# Investigation of the effect of copper nanoparticles incorporated in ZnO buffer layer of inverted organic solar cell

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# Abstract

This study examines the effect of cheap and easy to synthesize copper nanoparticles (CuNPs) for its surface plasmon resonance behavior on the performance of organic solar cells (OSC). A simple synthesis of stable CuNPs is reported together with a procedure for their optimized incorporation in the OSC architecture via dispersing them on ZnO interfacial layer. The CuNPs triggers the localized surface plasmon resonance resulting in enhancement of short circuit current density under AM1.5 illumination. Copyright © 2017 VBRI Press.

Keywords: Surface plasmon resonance, copper nanoparticles, organic solar cells.

### Introduction

Last decades has witnessed continued growing research interest in organic solar cells (OSC) owing to its numerous advantages such as light weight, mechanical flexibility, low-cost and low-temperature roll to roll processing [1]. Although power conversion efficiency (PCE) of OSCs has reached up to 12% [2], they are still far from commercialization and need focused efforts for improvement of performance. Due to limited charge carrier mobilities of organic materials, the typical organic active layer of OSC is very thin (~150 nm) and is insufficient for light absorption. Along with other factors of surface reflection and scattering [3], all are the causes for optical losses and thus the low performance of OSCs. Further improvements in PCE of OSCs can be expected, only upon reduction of such optical losses. The issues of optical losses have been addressed via improvement in device architecture [4], judicious choice of interface materials [5] and electrode materials [6-9], and the use of optical spacers [10-12]. Nevertheless, the insufficient light absorption and trapping is still a major issue that limits photocurrent generation and alternative strategies for reduction in optical losses are highly desired.

The integration of plasmonic structures which are generally metallic nanoparticles (NPs), within all generation of photovoltaic devices is one of the promising approaches to address the issue of photon absorption and trapping. **[13-14]**. NPs supports surface plasmon resonances (SPR) by creating multiple scattering sites that induces multiple absorptions. The incident light with matched SPR energy of the NPs is strongly scattered and

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absorbed, and the magnitude of the extinction (which is a combination of scattering and absorption) depends on the geometric features of the structure, such as size and shape of the NPs. The NPs could be incorporated into OSC device architecture either by their dispersion in the photoactive layer or the buffer layer or in between the interfaces of two [15]. The researchers have applied various metallic NPs such as gold (Au), silver (Ag), copper (Cu) and platinum (Pt) in OSC architecture [16 -24]. Among various NPs, noble metals such as Au and Ag have been of particular interest until now because of their strong SPR in the ultraviolet and visible regions of the electromagnetic spectrum. Many experimental and theoretical studies have reported the improved performance of OPVs with the incorporation of Au and Ag NPs [25-30]. However, the availability and cost of Au and Ag are possibly deterrent factors for their widespread implementation in the plasmonic OPV architecture. Among various other metal NPs, Cu has attracted much interest in recent years due to its high abundance, low cost and highly conductive nature [31, 32]. Although CuNPs shows relatively weaker SPR in the visible range, it has demonstrated enhanced photocurrent response in near infra-red region for Si-based devices [33-35].

Through present study, we investigated the SPR effect of CuNPs on the device characteristics of an inverted polymer/fullerene OSC while incorporating them with an electron transporting ZnO interfacial layer. We found PCE of CuNPs OSC, utilizing P3HT: PCBM as an active layer were enhanced to 2.74% from 2.57% in comparison to the reference OSC without metal NPs, due to SPR effect of CuNPs. This boost in performance was

mainly due to increased short-circuit current density (Jsc) which was supported by the formation of ZnO nano-ridge kind of structure after addition of CuNPs in low concentration. The study indicates that SPR effect of CuNPs could be a feasible alternative for improving the performance of future OSCs.

## Experimental

## Synthesis of CuNPs

The materials, Hydrazine hydrate (68%) and Ethylene glycol (EG), Copper (II) sulfate pentahydrate  $(CuSO_4.5H_2O)$ were directly used as received. For synthesis, first 0.04 mole of CuSO<sub>4</sub>.5H<sub>2</sub>O was dissolved in 40 mL of Ethylene glycol (EG) and was heated at 140 °C in an oil bath having a magnetic stirrer for 20 mins. After that 100 µl of hydrazine hydrate was added in solution, and the whole solution was further stirred vigorously for two hours. On addition, the color of the solution became darker, and solid reddish brown mesh precipitated at the bottom of the flask. After cooling the precipitate was isolated by centrifuging it three times with toluene at 6000 rpm for 15 mins each. Finally, the collected samples were dried at 60 °C for 24 h in vacuum.

### Preparation of Sol-gel ZnO

The sol-gel ZnO was used as an electron transport interfacial layer in all the OSC devices, and it was prepared by the mixed reaction of 0.5 M zinc acetate dehydrate in 0.5 M monoethanolamine and 2-methoxyethanol for 30 mins. Further, CuNPs were added to sol-gel ZnO solution in various concentration of 0, 1.5, 2.5, 5, 10 and 15%, respectively.

## Fabrication of OSC

All OSC devices were fabricated in the configuration of ITO/ZnO/P3HT: PCBM/MoO<sub>3</sub> (10 nm)/Ag (100 nm) with CuNPs (**Fig. 1**). For fabrication, the patterned indium tin oxide (ITO) glass substrate (sheet resistance =  $15 \ \Omega \ sq-1$ ) was sequentially cleaned through ultrasonication in distilled water, acetone, ethanol and isopropanol respectively. After exposing substrates for 20 min UV-ozone, the sol-gel ZnO was spin-coated at 6000 r.p.m. for 40 secs, followed by annealing at 150 °C for 5 min. After that, ZnO coated substrates were transferred to glove box; a solution P3HT/PCBM (Conc.) was spin coated at 1000 rpm for 60 secs, followed by annealing at 150 °C for 5 min. Finally, all devices were completed by thermal evaporation of MoOx hole transport layer and Ag cathode (100 nm).

## Thin films characterization

Ultraviolet-visible spectroscopy (UV-vis) of different Thin films were recorded on a JASCO-V-550 spectrophotometer at room temperature in air. X-ray diffraction (XRD) patterns of pristine ZnO and ZnO with different concentration of CuNPs deposited on substrates were obtained using a D2 PHASER X-ray diffractometer (BRUKER) equipped with a focusing-graded X-ray mirror. Scans were taken with a 1 mm wide source with X-ray generator settings of 30 kV and 10 mA. Photoluminescence (PL) spectra were measured with the HORIBA Jobin Yvon FluoroMax-3 spectrofluorometer. A scanning electron microscope (FESEM) of JEOL JSM-6500F was used for obtaining surface morphology and cross-sectional view of various thin films. The transmission electron microscopy (TEM) of samples was done using FEI Tecnai<sup>TM</sup> G2F-20S model. To confirm the presence of CuNPs on ZnO thin films X-ray Photoelectron spectroscopy (XPS) study was done using a VG Theta Probe instrument.



Fig. 1. Schematic illustration of device structures with nanoparticles in the buffer layer.

### OSC characterization

The device area was 0.04 cm<sup>2</sup>. J-V characteristics of the photovoltaic cells were measured using a Keithley 2400 source unit under a simulated AM 1.5G spectrum. With an Oriel 9600 solar simulator, the light intensity was calibrated by a KG-5 Si diode; the measurements are carried out at ambient conditions. The external quantum efficiency (EQE) measurements of the devices were taken by using an Enli tech (Taiwan) EQE measurement system.

### **Results and discussion**

Metallic NPs are known to exhibit strong absorption bands in the UV-visible region, and these bands are assigned to the excitation of plasmon waves on their surfaces. The physical origin of the enhanced absorption is the coherent oscillation of the free electrons induced by the electromagnetic field. The SPR in NPs is primarily characterized by UV-Vis spectroscopy because the peak positions and shapes are sensitive to a particle size of NPs. Fig. 2a shows the extinction spectra of CuNPs in solution. The extinction peak could be seen at around 580 nm, which is similar to the results of previous studies [36]. Inset image of Fig. 2(a) shows the high-resolution TEM (HRTEM) image of CuNPs. The synthesized CuNPs are spherical in shape and highly dispersed in solution with average diameters of  $50 \pm 5$  nm. Fig. 2b shows the absorption spectra of the samples with different

concentrations of CuNPs in ZnO solutions. We can see from **Fig. 2(b)** increased absorbance intensity with the increasing concentration of CuNPs, suggesting a direct correlation of concentration of NPs with its SPR effect.



**Fig. 2.** (a) UV–vis absorption spectra of CuNPs: inset image is TEM pictures of CuNPs and (b) UV-Vis spectra of the samples with various concentrations of Cu NP in ZnO solutions.

For confirming the presence and studying the impact of different concentration of CuNPs on ZnO layer, we fabricated thin films using ZnO solutions with a different CuNPs concentration on the glass surface and performed characterization with several techniques. Fig. 3(a) shows the SEM image of pristine ZnO and Figs. 3 (b-f) are the SEM images of ZnO films with different concentration of CuNPs. An addition of CuNPs caused a change in the surface morphology of ZnO nanostructures. At lower concentrations (1.5, 2.5 and 5%), CuNPs acted as heat trapper which resulted in the reorganization of zinc acetate dihydrate [Zn(CH<sub>3</sub>COO)<sub>2</sub>.2H<sub>2</sub>O] gel particles and shaped them into the ridge-like structure. When the concentration of CuNPs increased up to 10 and 15 %, the morphology underwent a further transformation forming an aggregation (Fig. 3e and f). For further confirming the presence of CuNPs on the ZnO layer, HRTEM was carried out on pristine ZnO and ZnO films with highest and lowest concentration of CuNPs. Fig. 4a clearly depicts lattice fringes of ZnO having an interplane spacing of 0.26 nm, which matches well with the spacing

between (002) crystal planes of ZnO. TEM micrographs of the ZnO with CuNPs revealed lattice fringes of D-Spacing of 0.26 nm and 0.21 nm corresponding to the (002) plane of zinc and the (111) plane of the facecentered cubic CuNPs, confirming the presence of CuNPs in ZnO thin film surface (**Fig. 4b and c**). The structural evolution of ZnO with and without CuNPs was also examined through the XRD spectra, as shown in **Fig. 5a**. Two major diffraction peaks can be assigned as (100) and (002) lattice planes, respectively, according to JCPDS no.36-1451. The spectra clearly reveal the formation of hexagonal wurtzite phase ZnO. The XRD patterns of ZnO/Cu nanoparticles exhibits one additional peak at 52° which correspond to (200) planes of Cu, confirming the existence of CuNPs in ZnO.



**Fig. 3.** (a) SEM images of buffer layers prepared with pristine ZnO and (b - f) ZnO with different concentration of CuNPs.



Fig. 4. HRTEM images of (a) pristine ZnO and (b,c) ZnO with 1.5 and 15 % CuNPs.

5 nm

As the surface energy of the ZnO film can affect the interfacial property of the ZnO and active layer, thus, the performance of the solar cell, water contact angle (WCA) measurements of various ZnO layers was carried out to study the surface energy, and the results are shown in **Fig. 5b**. The WCA somewhat increases with respect to the increasing of CuNPs concentration. However, the change is not huge that it could result in the substantial change of surface energy of ZnO.



**Fig. 5.** (a) X-ray diffraction (XRD) patterns of pristine ZnO and ZnO with different concentration of Cu nanoparticles (b) Measured water contact angles of ZnO films without and with various concentrations of CuNPs.

XPS was further employed to confirm the presence of CuNPs on ZnO thin film surface. The spectra of Zn 2p and Cu 2p are shown in **Fig. 5a** and **b**. The binding energy positions for Zn 2p3/2 and Zn 2p1/2 are obtained at 1021.1 eV and 1044.1 eV, respectively. The difference in binding energy between two Zn peaks is about 23.0, which matches well with the earlier reported value [37]. **Fig. 6b** showed the binding energies of Cu 2p orbitals having 2p3/2 and 2p1/2 peaks at 932.9 and 952.5 eV respectively, revealing the presence of Cu nanoparticles on the surface of ZnO.

To explore the effect of the SPR on the exciton generation behavior, we also carried out steady-state photoluminescence (PL) measurements on P3HT: PCBM thin films prepared on various ZnO layers (**Fig. 7**) as the PL directly associate with the photon absorption. The spectra show increased PL intensity with the increasing in concentrations of CuNPs from 1.5% to 5%. We infer that the enhanced PL was probably due to the increased level of photon absorption resulted from SPR effect. The higher the concentration of CuNPs, the more the plasmonic effect, thus, the higher degree of photon absorption. Thereby, the higher PL intensities. On the other hand, the PL intensity declined with further increasing in CuNPs concentration of 10 and 15%. Such decreasing in PL could cause by the severe aggregation of CuNPs in ZnO, which reduce the transmittance of ZnO layer and thereby the photon absorption of P3HT: PCBM layer. Also, the SPR effect highly depending on the CuNPs sizes and the distances between particles. Therefore, the apparent aggregation of 10 and 15% samples might lead to the deterioration of SPR, which also decline the photon absorption. Nevertheless, the PL results indirectly confirm the SPR effect of CuNPs.



Fig. 6. XPS spectra of (a) Zn2p and (b) Cu2p of ZnO layer with CuNPs.



**Fig. 7.** Photoluminescence spectra of P3HT: PCBM films prepared on ZnO layers without and with different concentrations of CuNPs.

Fig. 8a shows current density versus voltage (J-V) characteristics of devices with structure ITO/ZnO(with and without CuNPs)/P3HT: PCBM/MoO<sub>3</sub>/Ag (100 nm) incorporating various concentrations of CuNPs in the ZnO layer. The key cell parameters, including Jsc, Voc, fill factor (FF), series resistance (Rs), shunt resistance (Rsh) and power conversion efficiency (PCE) of all devices are listed in Table 1. The reference device without CuNPs showed Jsc of 8.17 mA/cm<sup>2</sup>, Voc of 0.58 V, FF of 54%, and power conversion efficiency of 2.57%. After the addition of CuNPs to the ZnO interfacial layer, an altering behavior could only be in Jsc while almost unaltered Voc suggesting the unchanged nature of the electrode-organics interface or donor/accept (P3HT/PCBM). It is rational as the only variation of The devices are the ZnO layers. The maximum power the conversion efficiency of 2.74 % was achieved with  $Jsc = 8.74 \text{ mA/cm}^2$ , Voc = 0.59 V, and FF = 53% in the case of 1.5% CuNPs addition. The increase could also be attributed to ridge-shaped nanostructure (Fig. 3) on ZnO surface at low CuNPs concentrations. The structure seems to have a better charge extraction capability due to the large interfacial area. However, at higher concentration due to reorganization CuNPs in aggregates, charge extraction capability decreases sharply. The impact could be observed with increasing of CuNPs (2.5, 5, 10 and 15 %), where Jsc was seen to be decreasing resulting in corresponding deterioration of device performances. However, it is noteworthy that the Jsc values of 2.5 and 5% devices are still higher than that of the pristine device, suggesting the positive impact of plasmonic effect due to CuNPs.

Device	PCE (%)	Jsc (mA/cm <sup>2</sup> )	Voc (V)	FF (%)	Rs (Ω.cm <sup>2</sup> )	$\frac{\text{Rsh}}{(k\Omega.\text{cm}^2)}$
Standard (0% CuNPs)	2.57	8.18	0.58	54.12	3.91	0.52
1.5 % CuNPs	2.74	8.75	0.59	53.00	4.09	0.56
2.5 % CuNPs	2.52	8.52	0.58	51.00	4.18	0.42
5 % CuNPs	2.16	8.21	0.55	47.87	4.58	0.41
10 % CuNPs	1.93	7.54	0.58	44.25	12.02	0.42
15 % CuNPs	1.07	6.55	0.59	27.20	29.77	0.17

On the other hand, the Rs increased from 3.91 to 29.77  $\Omega$  cm<sup>2</sup> and Rsh decreased from 0.52 to 0.17 k $\Omega$  cm<sup>2</sup> after incorporating the CuNPs, resulting in the drastic declining in FF. **Fig. 8b** presents EQE curves of the devices with and without CuNPs. The photocurrent within the wavelength range from 450 to 700 nm increased significantly after incorporating CuNPs into ZnO (except 10 and 15%) as compared to the standard cell device. Also, the degree of EQE on different ratios of CuNPs in ZnO matches well with the level of Jsc deduced from J-V

measurements. Although we could attribute enhancement of EQE to the presence of the CuNPs-induced surface plasmon, other possibilities such as multiple internal reflections or higher surface area due to ridge-like ZnO surface cannot be completely ruled out.

From the devices characterization results, it is clear that incorporation of CuNPs in ZnO layer led to two competition effects. On the one hand, the photon absorption of active layer increases due to SPR effect. On the contrary, the electrical properties of the OPV devices deteriorated with growing the concentration of CuNPs, possibility due to the reason that CuNPs might act as electrons trap [**38**] and aggregation of CuNPs could also hinder the electron transport through ZnO buffer layer. Therefore, to improve the overall efficiency of OPV device, it is imperative to determine the optimized concentration of CuNPs in ZnO layer.



**Fig. 8.** (a) Current density–voltage (J-V) characteristics of the OPV devices without and with different concentrations of CuNPs in ZnO buffer layer, under 100 mW/cm<sup>2</sup>, AM 1.5G simulated solar illuminations (b) EQE curves of the OPV devices without and with different concentrations of CuNPs in ZnO buffer layer.

### Conclusion

In summary, we have improved the device performance of OPV by blending CuNPs into the ZnO interfacial layer. By controlling the concentration of CuNPs in ZnO precursor solution, the SPR induced by the CuNPs led to a noticeable enhancement in the photocurrent of the device. We expect that the results reported here might be of further use in other material systems such as low bandgap polymers to achieve even higher PCEs.

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