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Compositional engineering of acceptors for highly efficient bulk heterojunction hybrid organic solar cells

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GRAPHICAL ABSTRACT



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ABSTRACT

The wet chemical synthesis of chromium oxide (Cr₂O₃) nanoparticles (NPs) and its application in active layer of inverted bulk heterojunction organic solar cells is documented in this research. Chromium oxide NPs of 10-30 nm size range having a band gap of 2.9 eV were successfully synthesized. These NPs were used in inverted organic solar cells in amalgamation with P3HT:PCBM and PTB7:PCBM polymers. The fabricated hybrid devices improves PCE significantly for P3HT:PCBM and PTB7:PCBM systems. The photophysical energy levels, optoelectrical properties and microscopic images have been systematically studied for the fabricated devices. The introduction of Cr₂O₃ nanoparticles (NPs) enhances light harvesting and tunes energy levels into improved electrical parameters. A clear red shift and improved absorption have been observed for ternary blended devices compared to that observed with controlled organic solar cells. Apparently, when the amount of NPs in the binary polymer blend exceeds the required optimum level, there is a breakdown of the bulk heterojunction leading to lowering of the optical and electrical performance of the devices.

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1. Introduction

In renewable energy resources, electricity through solar energy offers an attractive alternative but it accounts for only $\sim 1\%$ of total world's power consumption [1]. Tremendous research in the field has resulted in the development of various types of photovoltaic cells (PVCs). Among these, solution-processed organic bulk hetero-

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https://doi.org/10.1016/j.jcis.2018.05.027 0021-9797/© 2018 Elsevier Inc. All rights reserved. junction solar cells (OBHJSC) have attracted extensive research attention owing to benefits of reduced fabrication cost, and promising features like eco-friendliness, use of earth-abundant materials, large area, lightweight, mechanical flexibility and tunable energy level alignment [2-5]. Hence, rapid and significant investigations have been carried out to improve the power conversion efficiency (PCE). The several approaches that can increase PCE include interface engineering, surface modification of charge transport layers, insertion of buffer layers/optical spacer, combination of new donor (D) and acceptor (A) materials, use of additives, various







processing methods, and device architectures [6,7]. The efficiencies of OBHJSCs is apparently approaching to its limit i.e. 10–12%. [8]. PCBM based devices have attained the maximum efficiency of 11.6% and 13% is obtained with non-fullerene acceptors. [9,10]. However, further progress in the performance of the binary structure is rather challenging due to low charge mobility, short exciton diffusion length and recombination losses. The weak absorption of the devices in the visible region also restricts further advancement.

A facile strategy could involve blending organic and inorganic semiconductors in the active layer of BHJ as organic-inorganic hybrid bulk heterojunction solar cells-HBHJSCs exhibit rising prospects to integrate the potential advantages of both materials. Semiconducting nanoparticles (NPs) have high physical-chemical stability and strong electronic-photoconducting properties [11]. The combined absorption band of polymer and nanocrystals can effectively enhance the absorption range and build efficient pathways for exciton dissociation at D/A interfaces [12].

In past, hybrid devices with polymer/nanomaterials binary blends have been reported by many researchers [13–17]. There are also reports on the use of ternary and quaternary blends-based hybrid organic solar cells with promising results, generating a lot of interest in PV research [18–20]. The reported increase in efficiencies for TiO₂, ZnO, CuO, NiO, CdSe, PbS, CuInS₂ are 7%, 23%, 18%, 40%, 28%, 23%, 47%, 97%, respectively [21–27].

In this work, we attempted to fabricate ternary HBHJSC devices and to study the influence of incorporation of Cr_2O_3 nanoparticles in the active layer consisting of P3HT, $PC_{60}BM$ and PTB7, $PC_{70}BM$. Chromium oxide is one of the extensively studied p-type semiconductor metal oxide and has a large band gap of 3.0–3.3 eV [28,29]. The incorporation is expected to upsurge the device performance as Cr_2O_3 is highly stable with high electrical conductivity [28] and its large band-gap can effectively block holes and excitons which can redistribute light in the active layer [30].

2. Experimental details

2.1. Materials

Glass substrates coated with ITO of sheet resistance $8-12\Omega$ were obtained from Delta Technologies, USA. Poly[[4,8-bis](2-*ethylhexyl*) *oxy*]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethyl hexyl)carbonyl]thieno[3,4-b] thiophenediyl]][6,6]) (PTB7) was received from 1-Material (Canada) and poly(3-hexylthiophene-2,5-diyl) (P3HT) from Ossila, UK. 99.99% pure PCBM was purchased from Solenne. Chromium chloride (CrCl₃) was procured from Sigma-Aldrich, and sodium hydroxide (NaOH) from Panreac.

2.2. Synthesis of Cr₂O₃ nanoparticles

The Cr_2O_3 nanoparticles with rhombohedral structure were synthesized by a wet chemical method. Typically, 0.1 M solutions of $CrCl_3$ and NaOH were prepared separately in distilled water. The solutions were then mixed and stirred for 2 h at room temperature.

$CrCl_3$ +3NaOH \rightarrow Cr(OH)₃+3NaCl

 $Cr(OH)_3$ precipitate was collected and washed several times with distilled water to remove impurities. The washed precipitates were aged at 60 °C for 48 h and the dried $Cr(OH)_3$ was heated in a muffle furnace at 450 °C for 30 min to obtain Cr_2O_3 .

 $2Cr(OH)_3 \rightarrow 2Cr_2O_3 + 3H_2O$

2.3. Device fabrication

P3HT:PC₆₀BM (1:0.8) and PTB7:PC₇₀BM (1:1.5) solutions were prepared in 1, 2 dichlorobenzene (DCB) and chlorobenzene (CB),

respectively. The prepared solutions were stirred overnight at 70 °C to ensure complete dissolution of the polymers in solvents. Chromium oxide NPs were separately dispersed in DCB and CB and then added into the polymer solutions at different weight ratios (1-4%) of polymer to nanoparticles.

To fabricate the devices, first a thin layer of electron transport layer of ZnO was spin coated on cleaned and patterned ITOs at 5000 rpm for 45sec and heated on a hot plate at 150 °C for 20 min [31]. The binary and ternary hybrid solutions (composed of polymers and nanoparticles) were spun cast onto ZnO coated substrates at 1500 rpm for1min and P3HT-based devices were subsequently heated at 110 °C for 20 min. Finally, a thin layer of 10 nm-MoO₃ and 100 nm-Ag were thermally evaporated at high vacuum (10⁻⁷ mbar) on the devices.

Hereafter, ITO/ZnO/P3HT: Cr_2O_3 :PC₆₀BM/MoO₃/Ag structuredbased inverted devices are referred to as D1 (0%), D2 (1%), D3 (2%), D4 (3%), D5 (4%) and ITO/ZnO/PTB7: Cr_2O_3 :PC₇₀BM/MoO₃/Ag architecture inverted devices as D6 (0%), D7 (1%), D8 (2%), D9 (3%), D10 (4%).

2.4. Characterization

X-ray diffractometer (model: PAN Analytical X'pert PRO) equipped with Cu-K α radiation (1.54 Å) and 2 θ values 20–80°, was used to obtain structural and phase information on the samples. Absorption profile of NPs in solution form, polymer:PCBM and polymer:PCBM:Cr₂O₃ blend in the thin film were recorded using UV–Vis spectrophotometer (model: Genesys 10S). To characterize electrical parameters at 1 sun-1000 Wm⁻² (Xenon bulb coupled with AM 1.5 filter) the solar simulator CT 100AAA with Keithley 2420 source meter unit was used. AFM images were taken on a JEOL JSPM-5200 in tapping mode. External quantum efficiency (EQE) measurements were recorded via model QEX 10PV.

3. Results and discussion

X-ray diffraction pattern obtained on the chromium oxide powder is shown in Fig. 1. The observed prominent peaks were indexed to the rhombohedral phase (Cr₂O₃) based on JCPDS No. 38-1479, except the peak at $2\theta = 31.63^{\circ}$ (2 0 0) plane was indexed to orthorhombic phase (CrO₃) based on JCPDS No. 03-065-1388. The characteristic peak intensity and positions at $2\theta = 24.7^{\circ}$, 33.7°, 36.29°, 41.59°, 45.38°, 50.3°, 54.70°, 63.37°, 65.18°, 73.09°, and 76.9° correspond to the crystal plane of (0 1 2), (1 0 4), (1 1 0), (1 1 3), (2 0 2), (0 2 4), (1 1 6), (2 1 4), (3 0 0), (1 1 9), and (2 2 0) of crystalline Cr₂O₃ and CrO₃ phases without any traces of impurities [29,32]. The average crystallite size of the particles was estimated using the Debye-Scherrer equation shown below and applied to all diffraction peaks.

$$d = k\lambda/\beta_{\theta}\cos\theta \tag{1}$$

where k is a shape factor which normally ranges between 0.9 and 1.0 (in this study K = 0.9), λ is the X-ray wavelength, and β and θ are the half width of the peak and half of the Bragg angle, respectively. Using the equation, the crystallite sizes of Cr₂O₃ nanoparticles were found to be in the range of 10–40 nm. FESEM photograph image collected at 20,000× magnification and shown in Fig. 1(b) reveals the presence of an uniform and homogenous distribution of NPs in the form of cluster agglomerations. The inset demonstrates the transparent magnified image of nanoparticles in the range 10–13 nm from higher resolution FESEM image is in confirmation with the average crystallite size obtained from XRD. The UV–Vis absorption spectrum of Cr₂O₃ NPs is shown in Fig. 1(c). As displayed, Cr₂O₃ NPs have intense absorption in the ultraviolet

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