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journal homepage: www.elsevier.com/locate/jiec1 Surface modification of line-patterned electron transfer layer for
2 enhancing the performance of organic solar cells

3 Q1 Seung E. Jung, Eui J. Lee, Doo K. Moon*, Jung R. Haw*

4 Department of Materials Chemistry and Engineering, Konkuk University, 120, Neungdong-ro, Gwangjin-gu, Seoul 05029, Republic of Korea

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ABSTRACT

In this study, we fabricated inverted organic solar cells (i-OSCs) using line-patterned interlayer for electron extraction. The short circuit current density (J_{SC}) of line-patterned interlayer device increased drastically, while the fill factor (FF) decreased rather than flat-type interlayer device. So, an additional flat-type Zinc oxide (ZnO) layer with 1% vol. ethylene glycol (EG) added as an additive between the line-patterned ZnO layer and the ITO electrode was fabricated. The addition of EG in ZnO precursor has reported that the oxygen molecules in EG passivate the surface defects of ZnO and alters its electron affinity of ZnO. As a result, the power conversion efficiency (PCE) of the line-patterned devices introduced an additional flat-type ZnO layer and a ZnO precursor with 1% vol. EG was enhanced from 6.5% to 7.6%.

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5 Introduction

6 Organic solar cells (OSCs) based on conjugated polymers offer
7 many advantages including low cost, transparency, light weight,
8 and flexibility. They have tremendous attention as next generation
9 renewable power sources for portable and wearable devices such
10 as tablets, laptops, smart watches, and smart phones [1,2]. Several
11 studies have recently been published on OSCs with power
12 conversion efficiencies (PCE) of 10% or more [3]. However, the
13 commercialization potential of OSCs remains questionable, owing
14 to their low efficiency. Hence, a number of studies have been
15 carried out to improve the efficiency of OSCs [4] by using materials
16 with superior photon harvesting [5–7], optimization of phase
17 segregation through additives [8–10], and improving the charge
18 carrier collection through interfacial modifications [2,11,12].

19 In OSCs, a bulk heterojunction consisting of acceptor and donor
20 polymers is used as the photoactive layer. This layer is positioned
21 between an indium tin oxide (ITO)/poly(3,4-ethylenedioxythio-
22 phene:polystyrene sulfonate) (PEDOT:PSS) anode and a low work
23 function metal such as Ba, Al, or Ca [13]. PEDOT:PSS is a transparent
24 electrode material with exceptional electrical and optical proper-
25 ties. However, the acidic and hygroscopic properties of PEDOT:PSS
26 are known to reduce the lifetime of a device [14,15]. This problem

can be addressed by using an inverted structure. Kim et al.
proposed that the hole collection efficiency of OSCs can be
increased by using high work function metals such as Au or Ag and
that the device stability can be improved by using air stable metal
oxides such as ZnO, TiO_x, or ZMO [16].

Among the aforementioned metal oxide candidates for OSCs,
ZnO stands out owing to its unique properties such as optical
transparency, high electron mobility, and environmental stability.
In addition, its solution processability allows an easy morphology
control. Owing to its aforementioned characteristics, ZnO is often
used as an electron transfer layer (ETL) [13,17,18]. However,
calcined ZnO suffers from nanoripples on its surface and charge
trapping. This affects the energy band alignment of devices
adversely [19]. To eliminate these defects, various methods such as
the use of additives or the fabrication of nanostructured ZnO are
employed. Poly(ethylene glycol) (PEG) and poly(ethylene oxide)
(PEO) are the commonly used additives for controlling the
morphology of ZnO. Jo et al. have reported that the addition of
PEG to ZnO alters its electron affinity. This is because the oxygen
molecules in PEG passivate the surface defects of ZnO, and hence
reduce its domain boundaries [20]. Sui et al. and Shao et al.
obtained similar results by using PEO as the additive [21,22].
However, their results also showed that as the amount of PEO
increases, the performance of OSCs decreases [22].

Fabricating a nanostructure in the buffer layer increases the
optical absorption property. As light trapping of an active layer is
related to its thickness and morphology, controlling the thickness
of the active layer can improve its light absorption property [10].

* Corresponding authors. Fax: +82 2 444 0765.

E-mail addresses: dkmoon@konkuk.ac.kr (D.K. Moon), jrhaw@konkuk.ac.kr (J.R. Haw).

55 Additionally, a functional microprism can be implemented on the
56 top of the glass to further increase the light trapping, which in turn,
57 decreases light scattering [12]. Hu et al. used nanoimprinting
58 lithography on a donor polymer to create numerous devices
59 depending on the pattern size. They found that by decreasing the
60 pattern size, the current density, J_{SC} , can be enhanced since the
61 light absorbance of long-wavelength light increases as the pattern
62 size decreases. However, the Si mold used by them was costly and
63 the process was complicated as the acceptor was thermally
64 evaporated [10]. Jeong et al. and Genevet et al. achieved improved
65 crystallization, chain alignment, and phase separation by imple-
66 menting nanostructures in photoactive films [2,23].

67 In the present study, an ordinary digital versatile disc (DVD)
68 was used as the imprinting mold instead of the aforementioned Si
69 mold. To overcome the defects due to imprinting and to enhance
70 the fill factor (FF), a patterned ETL was fabricated on a flat ETL. We
71 used a ZnO ETL since its morphology can be tuned easily, which is
72 favorable for the formation of nanostructures. Ethylene glycol (EG)
73 was added to the flat ETL as an additive for reducing its work
74 function and charge recombination. All this altered the surface
75 electron affinity of the ETL, which in turn, enhanced the energy
76 alignment of the device. As a result, the PCE of the patterned
77 devices increased by approximately 14%.

78 Experimental section

79 Preparation of ZnO precursor solution

80 The ZnO solution was synthesized according to previously
81 published procedures [24]. A gram of zinc acetate dehydrate
82 ($C_4H_6O_4Zn \cdot 2(H_2O)$ 99.5%, Aldrich) and 0.28 g of monoethanol-
83 amine ($HOCH_2CH_2NH_2$, 98%, Aldrich, 1:1 mol ratio) were dissolved
84 into 10 ml of 2-methoxyethanol ($CH_3OC_2H_4OH$, 99.8%, Aldrich) and
85 hydrolyzed for 12 h. The ethylene glycol (EG, $C_2H_4(OH)_2$ 99.8%
86 Aldrich) modified ZnO thin film (ZnO-EG) was prepared by adding
87 1 vol.% of EG into the 1 ml of ZnO solution.

Preparation of PDMS stamp

88 To form a nanostructure on ETLs, Polydimethylsiloxane (PDMS)
89 stamp was used. The Shin-Etsu KR-106 and CAT-RG were mixed
90 with a ratio of 10:1 (w/w) [25]. The mixture was stored in vacuum
91 chamber for 30 min to deforming process. After deforming, the
92 mixture was poured on the mold, which was prepared from DVD
93 discs, and annealed at 80 °C for 1 h.
94

Fabrication of inverted OSCs

95 To fabricate inverted OSCs, patterned ITO glasses were washed
96 by ultrasonicator with acetone, detergent, 2-propanol and
97 deionized water in sequence. After cleaning, the ITO glasses were
98 baked at 120 °C and UV-ozone-treated for 15 min. The ZnO and
99 ZnO-EG solutions were separately spin-coated onto different ITO
100 glasses and annealed at 150 °C for 1 h in air to obtain an electronic
101 transport layer (ETL, ~10 nm). For the nanostructured devices,
102 after the solutions separately were spin coated on the ITO or ETLs,
103 the PDMS stamp was placed on the surface of the films and
104 pressure of 0.6 MPa was applied on PDMS stamp with 60 °C for
105 5 min. After imprinting process, the PDMS stamp was peeled off
106 and nanostructured ETLs were annealed at 150 °C for 1 h in air. The
107 prepared samples were then transferred to a N_2 -filled glove box,
108 and a solution of polythieno[3,4-b]thiophene-co-benzodithio-
109 phene (PTB7):[6,6]-phenyl C_{71} butyric acid methyl ester (PC₇₁BM)
110 (1:1.5, w/w) in chlorobenzene/1,8-diodooctane (97:3, v/v) was
111 spin-coated to form the photoactive layer (80 nm). Finally, MoO₃
112 (5 nm) and Ag (100 nm) were sequentially deposited by thermal
113 evaporation in a high-vacuum chamber ($<10^{-7}$ Torr). The active
114 area of the fabricated devices was 12 or 7 mm².
115

Measurement

116 The current density–voltage (J – V) characteristics of the
117 fabricated inverted OSCs were assessed using a Keithley
118

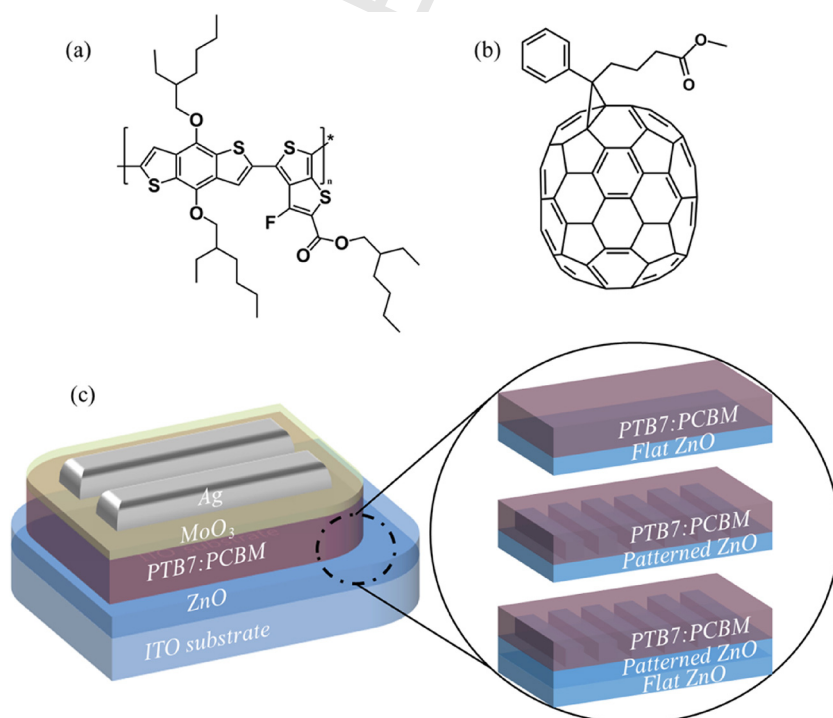


Fig. 1. Molecular structure of (a) PTB7 and (b) PC₇₁BM, and (c) device structure of inverted OSCs.

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