

## Doping effects for organic photovoltaic cells based on small-molecular-weight semiconductors

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### ABSTRACT

We have studied highly efficient organic photovoltaic (OPV) cells based on small-molecular-weight semiconductors: zinc phthalocyanine (ZnPc) and C60. To improve the efficiency furthermore, open-circuit voltage ( $V_{oc}$ ) has to be increased. We reported that 5,6,11,12-tetraphenyl-naphthacene (rubrene) produces the highest  $V_{oc}$  of 0.91 V by p–n heterojunction OPV cells with C60. In this paper, we report rubrene doping effects for OPV cells based on ZnPc to obtain higher  $V_{oc}$ .

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### 1. Introduction

Organic photovoltaic (OPV) cells have attracted considerable attention because of their potential for low-cost solar energy conversion. We have already reported highly efficient OPV cells based on small-molecular-weight organic semiconductors. By inserting the i-layer (mixture of p- and n-type molecules) at the p–n interface, power conversion efficiency (PCE) is improved up to approximately 4% under Air Mass 1.5 Global (AM 1.5G) solar-simulated light irradiation [1]. Although we obtain higher short-circuit photocurrent density ( $J_{sc}$ ) by the bulk-heterojunction layer, open-circuit voltage ( $V_{oc}$ ) is under 0.6 V. To improve PCE furthermore,  $V_{oc}$  has to be increased. Various research groups have tried obtaining higher  $V_{oc}$ . A few p-type semiconducting polymers for bulk-heterojunction OPV cells show high  $V_{oc}$ , i.e., 1.04 V for polyfluorene copolymer and 0.77 V for polyphenylene vinylene derivative [2,3]. On the other hand, small-molecular-weight materials had not yet shown such high  $V_{oc}$ . Fig. 1 shows the chemical structures of 5,6,11,12-tetraphenyl-naphthacene (rubrene), zinc phthalocyanine (ZnPc) and C60. Rubrene is well-known as a p-type organic semiconductor with the highest hole mobility. It was reported that single-crystal rubrene organic field-effect transistors show over  $10 \text{ cm}^2/\text{Vs}$  hole mobility [4]. By introducing this rubrene semiconductor into the OPV cell, the highest  $V_{oc}$  of 0.91 V by a rubrene/C60 p–n heterojunction OPV cell

was reported in our previous paper [7]. Fig. 2 shows device structures and energy-level diagrams of ZnPc/C60 p–n heterojunction and rubrene/C60 p–n heterojunction OPV cells. According to a hypothesis of the photovoltaic mechanism based on organic materials, the introduction of a p-type semiconducting material with a lower highest occupied molecular orbital (HOMO) level is effective for obtaining higher  $V_{oc}$ . The HOMO of rubrene is 5.4 eV, which is far lower than that of ZnPc as shown in Fig. 2. Therefore, we succeeded in obtaining the highest  $V_{oc}$  of 0.91 V, which is 50% higher than that of the ZnPc/C60 p–n heterojunction OPV cell. Unfortunately, the rubrene/C60 p–n heterojunction OPV cell shows poor  $J_{sc}$  of  $3.2 \text{ mA}/\text{cm}^2$ , which is almost half of the p–n heterojunction OPV cell based on ZnPc ( $5.2 \text{ mA}/\text{cm}^2$ ). The reason as to why the OPV cell based on rubrene shows such low  $J_{sc}$  is because of the poor absorption of rubrene semiconductor in a long-wavelength region. Fig. 3 shows the comparison of normalized incident photon-to-electron conversion efficiency (IPCE) spectra between rubrene/C60 p–n heterojunction OPV cells and p–i–n OPV cells based on ZnPc. Although the IPCE spectrum in a short-wavelength region (400–550 nm) of the OPV cell based on rubrene is almost similar to the IPCE spectrum of the OPV cell based on ZnPc, the IPCE spectrum in long-wavelength region of the OPV cell based on rubrene is far lower than that of the OPV cell based on ZnPc. Therefore, OPV based on rubrene cannot show enough  $J_{sc}$  for high-performance OPV cells.

Rubrene is also well-known as a dopant for organic light emitting diode (OLED) [5]. Therefore, rubrene doping is a useful method for organic electronic devices to obtain high performances.

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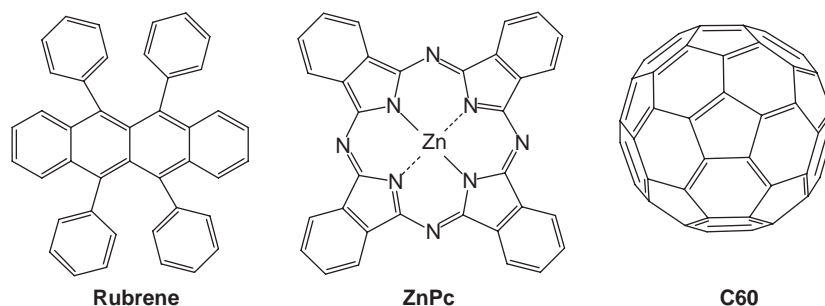


Fig. 1. Chemical structures of rubrene, ZnPc and C60.

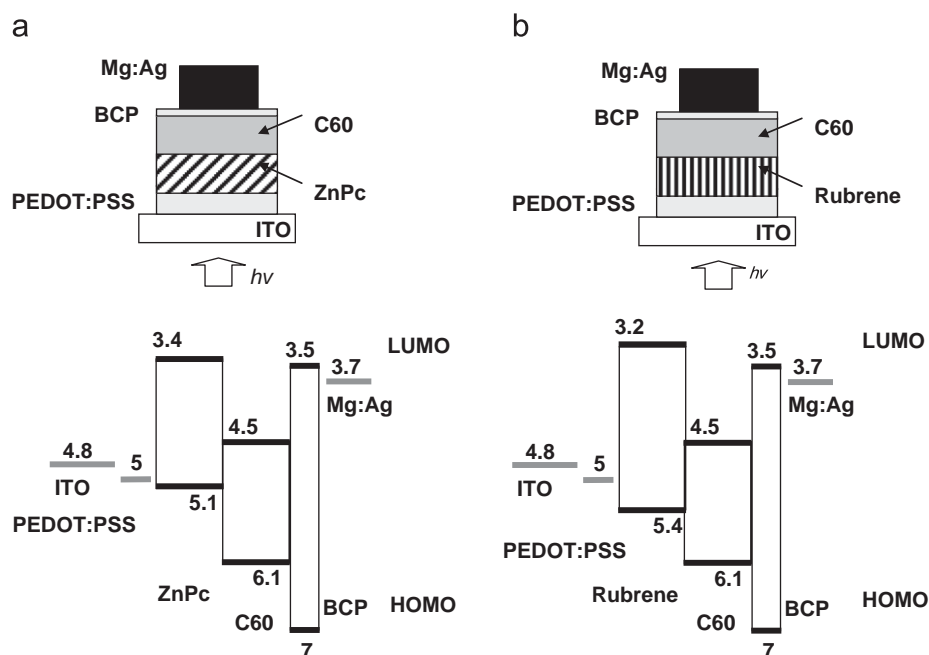


Fig. 2. Device structure and energy-level diagram of the OPV cell of a p–n heterojunction OPV cell based on ZnPc (a) and rubrene (b).

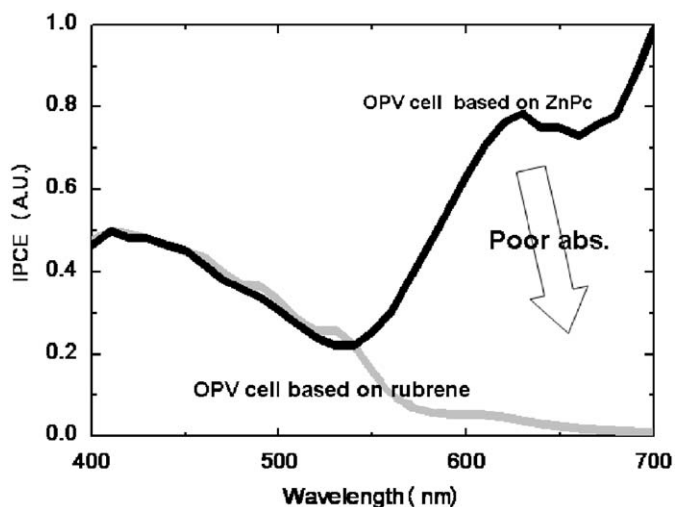


Fig. 3. Normalized IPCE spectra of an OPV cell based on ZnPc and rubrene. Device structure of the OPV cell based on ZnPc was a p–i–n structure.

Rubrene doping effects for OPV cells had already been investigated by Lee's group [6]. Rubrene was directly doped into each layer of a copper phthalocyanine (CuPc)/C60 p–n heterojunction

OPV cell in order to obtain high  $J_{sc}$ . They insisted that high  $J_{sc}$  is attributed to light absorption by rubrene in the range of 460–530 nm where CuPc and C60 have low absorbances. They focused on the improvement of  $J_{sc}$  by using rubrene semiconductors. However, the absorption coefficient of rubrene is too low to obtain enough  $J_{sc}$ . Therefore, rubrene is not proper for the improvement of  $J_{sc}$ .

Here, we investigate the effects of rubrene doping onto the p-layer of a p–i–n junction OPV cell based on ZnPc in order to obtain higher  $V_{oc}$  by rubrene and higher  $J_{sc}$  by ZnPc. In addition, we expect to obtain high Fill factor (FF) due to the high-performance hole transport layer (p-layer) produced by the high hole mobility of rubrene semiconductors.

## 2. Experiments

We used 97% purity ZnPc (Sigma-Aldrich) and 99.95% purity C60 (MTR Co. Ltd.) as p-type and n-type semiconductors, respectively. The ZnPc was purified three times by the gas-flow sublimation method. To fabricate OPV cells, we initially spin-coated poly(ethylenedioxythiophene):polystyrenesulphonic acid (PEDOT:PSS) (Bytron 4083, Bayer) on an indium-tin oxide (ITO) substrate to make its surface smooth. Semiconducting layers were deposited onto the PEDOT:PSS layer by vacuum evaporation using

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