

Unified assay of adverse effects from the varied nanoparticle hybrid in polymer–fullerene organic photovoltaics

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ABSTRACT

Nanoparticles (NPs) having different surface capping agent, variant electrical conductivity and sunlight absorption have been studied for the ternary hybrid containing poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C-61-butyric acid methyl ester (PCBM) in bulk heterojunction (BHJ) organic photovoltaics (OPVs). These NPs are composed of conducting gold, semiconducting CdS or PbS, or insulating cage-like molecular silica (POSS). We use a series of microscopic methods including TEM, AFM, SEM, optical, and fluorescence microscopy to estimate NP size and to probe the agglomeration and/or aggregation of NPs in P3HT/PCBM blends. Surface capping agent aromatic thiophenol (SPh) was found to be poor in the dispersion of NPs in P3HT/PCBM blends. The light harvesting of these NPs ranges from transparent (POSS NP), near transparent (CdS NP), visible light absorbing (Au NP), to near-infrared absorbing (PbS NP). Nevertheless, the absorbance of these NPs is all too small relative to that of P3HT polymer. Concerning the charge separation of P3HT exciton, the LUMO energy levels of these NPs have been determined by the combination of optical band-gap energy and HOMO energy levels. By the transient photocurrent time-of-flight method, charge carrier mobility of P3HT/PCBM/NP(CdS-SPh) ternary hybrid was found to be improved, although fluorescence quenching studies imply insufficient or ineffective contact between P3HT and all NPs in the present study. NPs hybrid P3HT/PCBM BHJ OPVs were fabricated by solution process. Regardless of conductivity or sunlight absorbance, all NPs show no improvement on power conversion efficiency of ternary hybrid OPVs, which is 3.03–3.91% less than 4.0–4.1% of P3HT/PCBM OPVs without NPs. Based on the present study, a few problems that associate with the inferior performance of NPs hybrid P3HT/PCBM BHJ OPVs are delineated.

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1. Introduction

Polymer-based organic photovoltaics (OPVs), the so called third generation solar cells, provide a potential solar energy utility with favorable features, such as low-cost, all-solution process, large-area, and mechanical flexibility [1–3]. Not long ago, power conversion efficiencies (PCEs) surpassing 5% have been achieved with the blends of poly(3-hexylthiophene) (P3HT) and a fullerene derivative, [6,6]-phenyl-C-61-butyric acid methyl ester (best known as PCBM) [4]. Recently, OPV PCE more than 9% has been realized by renovated

device structure and newly developed low-band-gap polymers, which show major absorption wavelength longer than 600 nm and more efficiency in harvesting solar energy [5–18]. Due to the unique structural feature, namely regioregularity, which facilitates the packing of polymer chains, the long wavelength absorption (the vibronic feature in the absorption spectra) of P3HT can be substantially improved by device processing conditions, such as thermal annealing and solvent annealing methods [19–22]. Other than extending absorption wavelength in matching solar spectrum, extensive polymer chain packing often facilitates the photocurrent output of polymer-based bulk heterojunction (BHJ) OPVs because of the favorable active layer nanoscale morphology [23–25], namely interdigitated charge transporting channel (percolation pathway).

In this regard, a wide range of nano-size materials in different forms, such as nanoparticles (NPs), nanocrystals, nanorods, nanofibers, nanoclusters, or quantum dots, have been incorporated into polymer-based OPVs to form the hybrid system [26–28],

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which may bring about interdigitated charge transporting channel and thus higher PCE of OPVs can be acquired. These nanomaterials can be electrical conducting (metallic) like Au, Ag, and Pt. They can also be semiconducting, such as CdS, CdSe, CdTe, PbS, PbSe, CuInSe₂, ZnO, and TiO₂ [29–31]. Although many of them absorb visible light (such as Ag, Au, CdSe, CdTe, HgTe, and CuInSe₂) or near-infrared (IR) light (such as PbS and PbSe), some of them are transparent (such as ZnO and TiO₂) or near transparent (such as Pt NP having no distinct plasmonic resonant absorption or small size CdS NPs) [29–31]. In the ordered organic–inorganic hybrid BHJ OPVs [32], electron transporting inorganic materials (Si, TiO₂, ZnO, CdS, and CdSe) in the form of nanofibers or nanorods are vertically aligned beneath polymer-based active layer on the electrode. However, the PCEs reported for such hybrid solar cells have been modest due to the problem of pore infiltration of the semiconducting polymer into the nanostructures [32–35]. Otherwise, one critical issue of such hybrid polymer-based OPV is the agglomeration and/or aggregation of the nanomaterials if they all blended and solution processed together in the fabrication of BHJ OPVs. Therefore, a surface capping agent or ligand, which is an organic species with an elongated saturated hydrocarbon chain, is implanted onto the surface of these nanomaterials to depress their agglomeration and/or aggregation and to enhance their polymer mixability.

Here in this report, we are particularly interested in analyzing the impact of organics capped NPs in P3HT/PCBM-based BHJ OPVs, of which the active layer is a ternary hybrid. These NPs are POSS-SC₁₆, CdS-SC₁₂, CdS-SPh, PbS-OA, Au-SC₁₂, and Au-SPh (Scheme 1). In addition to the different surface capping agent, either insulating saturated hydrocarbon chain (SC₁₆, SC₁₂, or OA oleic acid) or semiconducting aromatic benzene ring (SPh), the interior content of these NPs is distinctive in terms of electrical conductivity. They are conducting Au, semiconducting PbS and CdS, and insulating polyhedral oligomeric silsesquioxane (POSS). They are also quite different in sunlight absorption, such as transparent POSS, nearly transparent CdS NP, nearly panchromatic visible light absorption Au NP and near-IR absorption PbS NP. Surveying literature, a broad range of variances in PCE have been reported for NP hybrid P3HT/PCBM BHJ OPVs. A significant improved PCE from 2.63% to 4.08% has been found for conducting Pt NP hybrid OPVs [36]. Similar PCE enhancement (4.08% improved from 3.43%) also has been reported for conducting Ag NP hybrid OPVs [37]. In contrast, no beneficial effect on P3HT/PCBM OPV has been found for conducting Au NP hybrid in a more comprehensive study recently [38], although an earlier report had demonstrated a substantial PCE enhancement for Ag and Au NPs [39]. For semiconducting CdS NP hybrid P3HT/PCBM OPVs, the resulting PCE 0.95% is very low, from which it is hard to see the NP effect because of similarly low PCE 0.74% of P3HT/PCBM OPVs without CdS NP [40]. Very recently, a near-IR absorption PbS NP hybrid P3HT/PCBM system has been reported,

although it is a photodiode study irrelevant to OPV [41]. Similarly, long wavelength (~700 nm) absorption CdTe NP hybrid P3HT/PCBM system has shown no photovoltaic characteristic but photoconductive gain for photodetectors [42].

2. Experiments

2.1. Instrumentation and measurement

2.1.1. Transmission electron microscopy (TEM) measurements

P3HT:PCBM of 1:0.8 weight ratio or various weight ratios of NP-containing P3HT:PCBM samples were dissolved in *o*-dichlorobenzene. The solution was then added dropwise onto the carbon films on copper grid (200 mesh). Each sample was then heated at 60 °C for 2 h at ambient environment and then transferred to a vacuum oven to remove the residue organic solvent for 16 h. The measurements were performed using a JEOL JEM-2010 transmission electron microscope (200 kV accelerating voltage and LaB6 filament).

2.1.2. Atomic force microscopy (AFM) measurements

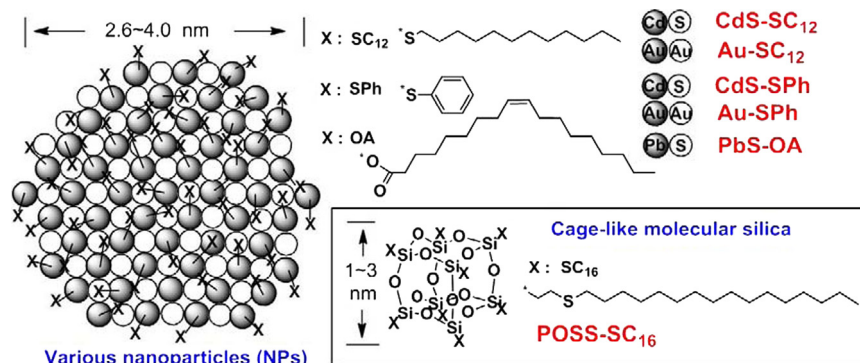
The substrates (ITO glass) were first coated with PEDOT:PSS and then P3HT:PCBM:NP hybrid. Each sample was heated at 150 °C for 10 min for the thermal annealing treatment. The measurements were carried out with a multimode atomic force microscope (Digital Instruments, Nanoscope III) using tapping mode with a silicon tip.

2.1.3. Scanning electron microscopy (SEM) measurements

The substrates (Si wafer) were coated with P3HT:PCBM:NP thin films and heated at 150 °C for 10 min for the thermal annealing treatment. The sample for cross-sections examination was prepared on ITO-coated glass. The measurements were performed using a JEOL JSM-6700 field-emission scanning electron microscope, having resolution of 1.0 nm (15 kV) or 2.2 nm (1 kV), accelerating voltage in the range of 0.5–30 kV, and the magnifying scale of 25–650,000.

2.1.4. Optical and fluorescence microscopy measurements

The substrates (quartz plates) were spin-coated with P3HT:PCBM or P3HT:PCBM:NP thin films and heated at 150 °C for 10 min for the thermal annealing treatment. The measurements were conducted using an Olympus BX51 microscope with an objective lens of Olympus MPlanFL N (100X/NA 0.9). For the fluorescence images, the excitation wavelength was set at 510–550 nm and the detecting wavelength was greater than 590 nm. We used CoolSNAP HQ2 air cooled CCD as the detector.



Scheme 1. Schematic illustration of the composition and size of NPs and the chemical structure of their surface capping agent (x).

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