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Solution-processed small molecular photocells with neat fullerene

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ABSTRACT

Because of ease in ensuring chemical integrity and purification, small molecular materials are promising as alternatives for polymeric electron-donor materials in solution-processed bulk heterojunction (BHJ) photocells. On the other hand, replacement of modified fullerene with neat one potentially reduces both economical and environmental costs of the photocells. In this paper, solution-processed BHJ photocells with a small molecular electron-donor 7,7'-[4,4-bis(2-ethylhexyl)-4*H*-silolo [3,2-*b*:4,5-*b'*]dithiophene-2,6-diyl]bis[6-fluoro-4-(5'-hexyl-[2,2'-bithiophen]-5-yl)benzo[*c*][1,2,5]thiadiazole] (*p*-DTS(FBTTh₂)₂) and neat C₇₀ are studied. The devices showed a power conversion efficiency of approximately 1.3% after optimal thermal annealing. Analysis of current–voltage characteristics using an equivalent circuit model suggests that the thermal annealing enhances not only the generation but also the internal recombination of photocarriers.

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1. Introduction

Solution-processed organic photocells have been attracting much attention of researchers, partially because of their compatibility with printing technology, which is thought to be one of the cheapest technologies. It is expected that photocells produced like sheets of news paper will be available at significantly lower cost than present Si-based photocells.

Among various material systems for solution-processed photocells proposed so far, the bulk heterojunction (BHJ) composites based on conjugated polymers and fullerenes are the most widely studied systems [1–3]. The findings of efficient and ultrafast electron transfer in conjugated polymers containing small amount of neat fullerene C₆₀ initiated the research area [4–6].

However, the solution-processed organic photocells based on neat fullerenes had been rarely studied, because suitable solvents for the preparation of BHJ composites, which consist of comparable amounts of the both components yielding transport paths for the electrons and holes to reach the electrodes, using neat fullerenes had not been widely known to the researchers in this area. Thus, chemically modified fullerenes such as [6,6]-phenyl-C₇₁-butyric acid methyl ester (C₇₀-PCBM) with high solubility play a major role in this area [7].

Nevertheless, the utilization of neat fullerene is attractive from both economical and environmental viewpoints, since all fullerene derivatives including C₇₀-PCBM developed so far are synthesized

from a relevant neat fullerene. Neat fullerenes have been routinely used in the dry-processed organic photocells [8–10]. It has been reported that the utilization of some not-so-common solvents such as 1,2,4-trimethylbenzene (TMB, also known as pseudocumene) make it possible to realize solution-processed BHJ photocells based on neat fullerenes [11,12]. The combination of neat fullerenes and non-halogenated solvents such as TMB is expected to offer a pathway to develop environment-friendly and low-cost photocells.

In addition, since the electron mobility of neat fullerenes is higher than their modified versions such as C₇₀-PCBM, the photocells with neat fullerenes potentially show superior performance compared with those using the modified fullerenes [13,14]. Although some researchers have addressed the development of photocells using neat fullerenes, the performance of such photocells are generally lower than those with the fullerene derivatives [11,12,15]. One of the probable reasons for this is the fact that the energy level of the lowest unoccupied molecular orbital (LUMO) of the neat fullerenes are slightly lower than that of the modified fullerenes, yielding a reduced open-circuit voltage [16]. This drawback will be overcome by the development of the electron-donor materials whose energy level of the highest occupied molecular orbital (HOMO) is tailored to the LUMO energy level of the neat fullerenes. The neat fullerenes are considered as the first choice for electron-acceptor materials in vacuum-processed organic photovoltaic devices [17–19].

Although the polymer materials show superior film-forming properties, they have inherently drawbacks regarding chemical integrity and purity of material compared with small molecular materials. That is, every small molecular material has a single and

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distinct molecular structure. Furthermore, the purity of a small molecular material can be very high after the purification by sublimation [20]. Contrarily, it is practically impossible to obtain a polymer material with a single chain length, and polymers are not compatible with purification by the sublimation technique. It has been recognized that purity of the materials has a critical impact on the device performance [21,22]. The neat fullerenes are attractive also from this aspect, since neat fullerenes with ultra-high purity have been realized [17].

Recently, small molecular electron-donor materials compatible with solution-processes have been developed [23–25]. Among them, 7,7'-[4,4-bis(2-ethylhexyl)-4*H*-silolo[3,2-*b*:4,5-*b'*]dithiophene-2,6-diyl]bis[6-fluoro-4-(5'-hexyl-[2,2'-bithiophen]-5-yl)benzo[*c*][1,2,5]thiadiazole] (*p*-DTS(FBTTh₂)₂) is one of the most successful molecules [25,26]. Besides C₇₀-PCBM with which PCE over 8% has been achieved [27], various non-traditional electron-acceptor materials such as copper indium nanocrystals [28] and a perylene diimide derivative [29] have been investigated as a counterpart of *p*-DTS(FBTTh₂)₂. However, to the best of the author's knowledge, the BHJ composites of these materials combined with neat fullerenes have never been reported so far. Here, preliminary results on the photocells with a BHJ composite consisting of *p*-DTS(FBTTh₂)₂ and neat C₇₀ are reported.

2. Materials and methods

p-DTS(FBTTh₂)₂ and C₇₀ were purchased from Aldrich and Nano-C, respectively. TMB used was the product of Tokyo Kasei. The molecular structures of *p*-DTS(FBTTh₂)₂ and C₇₀ as well as the schematic device structure of photocells are indicated in Fig. 1(a). The energy level diagram of the *p*-DTS(FBTTh₂)₂:C₇₀ BHJ photocells is shown in Fig. 1(b) [27,30]. A solution of *p*-DTS(FBTTh₂)₂:C₇₀ composite (1:1 by weight) was prepared by dissolving 15 g l⁻¹ of both materials in TMB. Although the solubility of C₇₀ in TMB has not been determined, it has been experienced that no serious precipitation was observed in a TMB solution containing 23 g l⁻¹ of C₇₀. Films of poly(3,4-dioxythiophene):poly(styrene sulfonate) salt (PEDOT) were deposited on glass plates by spin-coating from an aqueous suspension purchased from Aldrich, followed by annealing at 130 °C for 60 min. The thickness of the PEDOT films was approximately 50 nm. The films of *p*-DTS(FBTTh₂)₂:C₇₀ BHJ composite were deposited on PEDOT-coated substrates by spin-coating. The films with approximately 50 nm in thickness coated from the solution mentioned above at a spin-rate of 1200 rpm were used throughout this study. It is important to keep the solution at high temperature by placing a bottle of the solution on a hot-plate heated at 90 °C to obtain homogeneous films. Serious precipitation was observed in the solution in a bottle on a hot-plate at 50 °C. It was experienced that the PEDOT underlayer significantly improves the uniformity of the *p*-DTS(FBTTh₂)₂:C₇₀ BHJ composite films, as shown in Fig. 2.

Glass plates with a prepatterned indium-tin-oxide (ITO) electrode were used for the photocells. After the spin-coating of the composite films, Al cathodes were vacuum deposited to prepare the devices with 3 mm × 3 mm square active area, followed by a post-production thermal annealing at a designated temperature within the range of 70 to 160 °C for 20 min. Finally, the active area of the photocell was covered by a glass plate and a UV-curable resin in order to moderate prompt degradation. The preparation of the devices was carried out in a glove-box filled with nitrogen.

The optical absorption spectra of the composite films deposited on PEDOT-coated glass plates were recorded using a Shimadzu UV-1800 spectrophotometer. The current-voltage characteristics of photocells were measured using a Keithley 6517A electrometer under AM 1.5 G 100 mW cm⁻² (1 sun) illumination from an Asahi

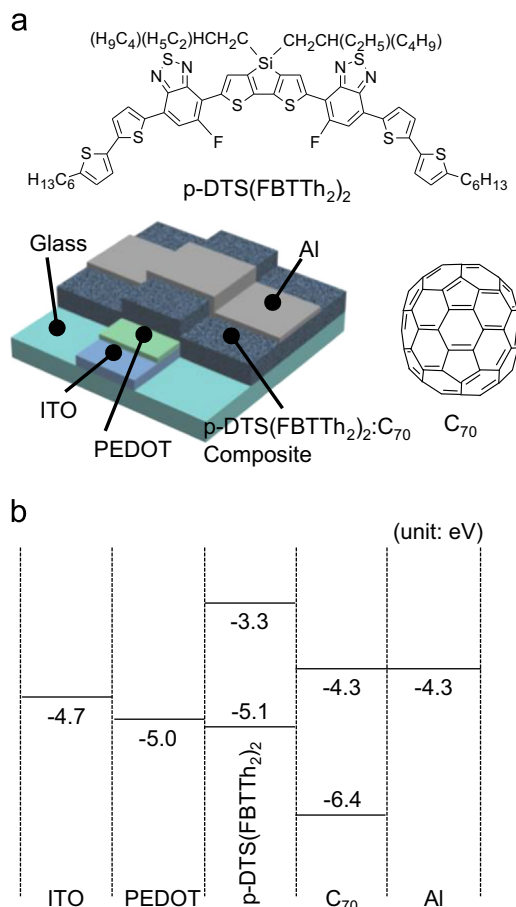


Fig. 1. (a) Schematic device structure with molecular structures of *p*-DTS(FBTTh₂)₂ and C₇₀. (b) Energy level diagram of the *p*-DTS(FBTTh₂)₂:C₇₀ BHJ photocells.



Fig. 2. Photograph of *p*-DTS(FBTTh₂)₂:C₇₀=1:1 BHJ composite films coated on a bare glass plate (left) and a PEDOT-coated glass plate (right). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Spectra HAL-C100 solar simulator. The Solver add-in program of Microsoft Excel 2007 was employed for fitting the current-voltage characteristics with an equivalent circuit model to extract the circuit parameters. Short-circuit photocurrent spectra were recorded by irradiating light from a 300 W Xe-lamp passing through a monochromator as well as appropriate long-pass glass filters. The short-circuit photocurrent spectra were converted to external quantum efficiency (EQE) spectra by using a short-circuit photocurrent

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