



# Incorporation of SiO<sub>2</sub> dielectric nanoparticles for performance enhancement in P3HT:PCBM inverted organic solar cells



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## ABSTRACT

It is well known that organic solar cells (OSCs) with inverted geometry have not only demonstrated a better stability and longer device life time but also have shown improved power conversion efficiency (PCE). Recent studies exhibit that incorporation of metal and/or semiconducting nanoparticles (NPs) can further increase the PCE for OSCs. In this present work, we have synthesized SiO<sub>2</sub> NPs of various sizes (25, 50, 75 and 100 nm) using the modified Stober method and incorporated them into P3HT:PCBM photoactive layer and ZnO based electron transport layer (ETL) in order to investigate the light trapping effects in an OSC. Absorption studies have shown a considerable increase in photo absorption in both cases. The fabricated devices demonstrated 13% increase in the PCE when SiO<sub>2</sub> NPs are incorporated in P3HT:PCBM photoactive layer, whereas PCE was increased by 20% when SiO<sub>2</sub> NPs are incorporated in ZnO based ETL. Mott–Schottky analysis and impedance spectroscopy measurements have been carried out to determine the depletion width and global mobility for both the devices. The possible reason for PCE enhancement and the role of SiO<sub>2</sub> NPs in active layer and ZnO ETL are explained on the basis of the results obtained from Mott–Schottky analysis and impedance spectroscopy measurements.

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## 1. Introduction

Owing to the promising features like low cost, light weight, easy processability, eco-friendly, and high-mechanical flexibility, bulk heterojunction based organic solar cells (OSCs) are always at the helm of research when compared to other energy harvesting devices [1,2]. In the last decade, extensive research on donor/acceptor system comprising of poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) have demonstrated the solar devices with power conversion efficiency (PCE) of ~6% [3]. The PCE was further improved to ~9% by using low band gap polymers as electron donors [4,5] in the system. However, the obtained PCE is still low when compared to silicon solar cells and it is due to the difficulty in achieving a trade-off between light absorption and exciton harvesting efficiency [6]. In most of the polymers, light absorption depth is slightly higher than the exciton diffusion length (which is nearly 5–10 nm), thereby restricting the thickness of active layer in the devices. The problem is more critical for many new low-band gap materials as their

absorption coefficients are typically lower than those of blue and green absorbers, which may lead to insufficient light absorption in OSCs [7].

Different strategies such as folded device architectures [8,9], a periodic dielectric stacks [10], diffraction gratings [9], textured grooves [11] and incorporation of metal [12] and semiconducting nanoparticles in active layer and in transport layers have been adopted to boost the light harvesting in OSCs. It was also found from the literature that incorporation of metal nanoparticles (NPs) such as Ag [12], Au [13], or semiconducting NPs such as CdSe [14], CdTe [15], PbS [16], PbSe [17], Cu<sub>2</sub>S [18,19], and ZnO [20] are helpful to increase the PCE. Recent research report explains that further improvement in the performance parameters of P3HT:PCBM based inverted OSCs are possible after the incorporation of SiO<sub>2</sub> dielectric and metal core/shell NPs [21]. The reason for improved PCE after incorporation of NPs in active layer and in transport layers is enhancement in the absorption of light by plasmonic resonance and scattering, respectively. Nunomura et al [22] demonstrated that submicron-sized SiO<sub>2</sub> NPs can be used as light scatters in a thin-film silicon tandem solar cell consisting of hydrogenated amorphous (a-Si:H) and microcrystalline (μc-Si:H) silicon. Their studies have shown that submicron-sized SiO<sub>2</sub> NPs lead to

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forward scattering owing to the Mie Scattering and thereby help to increase the optical path length. In addition, difference in the refractive indices of SiO<sub>2</sub> and silicon can induce subsequent internal reflection, which could lead to enhancement in the PCE. Based on this knowledge, in our previous work [23], the efforts have been manifested to understand the efficacy 200 nm SiO<sub>2</sub> NPs embedded into the 250 nm thick P3HT:PCBM active layer OSCs. Although, our simulations results have shown significant increment in the scattering cross-section, the devices ultimately failed to achieve higher efficiency which is the most intriguing factor for OSCs. Two reasons are responsible for the poor efficiency: (i) the thickness of active layer was higher than the charge carrier diffusion length which could lead to the recombination of excitons, and (ii) dielectric nature of SiO<sub>2</sub> offers poor mobility of charge.

To overcome these obstacles, in our present work, both the size of SiO<sub>2</sub> NPs as well as the thickness of active layer has been reduced to establish the synergy between light absorption and exciton harvesting efficiency. This work is on the first systematic study of different sizes SiO<sub>2</sub> NPs embedded into P3HT:PCBM based photoactive layer and ZnO based electron transport layer (ETL) in order to improve the PCE. We hypothesize that SiO<sub>2</sub> NPs will lead to forward scattering owing to the Mie Scattering and thereby increases the optical path length. To validate our hypothesis, at first, a systematic study has been carried out using 25 nm sized SiO<sub>2</sub> NPs to optimize the weight percentage of SiO<sub>2</sub> NPs to be incorporated in the photoactive blends of P3HT:PCBM so as to get maximum PCE. Thereafter, keeping the weight percentage as constant, SiO<sub>2</sub> NPs of different sizes were incorporated into the P3HT:PCBM based photoactive layer and ZnO ETL. The devices were fabricated in an inverted geometry where photoactive blends of P3HT:PCBM was used as an active layer, and ZnO and MoO<sub>3</sub> was served as an ETL and hole transport layer, respectively. Attempts have also been made to provide a clear understanding of device operation using a Mott–Schottky analysis and impedance spectroscopy measurements.

## 2. Experimental work

### 2.1. Materials and Methods

Ammonium hydroxide, tetraethoxysilane (TEOS, 99%) and Ethyl alcohol were purchased from Sigma–Aldrich and their aqueous solutions were prepared in deionized (DI) water (resistivity = 18 MΩ cm).

### 2.2. Synthesis of various sizes of silica nanoparticles

The conventional Stober method was utilized to synthesis the silica nanoparticles and the method was further modified for synthesizing different sizes of SiO<sub>2</sub> NPs [24–26]. The modifications on the Stober method were adapted from the work of Hamilton and co-workers [26]. SiO<sub>2</sub> nanoparticles were prepared by adding 0.045 mol L<sup>-1</sup> TEOS, followed by a mixture of 8 mol L<sup>-1</sup> ethyl alcohol, and [H<sub>2</sub>O]/[TEOS] 65.5 and 28 ammonium hydroxide mol L<sup>-1</sup>. The mixture was stirred for 12 hours. It was observed that the mixture turned milky in around 2 min and became dense opaque white in about 5–6 min. Then, it was centrifuged (3000 rpm for 10 min) and redispersed three times in ethanol. The experiments were repeated to prepare three more batches with varying concentrations/volumes of the reagents to get silica nanoparticles of various sizes. The as synthesized SiO<sub>2</sub> NPs of various sizes have been characterized using X-ray diffractometer (XRD) (BRUKER XRD), high resolution transmission electron microscope (HRTEM) (FEI Quanta FEG 200), and UV–Vis absorbance spectroscopy (Perkin Elmer lambda 750). The samples for HRTEM were prepared by

dropping 3 μl of the colloid solution onto copper grids. The average diameter of the SiO<sub>2</sub> NPs was confirmed from the HRTEM images. All the used batches are summarized below (Table 1) to prepare the different sizes of NPs.

### 2.3. Synthesis of ZnO solutions

ZnO was used as an ETL and its preparation procedures were adopted from the literature [27]. Initially, 0.2 M zinc acetate dehydrate solution was prepared in ethanol. The solution was then heated to 80 °C and rigorously stirred for 4 h. Subsequently, mono-ethanolamine (MEA) was added to the solution as a solution stabilizer. Finally, the solution was again heated to 80 °C and magnetically stirred for 24 h for mixing, and then used for ETL deposition using spray-coating technique.

### 2.4. Device fabrication and testing

Devices were fabricated by the method reported in our previous work [19]. However, for the devices with SiO<sub>2</sub> NPs, a fixed weight percentage of SiO<sub>2</sub> NPs of a desired size was incorporated into the P3HT:PCBM or ZnO ETL followed by used for deposition.

## 3. Results and discussion

Fig. 1(a) shows the X-ray diffraction pattern of SiO<sub>2</sub> NPs, whereas Fig. 1(b) shows the UV–Vis absorbance spectra for various sizes (25, 50, 75 and 100 nm) of SiO<sub>2</sub> NPs dispersed in the ethanol. The XRD pattern indicates a broad peak at  $2\theta = 22^\circ$ , which indicates the amorphous nature of the silica nanoparticles [29]. No significant peak or absorption edge was obtained in Fig. 1(b) and this observation was found to be similar with the earlier reported work on SiO<sub>2</sub> NPs [30]. X-ray photoelectron spectroscopy was utilized to determine the surface composition and oxidation states of synthesized SiO<sub>2</sub> NPs (shown in Fig. 1(c)). The Si 2P and O 1S peak is located at around 103.5 and 533.8 eV, respectively. These values are well agreement with the previously reported results [23,29].

Fig. 2 shows the HRTEM micrographs of the different sizes SiO<sub>2</sub> NPs. The average size of these SiO<sub>2</sub> NPs is found to be 25, 50, 75 and 100 nm and is shown in Fig. 2(a)–(d), respectively. All the nanoparticles are separated from each other with uniform particle-to-particle spacing and the surfaces of these NPs are found to be coarse and rough. It is known that the surface morphology and the device interfaces play a crucial role when one wants to achieve high performance from the OSCs. Therefore, atomic force microscopy (AFM) and field emission scanning electron microscopy (FESEM) was used to investigate the surface morphology of the deposited films (as shown in Fig. 3) and cross-sections of the devices (as shown in Fig. 4), respectively. The surface roughness for bare SiO<sub>2</sub> NPs (Fig. 3(a)) and P3HT:PCBM film (Fig. 3(b)) is found to be 10.21 and 1.28 nm, respectively. However, the surface roughness increases to 2.37 nm for the films with SiO<sub>2</sub> NPs incorporated in ZnO (as shown in Fig. 3(c)). Further, for the films in which P3HT:PCBM:SiO<sub>2</sub> NPs photoactive blend is deposited over SiO<sub>2</sub> NPs incorporated in ZnO, the surface roughness was found to be 4.32 nm (as shown in Fig. 3(d)). The increase in the surface

**Table 1**  
The different batches of synthesized silica nanoparticles.

Batch	Ethyl alcohol (mol L <sup>-1</sup> )	NH <sub>4</sub> OH (mol L <sup>-1</sup> )	TEOS (mol L <sup>-1</sup> )	[H <sub>2</sub> O]/[TEOS]	Avg. size (nm)
B-1	8	28	0.045	65.5	25
B-2	8	14	0.012	250	50
B-3	4	14	0.067	209	75
B-4	4	14	0.045	311	100

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