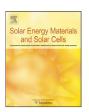
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## Studies of the optimization of recombination layers for inverted tandem polymer solar cells

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#### ABSTRACT

We report on an optimization study of recombination layers in inverted tandem bulk heterojunction polymeric solar cells. Modification of bilayers of  $MoO_x/Ag$  with a polymeric surface modifier, polyethylenimine ethoxylated (PEIE), is found to significantly improve the performance of these recombination layers. The improvement is assigned to a significant reduction by 1.5 eV of the work function of the Ag with PEIE. An analysis of the electrical, optical, and surface properties of the recombination trilayer is presented. Inverted tandem polymer solar cells, fabricated with two identical active layers comprising a donor polymer poly(3-hexylthiophene) (P3HT), and a fullerene derivative acceptor, [6,6]-phenyl  $C_{61}$  butyric acid methyl ester (P $C_{60}BM$ ), provide a proof-of-principle demonstration of the effectiveness of the  $MoO_x/Ag/PEIE$  trilayer as recombination layer. Such tandem cells yield an open-circuit voltage of 1042 mV and fill factors of 0.62, comparable to those of single cells (0.63) under simulated AM 1.5G, 100 mW/cm² illumination. Preliminary shelf lifetime studies suggest that these trilayer recombination layers show good air stability.

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#### 1. Introduction

Polymer-based solar cells (PSCs) have gained much interest for their potential as a promising renewable energy source with simple and low-cost processing [1,2]. Intensive efforts around the world in developing novel photoactive materials for PSCs have resulted in published power conversion efficiencies of 8.6% [3]. Compared to inorganic solar cells, the lower efficiencies obtained in PSCs arise primarily from relatively narrow absorption bands which do not provide an extensive overlap with the solar spectrum and from limitations in the thickness of the active layers that can be used due to limited extinction coefficients and short carrier lifetimes. Even with recently developed small band gap polymers, less than 60% of the incident solar power is absorbed [4]. Maximizing light absorption by increasing the thickness of the layers can reduce the carrier collection efficiency and make the cell too resistive, which will have an impact on its performance. Tandem cell geometries provide a potential solution to this tradeoff problem and a route to achieve PSCs with high power conversion efficiency.

In a tandem solar cell, two or more PSCs are connected in series by stacking them on top of one another. The interlayer connecting two adjacent cells, referred to as recombination layer, is critical for the tandem PSC performance since it needs to facilitate efficient recombination of opposite charge carriers coming from the adjacent PSCs. Hence, the recombination layer needs to present a first surface with a work function that is large enough to collect holes produced in the donor-like semiconductor in a first PSC, and a second surface with a work function that is low enough to collect electrons from the acceptor-like semiconductor in the second, adjacent, PSC. The order in which these two surfaces are distributed in the tandem structure depends upon the work function of the top and bottom electrodes. At the same time, the recombination layer needs to be optically transparent so that its presence does not limit the ability of the light to reach the innermost PSCs.

In inverted solar cells a transparent low-work function electrode, commonly a metal-oxide, is used as the bottom electrode and a high work function metal is used as the top electrode. Inverted solar cells avoid the use of reactive low-work function metal electrodes and therefore are attractive geometries to increase the air stability of PSC [5–7]. Inverted tandem solar cells have been demonstrated with power conversion efficiencies below 8.6% [8]. To date, a common route to realize a recombination layer in an inverted tandem PSC is to use a first layer

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comprising a metal-oxide with a high-work function (WF), typically  $MoO_x$  [9,10] (having a WF of 6.9 eV) [11], a second layer comprising an ultrathin (non-continuous) layer of a metal such as Al [9] or Ag [10] and a third layer comprising either a low-work function metal such as Ca [10] or a metal oxide such as ZnO [8,9] which assures good electron collection from the adjacent PSC. The use of a reactive metal such as Ca is not desirable in an inverted structure. On the other hand, the relatively large work function of ZnO (4.3 eV [6,12] when exposed to ambient conditions) may limit the choices for acceptor materials from which efficient electron collection can be achieved.

We recently reported on the use of polymers containing simple aliphatic amine groups such as polyethylenimine ethoxylated (PEIE), as an efficient and universal method to reduce the WF of conductors. These polymer layers are processed in air from aqueous solutions, and are physisorbed onto the conductor's surface. The physisorbed polymer layer creates a strong dipole at the interface of the conductor which causes a strong reduction of its WF. Using a Kelvin probe in air, we have measured reductions of the WF of Ag from 4.6 eV down to 3.7 eV [12].

In this study, we report on the use of PEIE in inverted tandem PSC structures. The tandem PSC uses an ITO/PEIE bottom electrode as the transparent electron collecting electrode and a recombination layer comprising a  $\text{MoO}_x/\text{Ag/PEIE}$  trilayer. Reductions of the work function of ITO and  $\text{MoO}_x/\text{Ag}$  are studied by Kelvin probe measurements and ultraviolet photoelectron spectroscopy (UPS). Inverted tandem solar cells with two identical sub-cells comprising a P3HT:PC\_{60}BM active layer showed a power conversion efficiency of  $2.1 \pm 0.1\%$  and an open circuit voltage of  $1042 \pm 6 \text{ mV}$  under  $100 \text{ mW/cm}^2$  AM 1.5G solar illumination. These cells have shown good shelf air stability for at least 140 h.

#### 2. Experimental section

#### 2.1. Solution preparation

Solutions of the organic films used in this study were prepared as follows: polyethylenimine ethoxylated (PEIE) [80% ethoxylated,  $M_w \sim 70,000~\rm g/mol,~35-40~\rm wt\%$  in water purchased from Aldrich, St. Louis, MO] (Fig. 1(a)) was diluted into 2-methoxyethanol (Aldrich, St. Louis, MO) to a concentration of 0.5 wt% and stirred overnight in ambient air; a solution of P3HT (4002E, Rieke Metals, Lincoln, NE):PC<sub>60</sub>BM (Nano-C, Westwood, MA) was prepared in chlorobenzene (Aldrich, St. Louis, MO) with a weight ratio of 1:0.7 (P3HT:PC<sub>60</sub>BM) and a total P3HT+PC<sub>60</sub>BM concentration of 17 mg/ml. The P3HT:PC<sub>60</sub>BM solution was stirred overnight in a nitrogen-

filled glove box, and heated at a temperature of 45  $^{\circ}\text{C}$  for 1 h before use.

#### 2.2. Device fabrication and characterization

ITO-coated glass (Colorado Concept Coatings LLC) with a sheet resistivity of 15  $\Omega$ /sq was used as the substrate. Substrates were cleaned in an ultrasonic bath of detergent (Liqui-Nox® Phosphate-Free Liquid Detergent, Alconox, Inc., White Plains, NY) water, rinsed with deionized water, and then cleaned in sequential ultrasonic baths of deionized water, acetone, and 2-propanol. Nitrogen was used to dry the substrates after each bath. On each ITO/glass substrate, a 300-nm-thick layer of SiO<sub>x</sub> (Silicon Monoxide, Kurt J. Lesker, Clairton, PA) was deposited by e-beam deposition (AXXIS, Kurt J. Lesker, Clairton, PA), through a shadow mask, at a rate of 0.4–0.5 nm/s and a base pressure of  $\sim 1 \times 10^{-7}$  Torr to electrically isolate around half of the glass/ITO area. Next, the substrates were ultrasonicated in 2-propanol for 10 min and dried with nitrogen. A 10 nm-thick PEIE layer was spin casted onto the substrates in air, at a speed of 5000 rpm for 1 min, by filtering the PEIE solution through a 0.2-µm-pore PTFE filter. The PEIE-casted substrates were dried on a hotplate at a temperature of 120 °C for 10 min in air. PEIE-coated substrates were loaded into a nitrogen-filled glove box. Films of P3HT:PC<sub>60</sub>BM around 70-nm thick were deposited by spin coating, at 1000 rpm for 1 min, by filtering the solution through a 0.2-µm-pore PTFE filter, and annealed at 160 °C for 10 min under a nitrogen atmosphere. Thicknesses of all layers were measured by spectroscopic ellipsometry (J.A. Woollam Co., Inc, Lincoln, NE).

For single cells, samples were loaded into a vacuum thermal evaporation system (SPECTROS, Kurt J. Lesker, Clairton, PA) connected to a nitrogen-filled glove box and a 6 nm-thick layer of MoO<sub>x</sub> was deposited through a shadow mask at a rate of 0.1–0.15 nm/s and a base pressure of  $\sim\!8\times10^{-8}$  Torr. Without breaking vacuum, a 150 nm-thick Ag layer was deposited through the previously used shadow mask, to define the active area, at a rate of 0.1–0.2 nm/s and a base pressure of  $\sim\!1\times10^{-7}$  Torr. The active area was around 0.1 cm² and determined accurately under a microscope for each individual device.

For tandem cells, a 6 nm-thick  $MoO_x$  and a 1 nm-thick (estimated from the crystal thickness monitor) Ag layers were sequentially deposited as previously described. In some samples, a second PEIE layer was spun on top of the Ag (1 nm) layer as previously described. Then, a second P3HT:PC<sub>60</sub>BM layer was deposited on top of PEIE-coated and uncoated samples as previously described. These samples were loaded back into the vacuum thermal evaporation system where a 6 nm-thick  $MoO_x$  layer and a 150 nm-thick Ag layer were sequentially deposited to

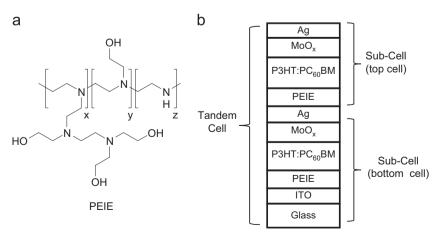


Fig. 1. (a) Molecular structure of polyethylenimine ethoxylated (PEIE) and (b) device structure of an inverted tandem P3HT:PC<sub>60</sub>BM solar cell with a recombination layer of PEIE modified MoO<sub>x</sub>/Ag layer (within left brace) and an inverted single P3HT:PC<sub>60</sub>BM solar cell (within right brace).

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