



Power conversion efficiency enhancement of organic solar cells by addition of gold nanostars, nanorods, and nanospheres



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ABSTRACT

The effects of gold (Au) nanoparticles (NPs) with different morphologies (star, rod, sphere) incorporated into buffer layer, poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), of polymer-based organic solar cells (OSCs) were investigated. Solar cells having gold nanoparticles exhibited significant improvement in device efficiency relative to the reference device. The observed improvement is most likely due to the surface plasmon and enhanced light reflection and scattering properties of the Au NPs. The power conversion efficiency (PCE) is increased ca. 29% with Au nanostars, ca. 14% with Au nanorods and 11% with Au nanospheres compared to the device with no Au NP (reference device). Au nanostars provide the strongest contribution to the efficiency among all NP morphologies studied as they have large size, sharp features, and strongest localized surface plasmon resonance effect associate with their morphology.

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1. Introduction

Photovoltaic (PV) devices emerge as an attractive way of converting energy directly from the sunlight [1,2]. Among all types of PV devices, polymer-based organic solar cells (OSCs) are particularly intriguing since they have the advantages of low production cost, processing at room temperature, mechanical flexibility, and suitability for mass production in addition to being relatively quiet, not producing toxic substances or greenhouse gases, and requiring little maintenance during operations [1–3]. However, these types of solar cells are struggling with some challenges, which limit photon absorption efficiency, thus power conversion efficiency (PCE). For instance, exciton diffusion lengths are short (~10–20 nm) for most conjugated polymers; therefore, recombination rates are higher

[2,4–6]. Excitons that recombine do not contribute to the photocurrent, and result in a decrease of PCE [2]. Moreover, photon absorption efficiencies are needed to be improved, especially on the red side of the electromagnetic spectrum. One solution might be an increase of the active layer thickness, however, this also increases the device resistance due to relatively low carrier mobility of organic semiconductors in an active layer, and rate of charge recombination and trapping [2,7–9]. Therefore, the quest for developing methods to improve the device efficiency without increasing active layer thickness has become an exciting and an active research field in the augmentation of OSCs.

The use of noble metal (e.g. gold (Au), silver (Ag)) nanoparticles (NPs) in solar cell structure is a promising way of addressing these problems associated with OSCs and improving the device efficiency [10,11]. Noble metal NPs show strong localized surface plasmon resonance (LSPR) effect that contribute to the light absorption by intensifying the electromagnetic field on or near NP surface and light scattering by the surface [7,12–18]. Increased light scattering may result light trapping for a longer period

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within the film and lead to an increase in interaction time of light with the active layer. Such an increase in the optical path length will contribute to the enhancement in the photogeneration of mobile carriers [19,20]. Increasing number of studies are reporting incorporation of noble metal NPs into the layers (active or buffer layer) of OSCs [7,10,13,15,21]. The addition of NPs in the active layer may increase exciton quenching; therefore, they are generally preferred to be placed within the buffer layer where some part of the NPs may extend into the active layer for extending the LSPR effect and to reduce the device resistance [20]. Majority of these studies are focused on the use of noble metal NPs with spherical morphologies [13,15,21]. However, the LSPR effect is much stronger for NPs with sharp features (i.e. rod, cube, diamond and star), which are expected to provide greater contribution to the device efficiency. A few study reported the use of anisotropic-shaped noble metal NPs in OSCs [7,20,22,23] and to best of our knowledge there is no report on the addition of star-shaped Au NPs in the solar cell structure.

Herein, we report an incorporation of Au NPs with three different morphologies (star, rod and spherical) into poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) layer of OSC and compare the effect of NP morphology on device efficiency. Among the studied NP morphologies, the highest PCE is obtained with the use of star-shaped Au NPs. This result was expected considering the superior optical properties of Au nanostars over nanorods and nanospheres as demonstrated with their use as surface enhanced Raman spectroscopy (SERS) substrates [24]. The special optical properties associated with their distinct morphologies, this time, endow Au nanostars for contributing to the development of solar cell technology.

2. Experimental

2.1. Materials

Gold (III) chloride trihydrate ($\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$), sodium borohydride (NaBH_4), ascorbic acid (AA), silver nitrate (AgNO_3), and sodium citrate (Na-citrate) were purchased from Sigma–Aldrich. Cetyltrimethylammonium bromide (CTAB) was purchased from Fluka. Indium-tin-oxide (ITO) coated glasses were purchased from VisionTek. Poly(3-hexylthiophene) (P3HT) was purchased from Sigma Aldrich and phenyl-C60-butyric acid methyl ester (PCBM) was purchased from Solenne BV. Ultra-pure deionized (DI) water (18Ω) was used for all the experimental solutions.

2.2. Methods

Au nanoparticles (star, rod, and sphere) were synthesized with methods described in the literature [24–26]. The methods used in NP synthesis and photovoltaic device preparation are described below.

2.2.1. Synthesis of Au nanostars

First, 0.2 mL of an aqueous 0.01 M solution of $\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$ was added to 4.75 mL of a 0.1 M CTAB solution in

a test tube. The solution was gently mixed. Then, 0.03 mL of a 0.01 M AgNO_3 solution and 0.032 mL of a 0.1 M AA solution were added in this order, results a colorless solution. Finally, 10 μL of commercially available 10 nm Au NP seed solution was added. After the reaction mixture was gently mixed, the solution was kept undisturbed for 3 h at room temperature. The color of the nanorod solution became blue–purple indicating the formation of Au nanostars [24]. The concentration (in number of particles/mL) of as-synthesized nanostars was determined by TEM analysis as ca. 3×10^{10} nanostars/mL. The details of this analysis were reported in our previous publication [24].

2.2.2. Synthesis of Au nanorods

(i) *Preparation of seed solution.* First, 1.25 mL of an aqueous 0.002 M $\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$ and 2.74 mL ultra-pure DI water were added in a test tube. Then, 3.76 mL of a 0.2 M CTAB solution was added onto the mixture. After mixing, 0.9 mL of a 0.01 M ice-cold NaBH_4 solution was added and the color of the solution turned from orange to light-brown color. The solution was kept at room temperature in water bath for 2 h. (ii) *Preparation of growth solution.* The concentrations of $\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$, CTAB, AgNO_3 , L-ascorbic acid, and molar ratio of L-ascorbic acid to $[\text{AuCl}_4^-]$ were adjusted as 4×10^{-4} M, 9.5×10^{-2} M, 6×10^{-5} M, 6.4×10^{-4} M, and 1.6:1, respectively and added in a test tube in that order. Then, the seed solution was added into the mixture. Final ratio of $[\text{Au}]_{\text{seed}}$ to Au ion concentration, $[\text{AuCl}_4^-]$, was 1/320. Finally, the solution was mixed for 20 s and left undisturbed at room temperature for 24 h [25]. The concentration (in number of particles/mL) of as-synthesized nanorods was determined as ca. 6×10^{10} nanorod/mL [24].

2.2.3. Synthesis of Au nanospheres

Three-step procedure was used to synthesize spherical Au NPs. In the first step, 9.5 mL ultra-pure DI water, 0.25 mL, 0.01 M $\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$, and 0.25 mL, 0.01 M Na-citrate were added in a test tube. After mixing homogeneously, 0.3 mL, 0.1 M, freshly prepared, ice-cold NaBH_4 solution was added and the color of the solution turned to pink. The citrate-stabilized spherical Au NPs (3.5 nm in diameter) solution was kept undisturbed for 3 h after mixing by rapid inversion. In the second step, 45 mL, 0.08 M CTAB, 1.125 mL, 0.01 M $\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$, and 0.25 mL, 0.1 M AA were added in a test tube and mixed by gentle inversion. Then, 5 mL of the citrate-stabilized seed solution was added. The resultant spherical Au NPs solution (8 nm in diameter) was mixed for 10 min. In the third step, 45 mL, 0.08 M CTAB, 1.125 mL, 0.01 M $\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$, and 0.25 mL, 0.1 M AA were added in a test tube. Finally, 5 mL of the spherical Au NP solution of 8 nm in diameter was added. The resultant spherical Au NPs solution (20 nm in diameter) was mixed for 10 min and kept undisturbed for 3 h [26]. The concentration (in number of particles/mL) of as-synthesized nanospheres was determined by TEM analysis as ca. 4×10^{10} nanorod/mL [24].

2.2.4. Photovoltaic device preparation

Photovoltaic devices based on conjugated polymers with a blend of Au nanorods are prepared as shown in

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