



Insight into the efficiency enhancement of polymer solar cells by incorporating gold nanoparticles

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ABSTRACT

Photovoltaic performances of polymer bulk-heterojunction solar cells (PSCs) with various sized (20, 35, 50, and 75 nm) Au nanoparticles (NPs) incorporated on indium tin oxide (ITO)-coated glass substrates are investigated in detail, wherein poly(3-hexylthiophene):phenyl-C61-butyrac acid methyl ester (P3HT:PCBM) blend serves as active layer and poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) as anode buffer layer on ITO. The optical and electrical properties of these devices incorporated with Au NPs with different space distributions in the interface of PEDOT:PSS buffer layer and P3HT:PCBM active layer are investigated. We find that, the optical property is improved as the Au NPs are large enough to penetrate into the active layer, while the performance of PSCs with small Au NPs can only benefit from the improved hole collection efficiency. Meanwhile, the exciton dissociation efficiency reduces remarkably as increasing the size of Au NPs. Finally, we demonstrated a maximum power conversion efficiency (PCE) improvement of ~23% in the PSCs by incorporating 35 nm Au NPs.

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1. Introduction

Polymer-fullerene-based bulk heterojunction (BHJ) solar cells are the highly promising candidates for photovoltaic devices because of their properties including simple fabrication procedure, low-cost materials, physical flexibility and semi-transparency [1–7]. Currently the power conversion efficiency (PCE) of these BHJ polymer solar cells (PSCs) has reached as high as 7–9% [8–10]. However, compared with inorganic solar cells, PSC has one important hindrance for further efficiency improvement: the useful thin active layer for efficient light absorption is limited by the short exciton diffusion length and the low carrier mobility [11,12]. As a result, enormous efforts are being made in the development impactful light-trapping techniques [13–15].

Among the various light-trapping techniques, one promising method is the incorporation of localized surface plasmon resonant (LSPR) metallic (e.g., Au, Ag, Cu) nanoparticles (NPs) into the BHJ PSCs. So far, many studies have shown the PCE improvement of PSCs by incorporating metallic NPs in either the buffer layer such as poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) or the active layer [16–25]. Recently, a high PCE of 8.79% (improved from 7.59%) was reported by utilizing both a metallic nanograting electrode as the back reflector and metallic NPs embedded in the active layer [9].

Although many reports on the metallic NPs enhanced PSCs the exact understanding on the photovoltaic performance changes remains inconclusive. On one hand, several studies have shown that the absorption of photoactive conjugated polymer in the active layer can be enhanced due to the plasmonic scattering or near-field enhancement caused by LSPR effect of the metallic NPs (mainly enhance the optical property) [26,27]. On the other hand, Choy et al. indicated that the strong near field around Au NPs due to LSPR effect mainly distributes laterally along the PEDOT:PSS layer, leading to minimal enhancement of light absorption in the active layer. Thus they contributed to the improvement of PCE to the enlarged interfacial area between active layer and PEDOT:PSS buffer layer as well as the improved PEDOT:PSS conductivity (mainly improve the electrical property) [28]. In these studies, they all investigated the effect of metallic NPs with a fixed size, however, there are distinct optical properties and space distributions in the PEDOT:PSS layer for various sized metallic NPs. Consequently, the investigation of the performance changes of PSCs with various sized metallic NPs is highly important and desirable to better understand the physics within the 'plasmonic' PSCs.

Herein we study the comparative performances of poly(3-hexylthiophene) (P3HT)/phenyl-C61-butyrac acid methyl ester (PCBM) BHJ PSCs incorporated various sized Au NPs (20 nm, 35 nm, 50 nm and 75 nm) on ITO-coated glass substrates. We investigate how the optical and electrical properties of these devices perform when Au NPs with different space distributions in the interface of PEDOT:PSS buffer layer and P3HT:PCBM active

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layer. At an optimized Au NPs size of 35 nm, a maximum PCE improvement of ~23% (from 2.37% to 2.91%) was demonstrated.

2. Experimental

2.1. Synthesis of Au NPs

The various sized Au NPs were prepared by a hydroquinone-reduction seed-growth method reported previously [29]. Briefly, a 15 nm diameter Au seeds solution was prepared according to the protocol by Frens [30], resulting in a colloidal suspension containing $[Au]=1.5$ mM. Then a 99 mL “growth solution” containing 3 mL 10 mM centrifuged $H AuCl_4$ solution, 250 μ L 1% w/w sodium citrate solution and a suitable amount of Au seeds solution (ranging from 11.5 mL to 0.16 mL, which yield particle sizes of 20 nm up to 75 nm) was prepared in a 250 mL flask. At last, 1 mL 30 mM hydroquinone was injected into the growth solution under vigorous stirring. These reactions completed within 1 h. In order to cap the Au NPs with poly-(vinylpyrrolidone) (PVP, MW=10,000, Aldrich), we added 3 mL 100 mM PVP solution to the reaction solution, and stirring for 24 h. Finally, all solutions were centrifuged to eliminate the excess ligands and redissolved in 1 mL of deionized water to avoid the aggregation of Au NPs.

2.2. Device fabrication

The ITO-coated glass substrates were ultrasonicated in detergent, acetone and isopropyl alcohol, dried with nitrogen, and treated by ozone-ultraviolet cleaner for 15 min. The same volume of various sized Au NPs aqueous solutions were spin-coated at 2000 rpm onto ITO-coated glass substrates, subsequently annealed at 140 °C for 10 min in air, and treated by ozone-ultraviolet cleaner for another 10 min (which can promote the spread of PEDOT:PSS solution). For the control device under the same conditions, we spin-coated the same volume of pure deionized water on ITO, and denoted as 0 nm Au NPs. The PEDOT:PSS solution (Baytron P 4083) were next spin-coated at 4000 rpm on the top of the layer of Au NPs and were subsequently annealed at 140 °C for 15 min in air, resulting in a thickness of ~30 nm layer. The mixed solutions consisting of P3HT (10 mg mL⁻¹, Aldrich) and PCBM (9 mg mL⁻¹, Aldrich) in 1,2-dichlorobenzene were then spin-coated at 500 rpm on the PEDOT:PSS layer in air. The thickness of the active layer is ~150 nm. After spin-coating photoactive layer, Al (100 nm) was thermal evaporated under high vacuum of $\sim 4 \times 10^{-4}$ Pa with a rate of 0.2 nm/s onto the polymer layer as a cathode to create a device with an active area of 9 mm² defined by a shadow mask. The final device structure is ITO/Au NPs/PEDOT:PSS/P3HT:PCBM/Al, as shown in Fig. 1a. Finally, thermal annealing was performed by directly placing the device on a hotplate at 150 °C for 10 min in an inert glove-box. It should be noted that the device fabrication conditions were optimized prior to this investigation.

2.3. Device characterization

The transmission electron microscope (TEM) images of Au NPs were measured using a JEM-1230EX TEM. The ultraviolet-visible (UV-vis) absorption spectra of solutions and films on ITO substrates were measured on a CARY 100 Bio UV-vis spectrophotometer. For the absorption tests of P3HT:PCBM films on ITO/Au NPs/PEDOT:PSS, we used the corresponding ITO/Au NPs/PEDOT:PSS sample as the reference, which can eliminate the absorption of ITO/Au NPs/PEDOT:PSS. The photoluminescent (PL) spectra of P3HT/PCBM films were measured on a Hitachi F-4500 spectrophotometer (excited at 470 nm). The current density-voltage (*J-V*) characteristics of devices under dark and illumination (100 mW/cm²) were measured

with a Keithley 2400 measurement source unit at room temperature in air. The light intensity for the solar simulator was calibrated with a standard photovoltaic (PV) reference cell. Incident photon to electron conversion efficiency (IPCE) curves were measured with a Stanford lock-in amplifier 8300 unit. Atomic force microscopy (AFM) image was measured by Veeco MultiMode scanning probe microscope operated in tapping mode in air. The film thickness was measured by an AMBIOS XP-1 high-resolution surface profiler.

3. Results and discussion

3.1. Photovoltaic characteristics of devices with various sized Au NPs

The current density-voltage (*J-V*) characteristics of OPV devices incorporated with various sized Au NPs (20 nm, 35 nm, 50 nm and 75 nm) in the PEDOT:PSS anode buffer layer are shown in Fig. 1b. It should be noted that all devices with all sized Au NPs were the ones with optimized Au NPs coverage density. The device photovoltaic performances are summarized in Table 1. It is observed that PCE of our PSCs is improved by 23% from 2.37% (0 nm) to 2.91% (35 nm), as increasing the size of Au NPs from 0 to 35 nm. Further increasing the size of Au NPs, the PCE values drop to 2.49% (50 nm) and 2.04% (75 nm). The improvement of PCE in the devices with small Au NPs is mainly attributed to the increase in fill factor (FF) and short-circuit current (J_{SC}), from 0.5 to 0.52 and 7.59 mA cm⁻² to 9.05 mA cm⁻², respectively. From the dark *J-V* curves in Fig. 1c, we can observe that the series resistance (R_s) of the devices, obtained from the inverse slope of the dark *J-V* curves at a voltage of 0.5 V, slightly decreases from 5.2 Ω cm² (0 nm) to 4.94 Ω cm² (20 nm), and then increase to 6.76 Ω cm² (75 nm) as increasing the size of Au NPs. Fig. 1d shows the IPCE characteristics of these devices, we can obtain the highest IPCE of 70% at 500 nm in the device with 35 nm Au NPs, with a ~19% improvement to the control device (0 nm). Further increasing the size of Au NPs, the peak IPCE detrimentally decreases to 60% at 500 nm in the device with 75 nm Au NPs. This is in good agreement with the trend of J_{SC} . We also do some preliminary investigation on the stability of the OPVs with and without Au NPs, and no obvious changes of the stability are observed after incorporating Au NPs. In the following sections, we will study the optical and electrical properties of our PSCs, which describes the underlying physics of the ‘plasmonic’ PSCs in a better way.

3.2. LSPR effect of various sized Au NPs on photovoltaic performance

Fig. 2a–d shows TEM images of various sized Au NPs used in our PSCs, it is observed that the average size of these Au NPs is approximately 20 nm, 35 nm, 50 nm, and 75 nm, respectively. The absorption spectra of these Au NPs in water are shown in Fig. 2e. Corresponding to the excitation of LSPR, the absorption peaks for these Au NPs are at 523 nm, 530 nm, 536 nm, and 549 nm respectively, which all match well with the absorption of the P3HT:PCBM film.

In order to verify the degree of contributions of the LSPR effects, we measured the absorption spectra of P3HT:PCBM active layers on top of ITO/Au NPs/PEDOT:PSS with various sized Au NPs, as shown in Fig. 3a. During the measurement of light absorption, we have eliminated the absorption of ITO/Au NPs/PEDOT:PSS as it absorbs light. So the absorption spectra represent light absorbance within the active layer only. Interestingly, the devices with smaller Au NPs (20 nm and 35 nm) have no significant difference in absorption compared with the control device (0 nm) while there are obvious absorption enhancements in the devices with larger Au NPs (50 nm and 75 nm). It has been reported that when Au NPs are incorporated into the PEDOT:PSS layer, the strong

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