Organic Electronics 15 (2014) 139–143

Contents lists available at ScienceDirect

Organic Electronics

journal homepage: www.elsevier.com/locate/orgel

Letter

Solvent-dependent morphology of thermally converted copper phthalocyanine for solution-processed small molecule organic photovoltaic devices

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ARTICLE INFO

Article history:
Received 31 July 2013
Received in revised form 26 September 2013
Accepted 18 October 2013
Available online 6 November 2013

Keywords:
Solar cells
Solution process
Copper phthalocyanine
Thermal conversion
Thermocleavable

ABSTRACT

We fabricated copper phthalocyanine (CuPc)-based organic photovoltaic (OPV) devices *via* a solution process. CuPc was obtained through thermal conversion of its precursor $CuPc(OMe)_2$, which has excellent solubility in various organic solvents, on a substrate. Solvent-dependent performance of the resulting devices was observed, which could be explained by considering film morphology. Using a 1:2 (wt/wt) mixture of chlorobenzene and chloroform produced a p–n type OPV device with a power conversion efficiency of 1.35% under 1 sun simulated AM1.5G solar illumination.

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1. Introduction

Over the past two decades, organic photovoltaics (OPVs) have received a tremendous amount of attention because of their potential application as green energy devices [1,2]. OPV devices are often categorized into polymer and small-molecule OPVs according to their constituent materials. Polymer-based OPVs have been intensely studied because of their good film quality, high fill factor (FF) and comparatively high power conversion efficiency (PCE). Compared with polymer-based devices, small-molecule OPVs have some important advantages, including structural versatility, and high charge mobility and open circuit voltage (V_{OC}) [3–5]. Recently, small-molecule OPVs with PCEs of >8% [5] have been obtained through careful molecular design and device optimization, revealing the possibility of commercialization of OPVs based on small molecules.

Tang published a pioneering work in the OPV research field [6]. He studied small-molecule OPVs with a two-layer structure containing copper phthalocyanine (CuPc). Devices based on CuPc have been studied continuously since because of the low cost, chemical and physical stability, and outstanding photophysical and electrochemical properties of CuPc. Accordingly, CuPc plays an important role not only in the colorant industry, but also in high-tech products such as optical disks, photodynamic therapy, and organic semiconductors [7]. With regard to OPV research, over the past decade, a PCE of 5.7 ± 0.3% [8] has been achieved for an OPV containing CuPc under 1 sun simulated AM1.5G solar illumination. However, thermal evaporation in a high-vacuum chamber under low pressure or a particular structure design was necessary to deposit the organic layers [9–15]. Such rigorous deposition conditions have limited the application of CuPc in OPVs. In addition, in this process, the most-used commercial acceptor materials [6,6]-phenyl-C_{61}-butyric acid methyl ester ($PC_{61}BM$) and [6,6]-phenyl-C_{71}-butyric acid methyl ester ($PC_{71}BM$) are not compatible with device fabrication, even though they show high electron mobility and good

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1566-1199/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.orgel.2013.10.023

solubility and miscibility [16–20]. Therefore, a solution process [21–23] to fabricate OPVs is preferred for academic study and industrial application.

CuPc is barely soluble in common solvents such as water, alcohols, acetone, and hydrocarbons. This solubility problem was successfully overcome with a thermal conversion concept [6,24–27]. Based on this approach, we fabricate CuPc devices using a solution process. Herein, we produce a p–n type OPV device with PCE = 1.35% under 1 sun simulated AM1.5G solar illumination using the CuPc derivative $CuPc(OMe)_2$ as an electron donor precursor and a mixture of PCBMs as electron acceptors. The morphology of the donor film after thermal conversion from $CuPc(OMe)_2$ to CuPc using various organic solvents is also investigated to elucidate the dependence of solvent on device performance.

2. Experimental

2.1. Reagents

Mix-PCBM (mixture of $PC_{61}BM$ and $PC_{71}BM$) and $PC_{71}BM$ [28] were purchased from Frontier Carbon Co. $CuPc(OMe)_2$ was synthesized according to a previous report [25]. Poly-(3,4-ethylenedioxythiophene)-polystyrene-sulfonic acid (PEDOT–PSS) dispersion in water (Clevios P VP, AI4083) was purchased from Heraeus Precious Metals GmbH & Co. All solvents used in this work were of analytical grade.

2.2. Fabrication of OPVs

The structure of the OPVs was: indium tin oxide (ITO)/PEDOT:PSS/CuPc/PCBM/Ca/Al, as shown in Fig. 1a. OPVs were fabricated in an N_2-purged glove box. The active area of each device was 0.04 cm^2. Before fabrication, a clean glass substrate precoated with a 145 nm-thick ITO layer was treated in a UV–ozone generator for 5 min. PEDOT:PSS was then spin-coated (3000 rpm for 30 s) as an electron-blocking layer (30–40 nm thick) and annealed at 120 °C for 10 min. A 100–200 nm-thick CuPc layer was fabricated on the PEDOT:PSS layer by spin-coating (700 rpm) $CuPc(OMe)_2$ dissolved in organic solvent. The whole device was then heated at 165 °C for 10 min to convert $CuPc(OMe)_2$ to CuPc (p-layer). A solution of mix-PCBM in chlorobenzene was spin-coated the prepared p-layer and then annealed at 65 °C for 10 min (n-layer). Ca (20 nm) was then deposited under vacuum ($<2.0 \times 10^{-4}$ Pa) to act as a hole-blocking layer. After that, Al (80 nm) was deposited under vacuum as a back electrode. Finally, the fabricated OPV cell was encapsulated with a backing glass using a UV-curable resin under N_2 atmosphere. Donor and acceptor material structures are shown in Fig. 1b and c, respectively.

2.3. Characterization of OPVs and thin films

The current density–voltage (J–V) behavior of the encapsulated OPVs was measured under both dark and irradiated conditions. Current–voltage sweeps were obtained with a Keithley 2400 source measurement unit controlled by a computer. The light source for determination of power conversion efficiency was an AM1.5G solar simulator system (Sumitomo Heavy Industries Advanced Machinery) with an intensity of 100 mW/cm^2.

Scanning electron microscopy (SEM) was performed with a Carl Zeiss Ultra55 SEM instrument. Film thickness was measured with a DEKTAK 6 M stylus profilometer.

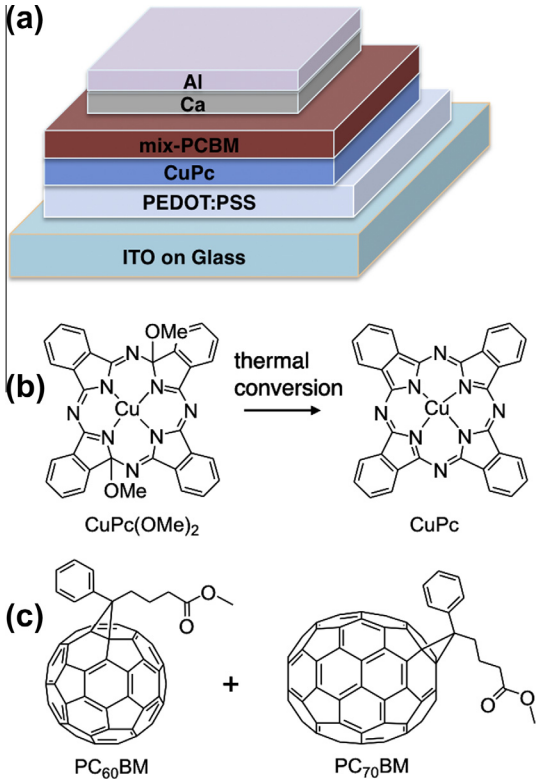

Fig. 1. (a) OPV device configuration of ITO/PEDOT:PSS/CuPc/mix-PCBM/Ca/Al. Structures of (b) donor and (c) acceptor materials.

3. Results and discussion

We first optimized the concentrations of donor precursor $CuPc(OMe)_2$ and acceptor mix-PCBM in the devices. When we used chlorobenzene as a solvent for the donor precursor to prepare the p-layer, a donor concentration of 26 mg/mL and an acceptor concentration of 17 mg/mL gave the device that exhibited the best performance (J_{SC} = 4.82 mA/cm^2, V_{OC} = 0.52 V, FF = 0.39, PCE = 0.98%; Table 1, entry 2) among tested concentrations (Table S1). Changing the concentration of the donor material much influenced on PCE; in contrast, PCE was less affected by the concentration of the fullerene acceptor. The abrupt change of PCE was caused by the increase of J_{SC} with p-layer thickness (Table 1).

Next, we investigated the effect of the solvent of the donor solution on OPV performance. The obtained photovoltaic performance is summarized in Table 1, and J–V curves of OPV devices are depicted in Fig. 2. Although donor pre-

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