

# Inverted-structure polymer solar cells fabricated by sequential spraying of electron-transport and photoactive layers



Hye-Yun Park<sup>a</sup>, Dongchan Lim<sup>b</sup>, Seung-Hwan Oh<sup>c</sup>, Phil-Hyun Kang<sup>c</sup>, Giseop Kwak<sup>d,\*</sup>,  
Sung-Yeon Jang<sup>a,\*</sup>

<sup>a</sup> Department of Chemistry, Kookmin University, 861-1, Jeongneung-Dong, Seongbuk-Gu, Seoul 136-702, Republic of Korea

<sup>b</sup> Surface Technology Division, Korea Institute of Materials Science, Changwon 641-010, Republic of Korea

<sup>c</sup> Radiation Research Division for Industry and Environment, Korea Atomic Energy Research Institute (KAERI), 29 Geungmu-gil, Jeongeup-si, Jeollabuk-do 580-185, Republic of Korea

<sup>d</sup> School of Applied Chemical Engineering, Major in Polymer Science and Engineering, Kyungpook National University 1370 Sankyuk-dong, Buk-ku, Daegu 702-701, Republic of Korea

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## ABSTRACT

Inverted-structure polymer solar cells (I-PSCs) containing sequentially sprayed electron-transporting layers (ETLs) and photoactive layers were fabricated. Low-temperature sol-gel-derived ZnO thin films were used as the ETLs and films of a poly(3-hexylthiophene) (P3HT)/[6,6]-phenyl-C61-butyric acid methyl ester (PCBM) blend were used as the photoactive layers. Nanoripples-containing ZnO ETLs could be successfully fabricated by controlling the spraying rate of the ZnO precursor solution and the subsequent annealing conditions. The P3HT/PCBM active layers sprayed on the ZnO ETLs were optimized using a unique solvent-assisted post-deposition treatment, namely, the sprayed solvent overlay (SSO) treatment. The power conversion efficiency (PCE) of the I-PSCs based on the optimized ETLs and active layers was as high as 3.55%, which is comparable to that reported for I-PSCs fabricated using the conventional spin-coating method. The sprayed I-PSCs also exhibited high environmental stability, maintaining ~80% of their PCE even after 40 days of aging in air under ambient conditions without encapsulation. The I-PSCs based on the P3HT/PCBM photoactive layers optimized using the SSO treatment displayed much higher stability than those based on photoactive layers optimized using a conventional thermal annealing treatment. This result indicated that the SSO treatment is a suitable post-deposition treatment method for improving the morphological stability of P3HT/PCBM active layers. Further, the fabrication technique investigated in this study is a high-throughput low-temperature one and is suitable for fabricating high-stability PSCs.

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## 1. Introduction

Polymer-based solar cells (PSCs) are being considered as the next-generation solar cells, and their development and

commercialization should lead to the realization of low-cost, disposable, and flexible energy devices. Improving the power conversion efficiency (PCE) of PSCs has been a major research goal. The PCE of such devices has been improved continuously, to up to ~9%, through the design of novel semiconducting materials such as low-band-gap polymers and fullerene derivatives [1]. The development of devices with PCE values as high as this have made it feasible to consider the commercialization of PSCs for various

\* Corresponding authors. Tel.: +82 (53) 950 7558; fax: +82 (53) 950 6623 (G. Kwak). Tel.: +82 (2) 910 5768; fax: +82 (2) 910 4415 (S.-Y. Jang).

E-mail addresses: [kwak@knu.ac.kr](mailto:kwak@knu.ac.kr) (G. Kwak), [syjang@kookmin.ac.kr](mailto:syjang@kookmin.ac.kr) (S.-Y. Jang).

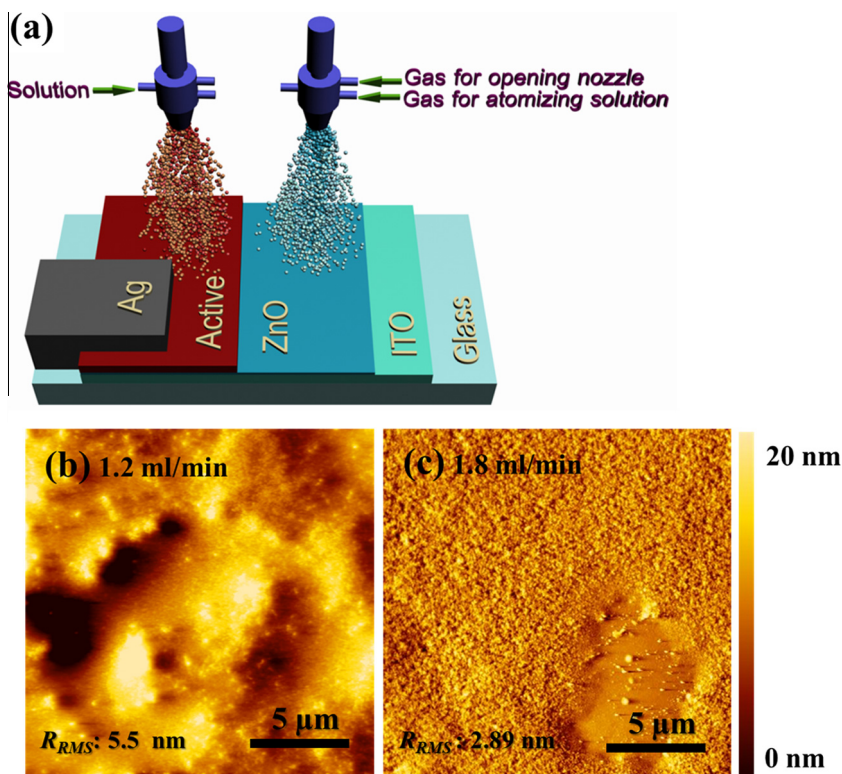
large-scale applications. However, in spite of these improvements, there remain a few obstacles to be overcome. These are the poor long-term stability of PSCs in ambient conditions and the infeasibility of fabricating high-PCE PSCs using industrial manufacturing techniques [2,3].

Conventional PSCs generally have a structure in which a hole-transporting layer (HTL), usually poly(3,4-ethylenedioxythiophene):poly(styrene sulfonic acid) (PEDOT:PSS), is coated on a layer of indium-doped tin oxide (ITO), and a low-work-function metal such as Al is used as a cathode. These normal PSCs are known to have low environmental stability because of the oxidation of the low-work-function cathode (Al) and the degradation of ITO by the acidic PEDOT:PSS HTL [4–7]. One of the simple routes to enhance the stability is to invert the charge collection direction, in which an electron transporting layer (ETL), regularly wide band-gap metal oxides, was deposited on ITO and a relatively high-work-function metal such as Ag or Au were used as anodes. This type of cells, known as inverted structured PSCs (I-PSCs) could considerably improve the environmental stability of conventional PSCs [8,9].

While the spin-coating method has been used almost exclusively as the fabrication method of choice for PSCs, other methods that are compatible with high throughputs and continuous processing have also been suggested [10]. These include screen printing [11,12], inkjet printing [13], and spray-based methods [14–19]. In most studies on these techniques, the polymer active layers were deposited using a number of different methods. The performances of the devices based on the thus-deposited layers were then

compared with those of devices based on spin-coated layers, as the performance of PSCs is determined primarily by the characteristics of the active layer. Controlling the morphology of the active layer at macroscopic level, along with the internal nanomorphology of the layer, by choosing the appropriate solvents, controlling the concentrations of the solutions used, and adjusting the solvent evaporation kinetics have been areas of intensive focus. Further, most of these studies have investigated conventional-structured PSCs alone. In order to allow for the commercialization of PSCs, the development of high-throughput processing techniques for fabricating I-PSCs is essential. In I-PSCs, the charge-selective layer between the active layer and the electrodes plays a crucial role in determining the open-circuit voltage ( $V_{OC}$ ) and the fill factor (FF) of the device. The ability to fabricate these layers using different deposition methods is a prerequisite. Given that the purported flexibility of PSCs is one of their most attractive qualities, the ability to fabricate I-PSCs using a high-throughput process at a low temperature is therefore of great importance.

In this study, we fabricated I-PSCs in which the ETL and the photoactive layer were fabricated by a spray-based method. Sol-gel-derived ZnO thin films were used as the ETLs, and films of a poly(3-hexylthiophene) (P3HT)/[6,6]-phenyl-C61-butyric acid methyl ester (PCBM) blend were used as the photoactive layers. The two types of layers were deposited using a gas-assisted spraying technique at relatively low temperatures. The embedded ZnO ETLs, whose surfaces contained nanoripples, could be deposited



**Fig. 1.** (a) Schematic showing the fabrication of I-PSCs by the spraying method. AFM images of the as-sprayed precursor films deposited using different spraying rates: (b)  $1.2 \text{ ml min}^{-1}$  and (c)  $1.8 \text{ ml min}^{-1}$ . The root mean squares roughness ( $R_{RMS}$ ) values of the films were 5.5 nm and 2.89 nm, respectively.

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