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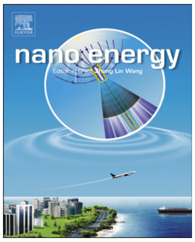

RAPID COMMUNICATION

# Low temperature efficient interconnecting layer for tandem polymer solar cells

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**Abstract**
Tandem solar cells are found to be promising in organic photovoltaic research because they can double open-circuit voltage ($V_{oc}$) and utilize photon energy efficiently, as compared to single junction solar cells. In a typical tandem polymer solar cell, two single subcells are staked and connected by an interconnecting layer, which is a prerequisite to achieving high photo-electron conversion efficiency. In this work, a low temperature solution processed and chemically stable PEDOT:PSS/AZO/PEIE interconnecting layer is reported. Tandem polymer solar cells based on this efficient layer can realize a high $V_{oc}$, which can sum the $V_{oc}$'s of subcells. Furthermore, the operation mechanism of the interconnecting layer is discussed to help better understand the electron recombination process in the interconnecting layer.

## Introduction

Organic solar cells have attracted much attention in the past decade, because of their solution based processing and potentially low cost as renewable energy resources. However, one problem to common organic solar cells is that the absorption spectral range of donor polymers is narrow, which limits the solar energy utilization of the entire spectrum. To achieve a broadband absorption, tandem structures have been employed by stacking two or more bulk heterojunction (BHJs) with complementary absorption spectra. The open circuit voltage ($V_{oc}$) of a tandem cell is typically the sum of $V_{oc}$'s of individual subcells, while the overall current density is limited by the subcell with lower current density. Tremendous efforts have been devoted to the development of desirable device configurations and interconnecting layers (ICLs) between subcells. Tandem cells with a power conversion efficiency (PCE) over 10.6% have been achieved [1]. To construct high efficiency tandem polymer solar cells, high-performance individual subcells

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using polymers with complementary absorption are needed [2]. Additionally, a tandem solar cell requires an ICL between subcells [3]. Ideally, the ICL and subcells should form ohmic contact, and the ICL also should function as a charge recombination layer to complete the circuit so that the $V_{oc}$ can add up. In inorganic tandem solar cells, the interconnecting layer is a tunneling diode consisting of heavily doped n- and p-type layers. In their organic counterparts, small moleculare tandem solar cells are simpler because the interconnecting layer and the top subcell are thermally evaporated. For example, Leo et al. evaporated highly p-doped N,N'-diphenyl-N,N'-bis(4-(N,N-bis(naphth-1-yl)-amino)-biphenyl-4-yl)-benzidine (DiNPB) and n-doped $C_{60}$ directly connecting two cells as a recombination layer [4]. The evaporation based approach has disadvantages including complicated process and high cost. Solution processed polymer tandem cells also have challenges. In addition to optical transparency and electrical functionality, the interconnection layer coating should not damage the existing subcells. Thus, the solvent selection is critical [2]. The penetration of solvents through the ICL may disturb charge carrier selectivity of the ICL between two subcells, and thus enhance the charge recombination loss [3]. Therefore, a reduced $V_{oc}$ is commonly observed for tandem cells. Furthermore, once coated, the ICL should provide protection so that the sequential BHJ active layer of the second subcell will not damage the underlying layers. Therefore, the ICL should have mechanical robustness in addition to device functionality.

The knowledge regarding interfacial layers accumulated in the organic light emitting diode (OLED) and organic photovoltaic (OPV) research fields provided strong support for tandem OPV interconnecting layer development. In 2006, Kawano et al. demonstrated a polymer tandem solar cell using sputtered indium tin oxide (ITO) combined with poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) as inter-connecting layer [5]. However, there was a loss in $V_{oc}$ of the tandem cell compared to the reference single cells because of the high contact resistance between the ITO and bottom cell interface. Janssen et al. showed an efficient ICL consisting of n-type and p-type layers, where the n-type layer is a bi-layer structure lithium fluoride/aluminum (LiF/Al) and the p-type layer is a bi-layer of gold (Au)/PEDOT [6]. However, the metal (Au) layer in this ICL is thick, which leads to a loss in transmittance of photons for the rear subcell. In addition, both the p- and n-type layers are formed through vacuum deposition, not all solution processed. Solution-processed electron transport layers such as zinc oxide (ZnO) and titanium dioxide ($TiO_2$) have been used as n-type interlayer in single junction polymer solar cells [7-13]. These electron-transporting layers are wide band gap semiconductors and are highly transparent in the visible and near-infrared (NIR) wavelength range where the polymer solar cells absorb. On the p-type side, PEDOT:PSS is an excellent hole-transporting layer for polymer solar cells [14]. It has been reported that the regular PEDOT:PSS layer is not strong enough to provide protection. The solution to this problem is using a low boiling temperature solvent (e.g., chloroform) for the deposition of the second subcell. High performance polymer solar cells are typically based on 1,2-Dichlorobenzene (ODCB) or chlorobenzene (CB) because they can facilitate morphology development for effective charge transport and collection. To overcome the challenge of making high performance tandem solar cells using higher boiling point solvent, Yang et al. introduced surfactant and dimethylformamide (DMF) into PEDOT:PSS to improve the mechanical properties and the conductivity of PEDOT:PSS. The modified PEDOT:PSS layer became very robust and showed excellent electrical conductivity and transparency [3,15]. PCE of 5.84% with a regular tandem device architecture utilizing an Al-$TiO_2$-PEDOT:PSS interlayer was achieved by Srista et al. in 2010 [1]. An inverted tandem cell with a PCE of 8.62% using modified PEDOT:PSS-ZnO interlayer was achieved by Dou et al. in 2012. [15] There were issues with ZnO stability such as aggregation of ZnO nanoparticles. Repeated ultraviolet (UV) illumination is required to remove the “S shape” current density-voltage (J-V) curves in ZnO/PEDOT:PSS interlayer and form ohmic contact at the interconnecting layer [16]. Room temperature processable metal oxides are ideal materials for developing efficient and stable single layer and multilayer organic solar cells, though several commonly employed metal-oxide interlayers such as ZnO demanding post-thermal annealing to attain improved conductivity [17]. Such high temperature processing is a drawback for realizing competitive organic solar cells. Generally speaking, an ideal interconnecting layer needs to possess an energy level matching with those of donor and acceptor molecules in the active layer, sufficient conductivity, high transparency, uniform coverage and good chemical stability. Several interconnecting layers including $TiO_x$/PEDOT:PSS [15], ZnO/PEDOT:PSS [3], molybdenum trioxide ($MoO_3$)/Al/ZnO [18], Al/$TiO_2$/PEDOT:PSS [1], LiF/Al/Au/PEDOT: PSS [6], LiF/Al/$MoO_3$ [19], and $MoO_3$/Ag/Al/Calcium (Ca) [20] have recently been demonstrated. More recently, graphene oxide (GO) based interconnecting layers, such as ZnO/GO:PEDOT:PSS [21] and ZnO/GO:single-walled carbon nanotube (SWNTs) [22] have also been reported. Because of the low conductivity of GO, however, certain highly conductive materials (e.g., PEDOT:PSS, SWNTs) have to be blended into the GO interconnecting layer, which complicated the device fabrication and limited the $V_{oc}$ of tandem cells to be only $\sim$80% of the sum of that of the subcells. There is a need to provide stable and optical transparent interconnecting layers that can be processed by low temperature solution [23].

Brabec et al. demonstrated aluminum-doped zinc oxide (AZO) layers from a sol-gel precursor, which can be processed at lower processing temperatures ($<150\,^{\circ}C$). AZO with lower processing temperatures ($<150\,^{\circ}C$) offers a suitable temperature for plastic substrates and deposition onto organic films [24] because some medium and low bandgap polymers cannot bear high temperatures. Low temperature solution processed interconnecting layer is a requirement for stacking two or more bulk heterojunction (BHJs) in tandem solar cells to achieve a broad absorption. Previous research revealed that the S-shape curves could be derived from an interfacial barrier [25]. Ethoxylated polyethylenimine (PEIE) can facilitate electron transport by lowering the energy barrier between the active layer and ITO cathode in BHJ devices [26]. Here, we report PEDOT: PSS/AZO/PEIE as a low temperature processable, efficient interconnecting layer for tandem polymer solar cells. Devices based on this efficient layer can realize high $V_{oc}$'s, which can sum the $V_{oc}$'s of subcells. The operation mechanism of the interconnecting layer is also discussed to help understand the electron recombination process in the interconnecting layer.