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Letter

Improvement of the efficiency of CuPc/C_{60}-based photovoltaic cells using a multistepped structure

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ABSTRACT

Organic photovoltaic cells with multistepped (MS) structures based on a CuPc/C_{60} materials system were fabricated. A detailed analysis of the effect of stepped distributions on the device's performance suggests that the highest efficiency is reached when the concentration gradient of donor and acceptor is largest. In this kind of device, owing to a great chemical potential energy gradient in addition to a large charge-separation (CS) interface, both the open-circuit voltage and short-circuit current density are improved remarkably, leading to a power conversion efficiency nearly one level higher than that of the bilayer device and 50% greater than the 1:1 homogeneously mixed device.

In 1986, Tang first reported on the bilayer heterojunction organic solar cells based on the Cu-phthalocyanine (CuPc)/perylene tetracarboxylic derivative (PV) [1]. In 1992, Sacriciftci et al. found that C_{60} can be a good electron acceptor material [2]. Soon after, these organic solar cells, which adopted C_{60} and its derivatives as an electron acceptor, became widespread [3–5].

In a P/N heterojunction bilayer device based on organic small-molecule dyes, only photogenerated excitons at the P/N interface will produce charges through separation. The comparatively great chemical potential energy gradient ($\nabla\mu_{hv}$) in this structure could also effectively impel electrons and holes towards opposite directions [6]. However, only the excitons within the range of the exciton diffusion length (L_D) can contribute to the cell photocurrent in this structure [4]. In the single layer device with a bulk heterojunction structure based on a mixture of polymer with fullerene and its derivatives, due to the D–A interpenetrating networks morphology that formed by phase separation, the efficiencies of exciton dissociation have been raised [6–10]. However, because the charges are evenly distributed on the whole photoactive layer, $\nabla\mu_{hv}$ is relatively small [6]. In addition, the recombination probability of photogenerated carriers becomes much higher than that of the bilayer P/N heterojunction [8–11].

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Great efforts have been made to address this problem by optimizing thin-film morphology to improve the charge transport and suppress recombination [12–15]. Moreover, the bulk heterojunction structure based on organic small-molecule dyes could be formed through donor–acceptor (D–A) co-deposition [16–19]. However, the carrier recombination rate in such type of mixed layer remains at a very high level [20].

In this paper, a multistepped (MS) structure of a device made from a CuPc:C_{60} heterojunction materials system is introduced. Through simultaneous dynamic alteration of the relative deposition rates of the two kinds of materials, the C_{60} content in the device ascends from the anode to the cathode, while that of CuPc descends from the anode to the cathode, which seems similar to the graded junction solar cell design for inorganic solar cells [22–24]. However, in organic solar cells, only Sullivan et al. [17], Drees et al. [13], and Koeppe et al. [5] have made similar structures by other methods. Sullivan et al. produced a five-layered device based on the CuPc/C_{60} materials system where the CuPc:C_{60} composition varies from purely donor to purely acceptor via three mixed layers of increasing acceptor concentration, which are 25%, 50%, and 75% [17]. Drees et al. propelled the interdiffusion of an initial MEH-PPV/C_{60} bilayer by simple heat treatment to obtain a bulk heterojunction with a concentration gradient of donor and acceptor [13]. Koeppe et al. used a pyrollidinofullerene compound with chelating pyridyl groups (PyF) that can interact with the Zn atom in the Zn-phthalocyanine (ZnPc) molecule in the bilayer solar cell, which may also induce a similar molecular architecture [5]. These devices all exhibit good photovoltaic performance, but its causes were discussed from the aspect of mixed film morphology only. Based on this concept, we envisioned MS device as a different approach to obtain a high photovoltaic performance. Thus, the effect of different sizes of concentration gradients of donor and acceptor on the solar cell performance is further discussed. Also, the optimization mechanism based on the theory of excitonic solar cells [6] is investigated to better understand the potential advantages of both bilayer heterojunction cell and homogeneously mixed cell in this structure.

Several devices with three different structures were designed for comparison (Fig. 1 inset). These are as follows:

- A: ITO/CuPc (30 nm)/C_{60} (30 nm)/TPBI (6 nm)/Al (80 nm);
- B: ITO/CuPc:C_{60} (MS, 60 nm)/TPBI (6 nm)/Al (80 nm); and
- C: ITO/CuPc:C_{60} (1:1,60 nm)/TPBI (6 nm)/Al (80 nm).

Among them, A is the bilayer heterojunction device (standard device), B is the MS device, and C is the 1:1 homogeneously mixed device. The CuPc:C_{60} (1:1) homogeneously mixed device is chosen owing to the fact that the total CuPc content is the same as that of C_{60} in the MS layers, and could thus be used to compare with these MS devices as the extreme case without the gradient. The 2,2,2-(1,3,5-benzenetriyl)tris-[1-phenyl-1H-benzimidazole] (TPBI) layer in each device acts as an exciton blocking layer.

Glass/indium tin oxide (ITO) substrate with a sheet resistance of 20 Ω/sq was cleaned in an ultrasonic bath using detergent water, de-ionized water, acetone, and ethanol in

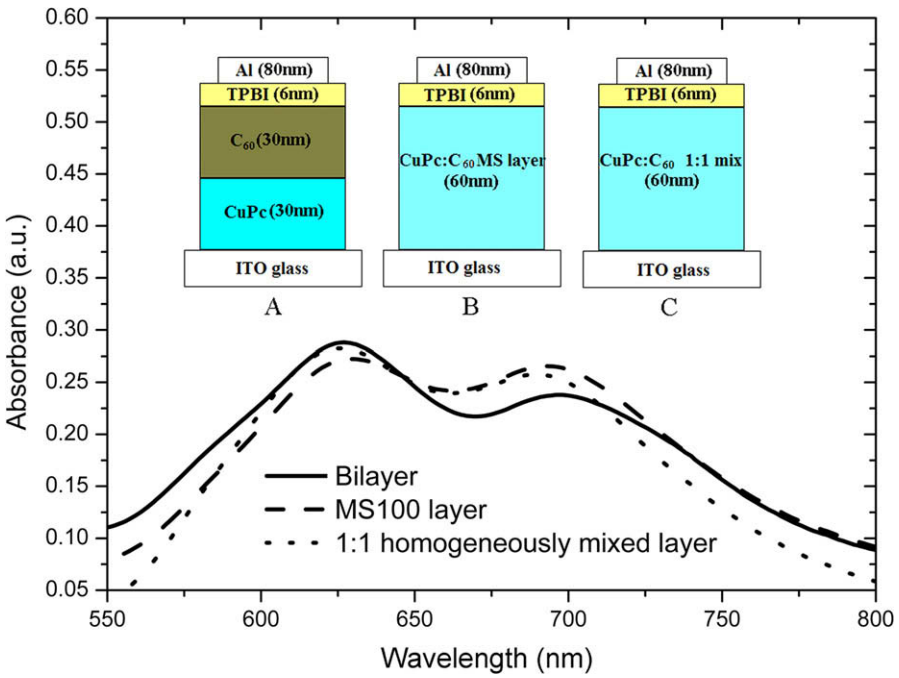

Fig. 1. Absorption spectra of CuPc/C_{60} bilayer, MS100 layer, and 1:1 homogeneously mixed layer deposited on quartz substrates. Inset: schematic of the PV devices investigated. A is the bilayer heterojunction device (standard device); B is the MS device; and C is the 1:1 homogeneously mixed device.