

Facile embedding of gold nanostructures in the hole transporting layer for efficient polymer solar cells

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ABSTRACT

Photon harvesting by the active layers of bulk heterojunction (BHJ) polymer solar cells (PSCs) greatly influences the power conversion efficiency (PCE) of the devices. Inclusion of novel metal nanostructures in photoactive layers is one of the effective ways to enhance light trapping without increasing the device thickness. Here, we demonstrate a dramatic enhancement (up to 43%) of the power conversion efficiency (PCE) by exploiting gold nanostructures (AuNSs)-embedded graphene oxide (GO) as the hole transporting layer (HTL) in BHJ PSCs. The enhancement of the device performance could be attributed to the increase in both short circuit current density (J_{sc}) and fill factor (FF). The increased FF is most likely the result of the enhanced charge collection through modified HTL and anode contact. Furthermore, the optical properties of the photovoltaic devices suggest that AuNSs cause light trapping via plasmonic effects resulting enhanced J_{sc} .

1. Introduction

Bulk heterojunction organic photovoltaic devices (OPVs), in particular polymer solar cells have received a great deal of attention from academia as well as industries for harvesting solar energy. This technology offers a variety of advantages such as low cost fabrication, simple solution processibility, light weight, mechanical flexibility, potential for large area and high throughput roll-to-roll production [1–5]. During the last decade, much research effort has been put forward to develop highly efficient PSCs. Various strategies have been adopted including the synthesis of low band gap polymers for more light harvesting and the design of novel device architectures. Till date, the best PCE achieved for single junction PSCs is 11.8% and 13.2% efficiency has been reported by Heliatek company for multi-junction PSCs [6,7]. However, PSCs are still suffering from low PCE compared to inorganic solar cells. Short exciton diffusion length (~ 5 – 10 nm) in PSCs limits the efficiency of exciton dissociation resulting in low short circuit current density (J_{sc}). It also reduces the thickness of the active layer leading to poor light absorption and hence low J_{sc} .

One of the key challenges in enhancing the PCE is to improve the light absorption without increasing the thickness of the active layer.

Recently, plasmonic resonant metallic nanostructures [8] have been exploited in PSCs to control the energy flow at nearly the atomic level by manipulating light-matter interaction. Plasmonic nanoparticles with different geometries including nanospheres [9–11], nanorods [10,12], nanocubes [10,12], nanoprisms [10,13], and nanooctahedra [10,14] have been introduced mainly in the active layer to enhance the optical absorption. The performance of organic photovoltaics has found to be enhanced with the use of these nanostructures [10,11,15–20].

In spite of efficient charge generation via exciton dissociation, the efficiency of PSCs is still low due to losses via charge recombination and inefficient charge collection. Interface (cathode/anode) layers play an important role in charge collection by the electrodes. The polymer, poly(3,4-ethylene-dioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) is widely used as HTL in PSCs [21–23]. However, its strong acidic nature not only etches the ITO electrode, but also its hygroscopic property allows the moisture to penetrate into the active layers resulting reduction of the device efficiency and lifetime [24,25]. Quasi two-dimensional (2D) GO has emerged as an interface material for organic electronic devices due to its unique chemical and electronic structures along with solution processability [26–28]. It is highly transparent like graphene in the solar spectral range and has a similar work function as

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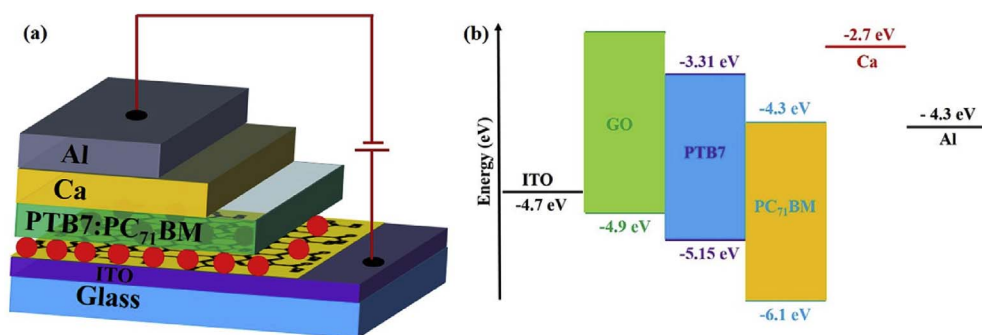


Fig. 1. (a) Schematic device structure and (b) energy level alignment of PTB7:PC₇₁BM solar cells.

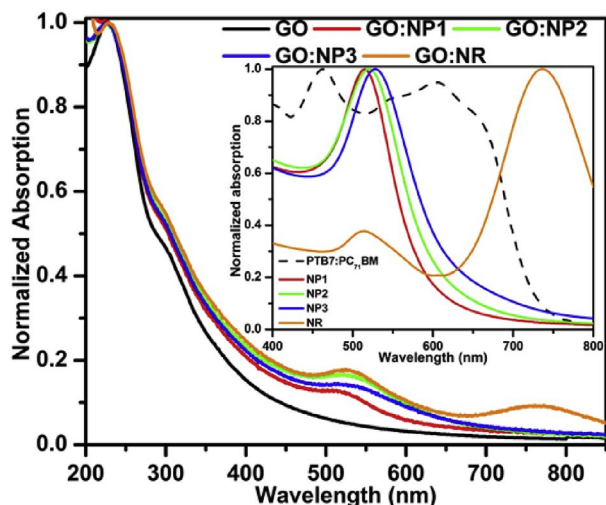


Fig. 2. Normalized absorption spectra of GO and AuNSs incorporated GO. Inset: Normalized absorption spectra of PTB7:PC₇₁BM blend and various AuNSs.

PEDOT:PSS [29]. Moreover, plasmonic effect can be incorporated in PSCs by dispersing the metal nanostructures on 2D-GO sheets in the HTL [30–33]. Recently, Chuang et al. [34] have shown enhancement in the photocurrent as well as the device performance by introducing AuNP decorated GO in the HTL or/and in the active layer. Plasmonic GO sheets were used in an anode buffer layer in inverted solar cells to improve the efficiency and stability of the devices [35]. Jang and co-workers [36] have employed AuNPs on the PEDOT:PSS doped reduced GO (rGO) as an HTL in plasmonic organic solar cells and demonstrated a 9.34% improvement in the PCE. Laser induced nucleated AuNPs on GO were utilized in the plasmonic solar cells [37]. The anchoring of AuNPs on the basal plane and edges of GO causes 13.6% enhancement in the device performance. Till date, mostly AuNPs (of spherical shapes) have been coupled to GO in the HTL, but not nanostructures of other shapes. The systematic study of the effect of the geometry of AuNSs on the optical and electrical properties of the PSCs is still lacking. Previous experimental studies suggest that the improvement of the device efficiency due to the incorporation of AuNSs is the result of more light absorption via plasmonic effect, but the exact mechanism is yet poorly understood.

In this study, we demonstrate the application of GO sheets decorated with AuNSs of various geometries as HTL in a low bandgap polymer Poly ({4,8-bis [(2-ethylhexyl)oxy] benzo [1,2-b:4,5-b'] dithiophene-2, 6-diyl} {3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno-[3,4-b]thiophenediyl}) (PTB7) and [6,6]-phenyl C₇₁ butyric acid methyl ester (PC₇₁BM) based BHJ solar cells. The efficiency of the photovoltaic devices is found to enhance significantly with the incorporation of AuNSs and depends on the size as well as the shape of AuNSs. Our results infer that the enhanced performance of the PSCs due to incorporation of AuNSs in the HTL could be attributed to both increased

light trapping by plasmonic antennas and improved charge collection via modified anode contact.

2. Experimental techniques

Indium tin oxide (ITO) coated glass substrates (resistivity 7 Ω/□) were sequentially cleaned in detergent (5% labolene solution), de-ionized (DI) water, acetone and isopropyl alcohol (IPA) for 20 min each in an ultrasonic bath. Cleaned substrates were treated with UV-ozone for 15 min to remove organic residuals. To prepare the solutions for interfacial layer, synthesized GO powder was redispersed in DI water following ultrasonication of about 60 min. Dispersed GO was then added to the AuNP (NP) and AuNR (NR) dispersions with 1:1 v/v ratio under a mild ultra-sonication. Thus, prepared solutions were spin coated on the ITO substrates at 3000 rpm for 60 s followed by low-temperature annealing for 10 min. The PTB7:PC₇₁BM (1:1.5 wt ratio) solution with a concentration of 25 mg/mL was prepared in chlorobenzene (anhydrous) and 1,8-diiodooctane (97:3 v/v) mixed solvent. Such blended solution was stirred inside the nitrogen-filled glovebox at 45 °C for several hours and heated at 75 °C for 60 min. An active layer of about 90 nm thickness was deposited on the top of HTL by spin coating the blended polymer solution. The films were dried overnight inside the glovebox. Finally, the photovoltaic devices were prepared by depositing Ca (20 nm) as an electron transporting layer (ETL) and Al (100 nm) as a cathode in a vacuum of 1×10^{-6} torr through thermal evaporation (using a thermal evaporator from Angstrom Engineering, Nexdep). The structure of the device (ITO/GO/PTB7:PC₇₁BM/Ca/Al) and corresponding energy level alignment are illustrated in Fig. 1. All the devices were characterized in inert atmosphere inside the nitrogen-filled glove box. The current density versus voltage (J-V) characteristics of the fabricated devices were measured using a Keithley 4200-SCS parameter analyzer. Solar irradiation was simulated using a class AAA solar simulator (Oriel Sol3A) with AM1.5 (air mass) filter calibrated with a standard Si solar cell. External quantum efficiency (EQE) was recorded by a QE measuring system (Enlitech technology limited).

3. Results and discussion

3.1. Morphology and optical property of AuNS loaded GO films

The AFM topography image of GO thin film, drop casted on Si substrate is shown in Fig. S1a. It is clear that the lateral dimension of individual GO sheet ranges up to micrometer. The AFM height profile (of a step-edge GO sheet) yields the GO sheet thickness of 2.13 nm (Fig. S1b) indicating the presence of tri-layers [29]. The Raman spectra of GO (Fig. S1c) display two prominent peaks: D band at 1360 cm⁻¹ corresponding to the breathing mode of k-point phonons of A_{1g} symmetry as well as from bonds between sp³-hybridized atoms, and a G band (ca. 1603 cm⁻¹) arising from the first order scattering of the E_{2g} phonons of sp²-hybridized C atoms [38].

TEM images of the synthesized AuNSs are shown in Fig. S2. Sizes of the synthesized gold nanoparticles are around 5 nm (NP1), 20 nm

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