoltaic (OPV) devices are promising for low-cost, flexible, lightweight, large-area energy generation applications, and much work on designing new materials, device structures, and processing techniques has been carried out to improve the power conversion efficiency (PCE) of OPV devices. So far, polymer solar cells (PSCs) based on conjugated polymers as electron-donor materials blended with [6,6]-phenyl-C71-butyric acid methyl ester (PC71BM) as an electron-acceptor material have achieved around 8% PCE using a bulk heterojunction (BHJ) device structure.^[2-5] Nonetheless, most of these materials suffer from the inherent disadvantages of either lacking a broad absorption range (bandgap [Eg] = 1.6 eV~2.0 eV), or relatively low carrier mobility. This reduces the external quantum efficiency (EQE) and photocurrent of the devices. To use solar radiation more effectively, an effective approach is to stack multiple photoactive layers with complementary absorption spectra in series to make a tandem PSC. Typically, such a tandem structure consists of a front cell with a high-bandgap material, an interconnecting layer (ICL), and a rear cell with a low-bandgap (LBG) material. By using more than one material, each material captures a different range of the solar spectrum and increase the open-circuit voltage (VOC) and thus the efficiency.^[6-7]

The photoactive materials play a critical role in determining the PCE; however, due to the great difficulties, there have been few reports to date on designing photoactive materials specifically for high-efficiency tandem PSCs. To be applied effectively in a tandem structure, sub-cell materials have several requirements, especially for rear-cell LBG polymers. First, a small energy bandgap (<1.5 eV) is critical so that the overlap of absorption spectra between the front and rear cells can be minimized.³⁴ Second, fine-tuning is required of the highest-occupied molecular orbital (HOMO) and lowest-unoccupied molecular orbital (LUMO) levels to achieve high VOC with a small bandgap while maintaining a LUMO level high enough for efficient charge separation. Third, high charge-carrier mobility and fine phase separation with the acceptor are required for high short-circuit current density (JSC) and fill factor (FF) in single-cell devices.²⁷⁻³¹ Therefore, a carefully designed LBG polymer will perform well for tandem cells only if it can achieve high current by efficiently using the low-energy portion of the solar spectrum. Currently, the existing LBG materials (Eg < 1.5 eV) show either low VOC, low JSC, or low FF values, which are far from ideal for the tandem structure. Thus, the PCE of tandem cells reported so far is less than 7%.

2. LOW BANDGAP POLYMER DESIGN

Here, we demonstrate the rational design of a novel LBG conjugated polymer poly{2,6'-4,8-di(5-ethylhexylthienyl)benzo[1,2-b;3,4-b']dithiophene-alt-5-dibutyloctyl-3,6-bis(5-bromothiophen-2-yl)pyrrolo[3,4-c]pyrrole-

1,4-dione} (PBDTT-DPP, Figure 1a) specifically for tandem solar cells. To achieve a small bandgap, a polymer backbone based on the diketopyrrolopyrrole (DPP) unit and benzodithiophene (BDT) unit was chosen. By replacing the oxygen atoms attached to the BDT unit with thiophene moieties to form the thienylbenzodithiophene (BDTT) unit, the HOMO and LUMO levels of PBDTT-DPP are moved deeper to increase VOC without losing the driving force for efficient charge separation, while keeping the bandgap within the ideal range. Furthermore, bulkier 2-ethylhexyl side chains on BDTT and 2-butyloctyl side chains on DPP are used to increase the solubility of the resulting polymers and thus obtain much higher molecular weights. Compared to PBDDT-DPP, PBDTT-DPP showcased improved solubility, higher molecular weight, and higher carrier mobility, which leads to a significantly higher JSC in single-cell devices. Finally, we applied this LBG polymer into a newly designed inverted structure of a tandem solar cell, and a PCE of 8.62% was certified by the National Renewable Energy Laboratory (NREL). Further optimization on materials and devices of this system has led to record breaking efficiency of 10.6%.

3. DEVICES PERFORMANCE

Single-layer BHJ photovoltaic cells based on PBDTT-DPP blended with PC71BM were fabricated with a regular and an inverted configuration. The optimized polymer:PC71BM blend ratio (by weight) was found to be 1:2 and the optimized film thickness was around 100 nm. The single-cell PV performance of PBDTT-DPP is shown in Figure 1. Of the more than 300 devices that we fabricated, the best devices gave values of V_{OC} around 0.74 V, J_{SC} around 13.5 mA/cm², fill factor around 65%, and PCE as high as 6.5% for both the regular and inverted structure.

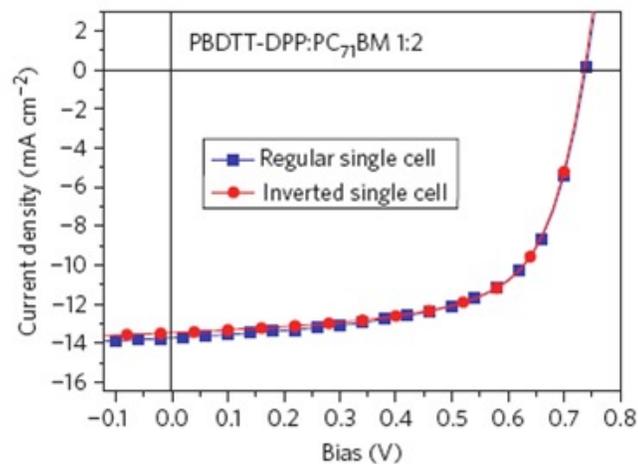


Figure 1. J–V characteristics of single-cell devices with regular and inverted structures under AM1.5G illumination from a calibrated solar simulator with an irradiation intensity of 100 mWcm².

Inverted tandem solar cells were fabricated using the rear-cell-specific LBG polymer PBDTT-DPP and the new device architecture. The J-V characteristics, EQE of a typical device are shown in Figure 2 (a) and (b), respectively. EQE of individual sub-cells in the tandem structure was measured by using light bias. As shown in Figure 4a, the front cell had photo-response from 300 to 600 nm, showed EQE as high as 60% at 530 nm, and its integrated JSC was 8.2 mA/cm²; the rear cell had broad photo-response from 300 to 850 nm, showed a maximum EQE of 47% at 770 nm, and its integrated JSC was 8.1 mA/cm². The incident light from 300 to 600 nm was strongly absorbed by the front cell and the EQE of the rear cell in this region was much lower than that of its single-cell devices; however, the rear cell can still provide enough photo-current (8.1 mA/cm²) to match the current that the front cell could supply (8.2 mA/cm²) because the new material PBDTT-DPP can very efficiently use the low-energy portion (from 600 to 850 nm) of the solar radiation.

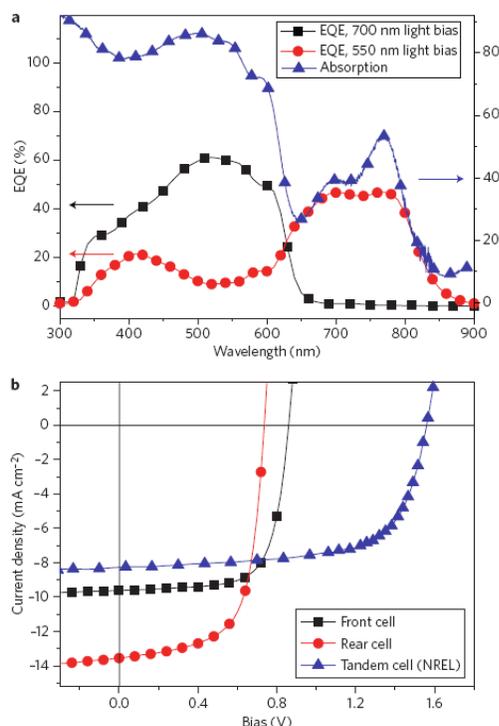


Figure 2 Quantum efficiencies, J–V characteristics and stability of single cells and tandem cells. a, EQEs of the inverted tandem solar cell and absorption spectrum (without metal electrode). b, J–V characteristics of the front cell, rear cell and inverted tandem solar cell, tested at NREL using the OSMSS simulator.

4. CONCLUSION

In summary, we have demonstrated a high-performance LBG conjugated polymer specifically for tandem PSCs by applying three design rules - lowering bandgap for spectral-matching with front cell, controlling HOMO/LUMO levels to enhance VOC, and increasing molecular weight to enhance JSC and FF. The novel polymer (PBDTT-DPP) has a small optical bandgap, deep HOMO level, and high hole mobility. Single-layer BHJ solar cells fabricated from PBDTT-DPP and PC71BM exhibited PCEs of more than 6%. The significance of the PBDTT-DPP polymer is clearly demonstrated in inverted tandem PSCs with 8.62% PCE under AM1.5G one-sun illumination as tested by NREL. This study opens up a new direction for polymer chemists to design new materials for tandem PSCs and also makes an important step forward toward commercialization of PSCs. Further optimization on materials and devices of this system has lead to record breaking efficiency of 10.6%.

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