



# Polymer surface modification to optimize inverted organic photovoltaic devices under indoor light conditions

Ji Soo Goo, Sang-Chul Shin, Young-Jun You, Jae Won Shim\*

Division of Electronics and Electrical Engineering, Dongguk University, Seoul 04620, Republic of Korea



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## ABSTRACT

The objective of this study is to determine the photovoltaic properties of inverted organic photovoltaic devices (OPVs) that employ indium–tin oxide (ITO) modified by ethoxylated polyethylenimine (PEIE) as an electron-collecting electrode under indoor lighting conditions. Owing to the reduced current generation at low light intensity, possessing a sufficiently large shunt resistance ( $R_p$ ) associated with a low leakage current is of utmost importance for performance maximization. The photovoltaic properties under this lighting condition were optimized by controlling the thickness of the PEIE layers, and the electrical, optical, and surface properties of the PEIE-modified ITO were determined. Inverted OPVs based on poly(3-hexylthiophene)–indene- $C_{60}$  bisadduct (P3HT–ICBA) with an 8.5-nm-thick PEIE layer yielded a maximum power-conversion efficiency (PCE) of 13.9% under an LED light with a luminance of 500 lx. Meanwhile, the same device showed poor performance with a PCE of 3.1% under 1 sun illumination owing to the insulating nature of PEIE.

## 1. Introduction

With the emergence of self-contained low-powered indoor electronic devices such as wireless sensor nodes (WSNs) for the Internet of Things (IoT), smart phones, tablet PCs, and laptops, the use of batteries as their power source is rapidly surging [1–3]. However, conventional batteries require periodic replacement and maintenance, thus spurring the rising demand for new types of energy-harvesting systems that employ ambient energy to operate indoor electronic devices. There are typically four main energy sources from the ambient environment: light, heat, radio waves, and mechanical vibrations. Among these energy sources, light and heat appear to be the preferred choices for indoor applications in terms of their performance. However, a very significant heat difference must be provided to supply enough energy with the heat source; photovoltaic technology that converts light energy into electricity is far more suitable than others for this purpose [4–6].

The conditions of indoor lighting are significantly different from those of outdoor lighting (defined as 1 sun illumination). Typical indoor light sources such as light-emitting diodes (LEDs), incandescent lights, and fluorescent lights have only 0.05–0.1% of the intensity of outdoor light (100 mW/cm<sup>2</sup>). Furthermore, the irradiation spectra of indoor lights are different from the solar spectrum (air mass (AM) 1.5 global (G)) (Fig. 1(a)) [7]. Among the photovoltaic technologies, organic photovoltaics (OPVs) offer the best performance in indoor power conversion, where a high absorption coefficient and a tunable absorption

range of a photoactive layer can allow effective absorption under weak indoor lighting. In addition, the flexibility, lightness, and various colors of OPV cells fit well with indoor applications. Finally, the viability of employing eco-friendly materials for the devices can be another advantage for interior utilization or building purposes [8–10]. For example, OPVs with photoactive layers composed of poly(3-hexylthiophene-2,5-diyl) (P3HT), one of the most widely employed donor polymers, and fullerene-based acceptors such as indene- $C_{60}$  bisadduct (ICBA) and [6,6]-phenyl- $C_{61}$ -butyric acid methyl ester (PCBM) yielded power-conversion efficiencies (PCEs; with ICBA: 4.90%; with PCBM: 3.68%) under indoor lighting conditions (e.g., illumination by LED) that are more than two or three times of those under 1 sun illumination (with ICBA: 13.05%; with PCBM: 8.90%) [11]. Recently, many reports have shown that OPVs have better photovoltaic performance than other photovoltaic technologies such as Si solar cells and copper–indium–gallium (di)selenide (CIGS) solar cells under indoor conditions [2,5,6].

To optimize the performance of OPVs under indoor lighting conditions, different approaches must be considered for 1 sun illumination. OPVs can be modeled with a current source, an ideal diode, and two parasitic resistances [a series resistance ( $R_s$ ) and a shunt resistance ( $R_p$ )] under illumination (Fig. 1(b)) [12–14].  $R_s$  accounts for the resistance of electrodes, the resistance where contacts meet the ideal diode, and the resistance in materials that form the diode.  $R_p$  is related to the leakage current through the ideal diode, e.g., by pinholes in the

\* Corresponding author.

E-mail address: [jwshim@dongguk.edu](mailto:jwshim@dongguk.edu) (J.W. Shim).

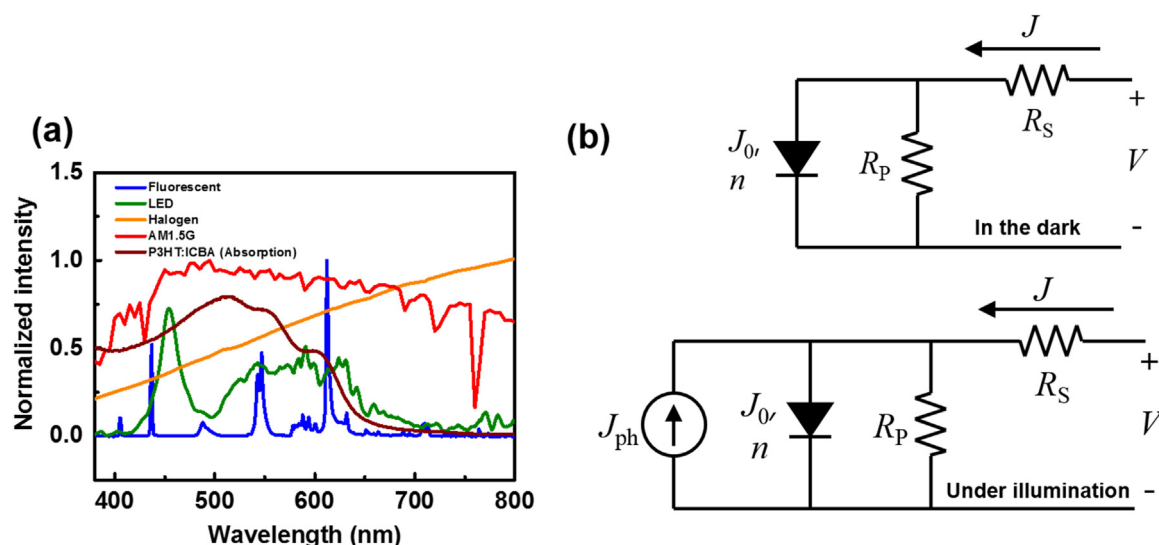


Fig. 1. (a) Spectra of various light sources employed in this study—AM 1.5G solar simulator (red), fluorescent lamp (blue), LED lamp (green), and halogen lamp (orange)—and the absorption spectrum of P3HT-ICBA (brown). (b) Equivalent-circuit models in the dark and under illumination. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

device, recombination at an interface [8]. Among the components of the circuit model,  $R_p$  plays a critical role because of the reduced current density under indoor lighting conditions. Recently, Steim et al. illustrated the effects of  $R_p$  and  $R_s$  on the indoor and outdoor performance of OPVs [7]. They suggested that lower  $R_s A$  ( $< 3 \Omega \text{ cm}^2$ , where  $A$  stands for the area) under outdoor conditions and higher  $R_p A$  ( $> 85 \text{ k}\Omega \text{ cm}^2$ ) under indoor conditions (1000 lx) could lead to better device performance [7]. Zhou et al. also showed the roles of  $R_p$  in device performance and demonstrated that sufficiently large  $R_p$  values are pivotal to optimized device performance under low light intensity [14].

The objective of this study is to optimize the performance of OPVs with P3HT-ICBA as the photoactive layer under indoor lighting conditions. The optimization was accomplished by modifying the surface of the ITO layer with a polymer like ethoxylated polyethylenimine (PEIE) that contains simple aliphatic amine groups, which is an efficient and universal method to reduce the work function (WF) of conductors to realize efficient electron-collecting electrodes (ECEs) [15–18]. Lechêne et al. recently showed the effect of polymer-based surface modification on the indoor performance of OPVs [1]. However, the so-called conventional geometry was employed in their research, in which reactive metals were incorporated at the top of a device, which is not a favorable configuration because of the potential to increase air instability. Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT-PSS) was also used as a hole-collecting interlayer in their devices, and the composite can cause chemical instability because of its acidic nature [19]. Thus, without using reactive or acidic components, the so-called inverted geometry was adopted as a more suitable geometry in this research [15]. To optimize the effects of  $R_p$  on the performance, the thickness of the PEIE layer was controlled by varying the concentration of PEIE (0.05, 0.1, 0.4, 0.6, 0.8, 1.2, and 1.6 wt%) in 2-methoxyethanol during the preparation process. To study the light-source-dependent performance, three different light sources (an LED lamp, a fluorescent lamp, and a halogen lamp) were utilized. The surface and optical properties of the PEIE layers were examined by atomic force microscopy (AFM) and ultraviolet–visible (UV–vis) spectrometry. Furthermore, the dependence of the optical density on the thickness of PEIE on the photoactive layer was simulated using the finite-difference time-domain (FDTD) method.

## 2. Experimental section

### 2.1. Preparation of precursor solutions

Precursor solutions of the organic films used in this study were prepared as follows: PEIE (80% ethoxylated,  $M_w \approx 70,000 \text{ g/mol}$ , 35–40 wt% in water, Aldrich, St. Louis, MO, USA) was diluted with 2-methoxyethanol (Aldrich, St. Louis, MO, USA) to concentrations of 0.05, 0.1, 0.4, 0.6, 0.8, 1.2, and 1.6 wt% and stirred overnight in ambient air. A solution of P3HT (4002E, Rieke Metals, Lincoln, NE, USA) and ICBA (Luminescence Technology Corp., New Taipei City, Taiwan) was prepared in 1,2-dichlorobenzene (DCB; Aldrich, St. Louis, MO, USA) to obtain a P3HT/ICBA weight ratio of 1:1 and a total P3HT-ICBA concentration of 40 mg/mL. The P3HT-ICBA solution was stirred at a temperature of 70 °C overnight in a nitrogen-filled glove box before use.

### 2.2. Device fabrication and characterization

Pre-patterned ITO-coated glass (OMNI.0782, Omniscience Korea, Suwon, Republic of Korea) with a sheet resistivity of  $10 \Omega/\text{sq}$  was used as the substrate material. The substrates were cleaned in an ultrasonic bath using detergent (Liqui-Nox® Phosphate-Free Liquid Detergent, Alconox, Inc., White Plains, NY, USA) and water, rinsed with deionized (DI) water, and then cleaned in sequential ultrasonic baths of DI water, acetone, and 2-propanol. Nitrogen was used to dry these substrates after each bath. Next, after filtering each PEIE solution through a polytetrafluoroethylene (PTFE) filter with pore size of 0.2  $\mu\text{m}$ , PEIE layers with thickness of 1.7 nm (0.05 wt%), 2.2 nm (0.1 wt%), 5.6 nm (0.4 wt%), 8.5 nm (0.6 wt%), 11.3 nm (0.8 wt%), 17.6 nm (1.2 wt%), and 22.8 nm (1.6 wt%) were spin cast onto the cleaned and dried substrates in air at a speed of 5000 rpm for 1 min. These PEIE-cast substrates were dried in air on a hotplate at a temperature of 110 °C for 10 min. The PEIE-coated substrates were then loaded into a nitrogen-filled glove box. After the P3HT-ICBA precursor solution was filtered through a PTFE filter with pore size of 0.2  $\mu\text{m}$ , films of P3HT-ICBA with thickness of around 180 nm were deposited on top of the PEIE-coated substrates by spin coating at 700 rpm for 30 s. They were then annealed at 150 °C for 10 min under a nitrogen atmosphere. The thickness of these PEIE layers was measured with a surface plasmon resonance spectrometer system (RT2005 SPR, Resonant Technologies GmbH, Framersheim, Germany) under illumination by p-polarized laser light (He-Ne, 632.8 nm, 10 mW). The thickness of the P3HT-ICBA layer was

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