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Effects of incorporation of copper sulfide nanocrystals on the performance of P3HT: PCBM based inverted solar cells

Sankara Rao Gollu [a], Ramakant Sharma [a], G. Srinivas [a], Souvik Kundu [b,1], Dipti Gupta [a,*]

[a] Plastic Electronics and Energy Lab (PEEL), Department of Metallurgical Engineering and Material Science, Indian Institute of Technology Bombay, Mumbai 400076, India

[b] School of Electrical Engineering and Computer Science, Oregon State University, 1148 Kelley Engineering Center, Corvallis, OR 97331, USA

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ABSTRACT

It has been reported that performance of bulk heterojunction organic solar cells can be improved by incorporation of an additive like metal and semiconducting nanoparticles in the active layer. Here in, we have synthesized Cu_2S nanocrystals (NCs) by chemical route and studied its dispersion in poly (3-hexylthiophene) [6, 6]-phenyl C61-butyric acid methyl ester (P3HT: PCBM) matrix. Variation in the performance parameters with change in the concentration of Cu_2S NCs into the P3HT: PCBM matrix has also been studied and it was found that the inverted geometry device with concentration of 20 wt% of Cu_2S NCs and having the structure ITO/ZnO (NPs)/P3HT: PCBM:Cu_2S NCs/MoO_3/Al has shown maximum efficiency of 3.39% which is more than 100% increase in comparison with devices without Cu_2S NCs. Photoluminescence measurements studies unveiled that the incorporation of Cu_2S NCs into a P3HT: PCBM matrix has helped in quenching photoluminescence which suggests more effective exciton dissociation at the interfaces between the P3HT and PCBM domains. The Nyquist plots obtained from impedance spectroscopy at 1 V bias in the dark has suggested the effective lifetime and global mobilities for P3HT: PCBM as 0.267 ms and 1.17×10^{-3} cm^2/V-S and for P3HT: PCBM:Cu_2S NCs (20 wt%) systems as 0.156 ms and 2.02×10^{-3} cm^2/V-S respectively. Based on observed photoluminescence quenching, calculated effective lifetime and global mobility, we have tried to explain the possible reason for improvement in the efficiency with the very well dispersion of Cu_2S NCs into the P3HT: PCBM matrix.

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1. Introduction

Recently, organic solar cells (OSCs) have attracted a significant attention because of several promising features such as low cost, light weight, easy processability, and high-mechanical flexibility, which could be well exploited in scalable fabrication techniques for achieving a long standing goal of economic and eco-friendly solar energy harvesting [1–9].

Among the various materials studied for their application as a photoactive layer in bulk heterojunction OSCs, poly (3-hexylthiophene) (P3HT) and [6, 6]-phenyl C61-butyric acid methyl ester (PCBM) blend system have shown promising power conversion efficiency (PCE) in the range of 6–7% [10]. These efficiency levels are predominantly achieved by obtaining control over morphology of the active layer [11], understanding absorption phenomenon, charge injection and charge transport processes in the device [12,13]. Experimentally, various routes such as

* Corresponding author.
E-mail address: diptig@iitb.ac.in (D. Gupta).
[1] Present address: Center for Energy Harvesting Materials and Systems (CEHMS), Virginia Tech, Blacksburg, VA 24061, USA.

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thermal annealing [14], solvent annealing [15], use of transport layers [16], incorporation of an additive in the active layer [17], etc. were employed to reach to these numbers. Further to these approaches, another concept of hybrid solar cells, wherein organic semiconductor blends mixed with inorganic nanocrystals (NCs) is investigated that utilize beneficial features of inorganic nanocrystals (like better charge transport and wide range absorption) and have resulted in significant efficiency improvements. The most commonly used NCs are metal nanoparticles (Ag [18] and Au [19] nanoparticles) and semiconductor nanocrystals (like CdS [20], CdSe [17], CdTe [21], ZnO [22] and Cu_2S [17,23], FeS_2 [24], PbS [25], $CuInS_2$ [26]) which are incorporated in the active layer. The metal nanoparticles (NPs) help in significantly enhancing the optical absorption, either via the formation of scattered waves at the large diameter NPs or due to the excitation of localized surface plasmon resonance (LSPR) modes at the smaller diameter NPs [27]. On the other hand, the semiconductor NCs not only offers the enhancement in spectral window but also provides an opportunity of energy band alignments that aids in exciton dissociation and charge transport [28]. However, benefits from such incorporation of NCs in the active layer of P3HT: PCBM can only be obtained when they make a type-II energy-offset with P3HT: PCBM so that excitons generated in both p- and n-type materials are dissociated into holes and electrons [29]. This point renders ZnO and CdS incompatible for incorporation into active layer. Additionally, nanoparticles of CdSe and CdTe cannot be used for large scale fabrication owing to the limited availability of selenium and tellurium as well as high toxicity of these semiconductors.

In this regard, p-type semiconducting Cu_2S nanocrystals with the band gap of 1.2 eV might emerge as a potential candidate as the valance band and conduction bands of Cu_2S will help in forming type-II energy-offset [29] with P3HT: PCBM. Further, Cu_2S NCs exhibit unusual properties such as high thermal and photo-chemical stability, broad absorption up to near IR range, high fluorescence quantum yields, high charge carrier mobilities and high electron affinities [30]. Based on these facts, it is expected that the incorporation of Cu_2S NCs in P3HT: PCBM matrix would enhance both absorption and charge carrier mobility. In a recent study by Lu et al. [23] and Liao et al. [17], it was shown that incorporating Cu_2S NCs and Cu_2S nanodisks in P3HT: PCBM matrix facilitates formation of nanostructure morphology that improves electron transport and enhances the device performance. These studies provide a good insight in pointing out the relevance of NCs in P3HT: PCBM matrix and in relating the device performance to the active layer morphology, however further studies are still required which provide a clear picture of the operating device mechanisms in these ternary systems. In this paper, we therefore outline a detailed picture of the device operation by including studies such as photoluminescence and impedance spectroscopy, which shed light on absorption phenomenon and carrier lifetime, respectively. The devices are fabricated in an inverted geometry using photoactive blends of P3HT: PCBM and P3HT: PCBM:Cu_2S NCs. ZnO NPs and MoO_3 has been used as electron and hole transport layers, respectively [31]. Variation in the performance parameters with change in the concentration of Cu_2S NCs into the P3HT: PCBM matrix has also been studied.

2. Experimental section

2.1. Synthesis of Cu_2S nanocrystals

Cu_2S NCs have been synthesized using a chemical route method reported elsewhere [32]. At first, a suspension 1 mM of copper (II) acetylacetonate and 3 mL of oleic acid is prepared and labeled as solution A. In a separate three-neck flask, 1.25 mM of ammonium diethyldithiocarbamate, 10 mL of dodecanethiol and 17 mL of oleic acid were mixed and then heated up to 110 °C under Argon gas flow. Then solution A is swiftly added into the reaction and reaction mixture is quickly taken up to 180 °C and kept at the temperature for 10–20 min. After the reaction is over, the solution containing precipitates of Cu_2S NCs is allowed to cool down naturally till 120 °C and then taken out of the flask for centrifuging at 5000 rpm for 5 min. The precipitates were re-dispersed in toluene and then again allowed to precipitate by adding isopropanol followed by centrifuging at 5000 rpm for 5 min. The procedure is repeated three times to clean away the residue of dodecanethiol and oleic acid. The as obtained powder of Cu_2S NCs is dried at 60 °C for 30 min and then used for characterization and fabrication of devices.

2.2. Synthesis of ZnO nanoparticles

ZnO inorganic nanoparticles (INPs) were synthesized at room temperature and details are described elsewhere [33]. It has been characterized for confirming the particle size and crystal structure.

2.3. Material characterization

X-ray Diffraction (XRD) pattern and X-ray photoelectron spectroscopy (XPS) studies were carried out by using Philips Analytical X-ray Diffractometer and ESCA Multilab 2000 respectively. HRTEM images were taken using JEOL JEM 2100F. Absorption of Cu_2S NCs and active materials was investigated by UV–VIS–NIR spectrophotometer (Perkin–Elmer Lambda 750). Emission of Cu_2S NCs and active materials was investigated by Varian, Cary Eclipse (Fluorescence spectrophotometer). Surface morphology of the blend films and Cu_2S NCs were analyzed by scanning probe microscopy (tapping mode atomic force microscopy – Veeco Systems).

2.4. Device fabrication and testing

Fig. 1 shows the device architecture. The process of fabrication began with a preparation of photoactive blends of P3HT: PCBM and P3HT: PCBM:Cu_2S NCs in a glove box. Regioregular P3HT (Average Mw ~64,000, (Aldrich)) and PCBM (Nano-C) were used in the preparation of blends. Initially four different blends of P3HT: PCBM (1:1 weight ratio) in dichlorobenzene with 20 mg/ml concentrations

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