

# Enhancement of Light Absorption in the Active Layer of Organic Solar Cells using Ag:SiO<sub>2</sub> Core-Shell Nanoparticles

Fahendri<sup>1</sup>, Ilham Perdana<sup>1</sup>, Zulfi Abdullah<sup>1</sup>, Mulda Muldarisnur<sup>1\*</sup>

<sup>1</sup>Department of Physics, Faculty of Mathematics and Natural Sciences, Universitas Andalas, Padang, Indonesia.

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Corresponding Author:

Mulda Muldarisnur

[muldarisnur@sci.unand.ac.id](mailto:muldarisnur@sci.unand.ac.id)

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**Abstract:** Organic solar cells still suffer from relatively low conversion efficiency despite significant progress. An Ag:SiO<sub>2</sub> core-shell nanoparticle embedded inside the active layer of an organic solar cell is expected to enhance light absorption. Light absorption within the active layer consisting of 70 nm thickness PEDOT: PSS and P3HT: PCBM was calculated using the finite element method for a variable diameter of the silver core, the thickness of the SiO<sub>2</sub> shell, and the relative position of nanoparticle inside the active layer. The diameter of the Ag nanoparticle varies from 20 to 50 nm, the thickness of SiO<sub>2</sub> varies from 1 to 4 nm, and the position was shifted vertically to 70 nm. The maximum light absorption in the active layer is obtained for silver nanoparticles with a diameter of 40 nm and a SiO<sub>2</sub> thickness of 1 nm. The optimum position of the core-shell nanoparticle was found to be not at the interface between the PEDOT: PSS and P3HT: PCBM layers but a little bit shifted down into P3HT: PCBM layer. The highest increase of light absorption, compared to without Ag:SiO<sub>2</sub>, is 210%, much higher than reported in the literature. Increasing absorption is related to the excitation of surface plasmon resonance of Ag:SiO<sub>2</sub> nanoparticles leading to the local field and scattering enhancement in the active layer of the organic solar cell.

**Keywords:** Organic solar cell; Light absorption enhancement; Finite element method; Ag:SiO<sub>2</sub> nanoparticle; Surface plasmon resonance

## Introduction

Fossil fuels dominate global energy sources with a nearly 80% share (Murdock et al., 2021). In addition to limited availability, fossil fuels are not environmentally friendly, the most significant contributor of greenhouse gases responsible for global warming and health problem. Concerns about sustainability, air pollution, and global warming encourage the development of renewable energy sources with nearly zero emitted harmful gasses (Casper, 2010). Compared to other renewable sources, sun radiation is the most abundant, easily accessible, sustainable, and non-destructive. Annually, the earth receives solar power of 79.000 TW, which is 3000 times the total global energy consumption. In other words, one-hour of radiation power reaching the earth is enough to fulfill yearly needs (Askari, 2014). The current solar energy utilization is still low, but it is estimated to reach 8.300 TW per year in the next 30 years (Perez et al., 2022).

Solar cells or photovoltaics are devices that can convert sunlight energy into electrical energy directly. High manufacturing and installation costs currently prohibit the rapid market penetration of solar cells. This bottleneck can only be solved by developing low-cost, durable, and highly-efficient solar cells. Silicon-based solar cells dominate the market (e.g., 95%) (Riede et al., 2021). Despite having the highest efficiencies, silicon-based solar cells are fragile and expensive due to the needs of the high cost of materials as well as expensive equipment and complex fabrication processes.

Continued efforts have been devoted to obtaining high-efficiency solar cells at lower prices. Organic solar cells (OSC) have attracted the attention of many researchers due to their lightweight, flexibility, semi-transparency, and low manufacturing cost (Shabani et al., 2022). The higher light absorption of organic molecules compared to silicon allows a much thinner active layer. Semi-transparency is appealing for decorative applications in buildings, airplanes, trains, or automobiles. Besides, the flexibility of organic thin films

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enables the manufacturing of solar cells on any flat or curved substrates. Currently available organic solar cells suffer from low efficiency and short lifetimes (Askari, 2014; Chen, 2019; Kim et al., 2012). One of the primary sources of inefficiency is the solar cell's active material's ability to absorb sunlight to create excitons. Low absorption is caused by the higher energy gap of organic molecules than that of inorganic semiconductors and also due to a relatively narrow absorption peak.

There are some strategies to enhance light absorption in organic solar cells, namely by using thicker active material (Chidichimo et al., 2010), tandem structures (Dou et al., 2012), an array of microlenses and mirrors (Tvingstedt et al., 2008), luminescent polymer doped active layer (He et al., 2022), and embedding metallic nanoparticles (Ahn et al., 2016; Feng et al., 2018). A thicker layer is prohibited by a short exciton lifetime and low carrier mobility (Falke et al., 2014). A tandem structure is much more expensive and tedious to fabricate. Enhanced light absorption with an array of microlenses requires careful lens placement with respect to incoming light. On the other hand, the use of luminescent polymer is susceptible to photobleaching resulting in fast degradation.

Metallic nanoparticles are more favorable because they increase light absorption without affecting charge transport from the active layer to the electrodes. In addition, the near-field nature of surface plasmon resonances makes them inherently suitable for thin-film solar cells. Metallic nanoparticles incorporated into solar cells enhance light absorption by two different mechanisms, namely: *first*, stronger scattering that reduces Fresnel reflection on the front surface and increases optical path length inside the active layer, *the second* enhanced electric field around nanoparticles boosts photon absorption (Atwater et al., 2010; Catchpole et al., 2008). The size, composition, and shape of plasmonic nanoparticles and the control of the nanometer distance between particles are critical factors in the design of plasmonic nanostructures to obtain higher absorption enhancement (Kozanoglu et al., 2013; Manzhos et al., 2021).

Several studies have been reported to increase sunlight absorption on OSC by adding metal nanoparticles into the active layer. The efficiency of OSC increases by 7.9% upon the addition of three prismatic nanometal shapes (Phengdaam et al., 2021). The vertical position of spherical silver nanoparticles inside the active layer influences light absorption. The highest absorption, nearly double compared to that without nanoparticles, is obtained when the nanoparticles are at the interface between the P3HT:PCBM and the PEDOT:PSS layers (Liu et al., 2013; Qu et al., 2011). The use of bare metal nanoparticles in the active layer should be done with care because they may act as

recombination centers that deteriorate solar cell efficiency (Rana et al., 2016).

Core-shell silver nanoparticles result in a significant increase in light absorption inside the active layer of an OSC. A 27% increase in OSC efficiency has been reported for Ag:SiO<sub>2</sub> core-shell nanoparticles (N'Konou et al., 2018). An increase of light absorption by 77.9% at a periodicity of 200 nm has been achieved (Perdana et al., 2020; Perdana et al., 2021). In these three works, nanoparticle size and position in the active layer have not been varied. The size and position of the nanoparticles will significantly affect the scattering of electromagnetic waves around the nanoparticles and their absorption by the active layer of an OSC.

Here, we report an attempt to increase the efficiency of OSC by enhancing light absorption in the active layer using Ag:SiO<sub>2</sub> core-shell nanoparticles. We focused on the influence of silver core diameter, SiO<sub>2</sub> thickness, and the position of the nanoparticles in the active layers. We found that metal nanoparticle size, shell thickness, and position of core-shell nanoparticles significantly influence light absorption. The highest obtained absorption enhancement is 210%, much higher than that reported in the literatures.

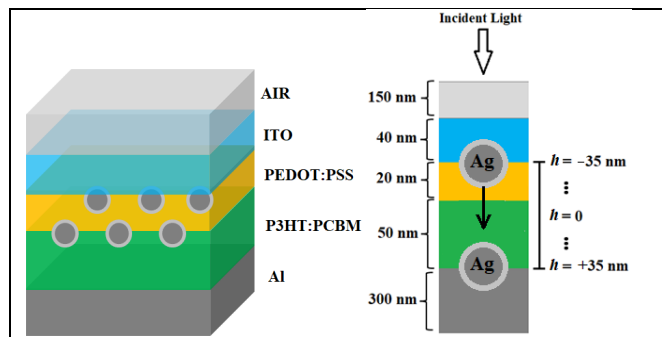
## Method

This research is aimed at increasing light absorption by the active layer of OSC numerically using the finite element method (Humphries, 2010; Jin, 2014). In this method, physical devices are divided into smaller elements where a function will be introduced to express the physical behavior of each element. This method transforms differential equations into a set of algebraic equations that can be written in matrix form. A perfectly-matched layer condition was imposed on the horizontal boundary to prevent non-physical reflection from entering the simulation area. Scattering boundary conditions have been applied for vertical boundaries.

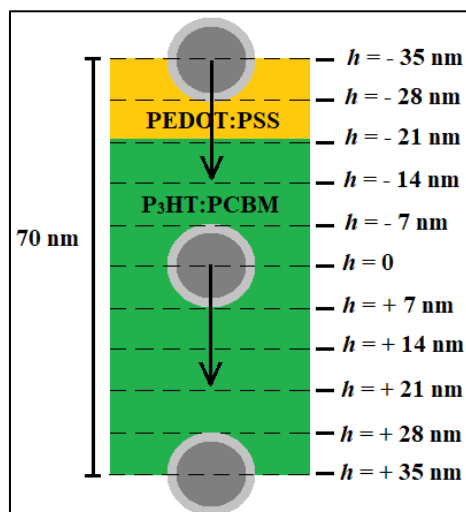
The studied bulk heterojunction OSC consists of ITO (indium tin oxide) layer as a transparent conductor or cathode, PEDOT:PSS (*poly 3,4-ethylenedioxy-thiophene: polystyrene sulfonate*) as a hole transporting layer, P3HT:PCBM (*poly3-hexylthiophene-phenyl-C6-butyric acid methyl ester*) as an active layer, and aluminum as an anode as well as reflector reflecting light into the active layer. The PEDOT:PSS polymeric interfacial layer enhances the cell mechanically and eases the charge extraction process. The P3HT is a p-type semiconductor polymer with high visible light absorption and greatly delocalized electrons. The P3HT and PCBM are commonly used as donor and acceptor materials in the photoactive layer where light is absorbed. Light absorption creates excitons that diffuse inside the material before reaching the donor-acceptor interface,

separating them into electrons and holes (Chidichimo et al., 2010).

The simulated organic OSC structure is shown in Figure 1. The thickness of each layer is 40 nm for ITO, 20 nm for PEDOT: PSS, 50 nm for P3HT: PCBM, and 300 nm for aluminum. The diameter of the silver (Ag) nanoparticle was varied from 20 - 50 nm, and the thickness of the SiO<sub>2</sub> shell was varied from 1 - 4 nm. The vertical position of Ag:SiO<sub>2</sub> core-shell nanoparticle was varied to find maximum absorption enhancement.



**Figure 1.** Organic solar cells structure with embedded Ag:SiO<sub>2</sub> core-shell nanoparticle (left), and the variation in the vertical position of the core-shell nanoparticles in the active layer (right)



**Figure 2.** Detail variation of Ag:SiO<sub>2</sub> core-shell nanoparticle in the organic semiconductor layer

The shift in the vertical position of the nanoparticles was varied along the thickness of the PEDOT: PSS layer with P3HT: PCBM that is 70 nm (See Figure 2). A relatively thin active layer is aimed at reducing exciton recombination. The 10-scale division distance range is 7 nm, with the zero-position located in the middle of the total thickness of the two layers. At the highest position (-35 nm), the nanoparticle penetrates the ITO layer partially. Similarly, the nanoparticle partially penetrates the aluminum layer at the lowest position (+35 nm). The penetration of nanoparticles into ITO or aluminum for the positions in between depends

on particle diameter. Physical and refractive indices data for all layers and nanoparticles needed for simulation were obtained from <http://www.refractiveindex.info> web page. Real and imaginary parts of the refractive index or permittivity were inputted into the simulation.

After solving Maxwell's equation using the finite element method, the dissipated power of the electromagnetic wave is calculated by performing volume integration of and Poynting vector.

$$L(\lambda) = \int^x \int^y \int^z -\nabla \cdot \mathbf{S}(x, y, z, \lambda) dx dy dz \quad (1)$$

where  $L(\lambda)$  is the dissipated power and  $\mathbf{S}$  is the Poynting vector. The absorption  $A(\lambda)$  of the active layer of the organic solar cell is defined as the ratio of the dissipated power to the incoming power from source  $P_{source}(\lambda)$ .

$$A(\lambda) = \frac{L(\lambda)}{P_{source}(\lambda)} \quad (2)$$

The absorption enhancement due to the incorporation of core-shell nanoparticles is defined as the ratio of the total absorbed power over the investigated wavelength range with nanoparticles to the absorption without nanoparticles.

$$AE = \frac{\int_{\lambda_1}^{\lambda_2} (\lambda / hc) A(\lambda) I(\lambda) d\lambda}{\int_{\lambda_1}^{\lambda_2} (\lambda / hc) A_o(\lambda) I(\lambda) d\lambda} \quad (3)$$

where  $A_o(\lambda)$  is the absorption of light without the addition of nanoparticles and  $I(\lambda)$  is the spectrum of solar radiation obtained from available standard data (American Society for Testing and Materials, 1987).

## Results and Discussion

Figure 3 shows absorption enhancement in the active layer of OSC for varying silver particle diameter, silica shell thickness, and vertical position of the nanoparticle. Light absorption enhancement tends to increase when the Ag:SiO<sub>2</sub> core-shell nanoparticle moves vertically into the active layer (P3HT:PCBM). The enhancement of light absorption increases from the position  $h = -35$  nm to  $h = +14$  nm before it decreases when nanoparticles become closer to the aluminum layer. The trend is similar for all nanoparticle diameters. The diameter of silver nanoparticles has a non-monotonous influence on absorption enhancement. The absorption enhancement increases when the silver nanoparticle diameter increases from 20 nm to 40 nm, then decreases for a larger particle diameter. For fixed silver nanoparticle diameter and particle position in the organic semiconductor layer, the absorption

enhancement decreases with the thickness of the SiO<sub>2</sub> shell. The highest enhancement is obtained for a shell thickness of 1 nm. When the nanoparticle is located inside the PEDOT:PSS layer, the absorption enhancement is less than one. In other words, embedding Ag:SiO<sub>2</sub> nanoparticles causes lower light absorption than without nanoparticles.

However, when the diameter of the metal nanoparticle is too large, the scattering type change from Rayleigh to Mie. The intensity distribution of Mie scattering is dominant in the forward direction instead of homogeneously distributed, as in the case of Rayleigh scattering. As a consequence, light is scattered more toward the aluminum electrode and less intensity to be absorbed by the active layer. The second mechanism of higher absorption enhancement for a larger particle is the stronger near-field resonance leading to much higher field confinement around the surface of the metal nanoparticle. According to Mie's theory, cubic dependence of field enhancement with particle size is expected.

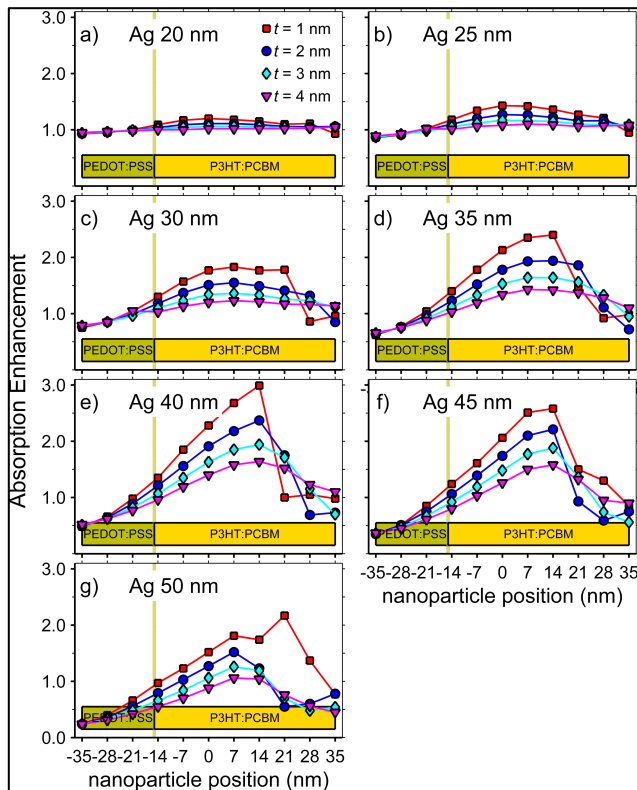


Figure 3. Graph of absorption enhancement as a function of nanoparticle position

The size of the silver nanoparticle core and SiO<sub>2</sub> shell determines the nanoparticle's scattering and absorption cross-sections. The absorption is dominant for nanoparticles having a diameter of less than 30 nm. On the other hand, instead of absorbing, larger particles tend to scatter light. Far-field scattering causes the lengthening of the optical path, increasing the absorption of the incoming light (Baek et al., 2013). These facts explain why the absorption enhancement in the active layer of OSC is much lower for small metal particles. When the nanoparticle size is too small, it is less helpful in extending the optical path in the active layer. Therefore, light absorption is enhanced slightly by the presence of the nanoparticle. Small nanoparticle absorbs photon to excite surface plasmon resonance on their surface, leaving fewer photons to be absorbed by the active layer.

When particles become bigger, light absorption by the metal particle decreases and is more scattered around the nanoparticle resulting in a higher light absorption enhancement in the active layer of OSC.

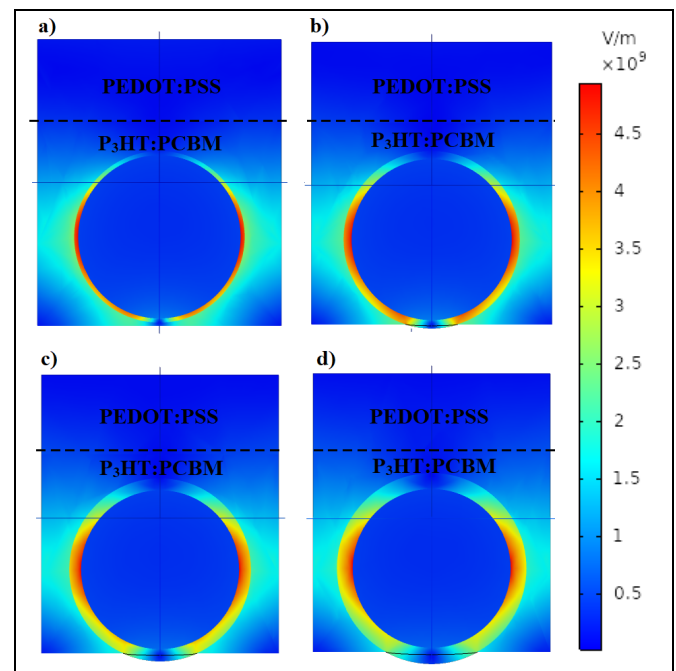
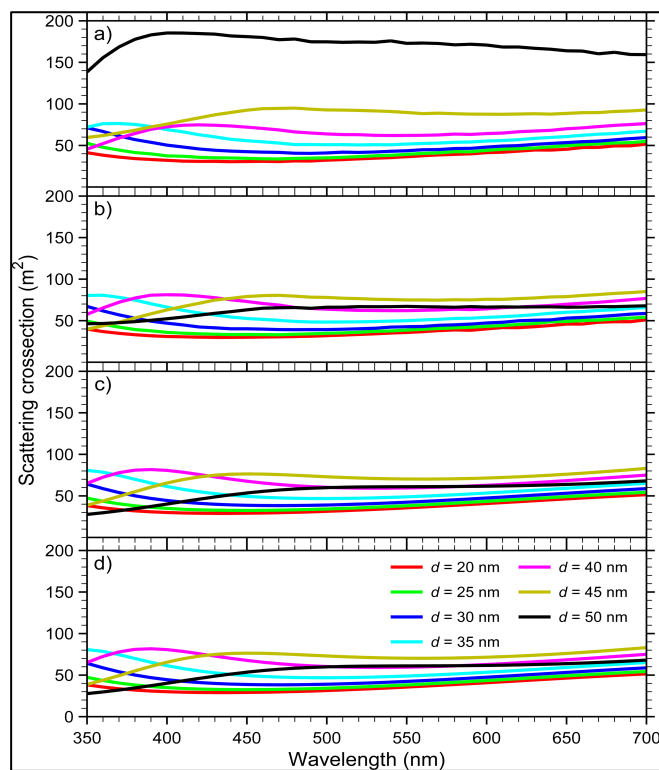


Figure 4. Electric field distribution for Ag nanoparticles with a diameter of 40 nm at the optimum position  $h = +14$  nm with variations in SiO<sub>2</sub> thickness (a) 1 nm, (b) 2 nm, (c) 3 nm, (d) 4 nm

Light absorption enhancement is lower when Ag:SiO<sub>2</sub> is located inside PEDOT:PSS layer. The reason is obvious because the active layer of OSC is P3HT:PCBM. PEDOT:PSS serves as a charge transporting layer after excitons are separated into electrons and holes. Since the enhanced field excited by surface plasmon resonance and the scattering field are concentrated in the PEDOT:PSS, the P3HT:PCBM would absorb less incident light than the OSC without nanoparticles. When the nanoparticle is located in the active layer, light absorption enhancement increases until the optimum value, around 14 nm. The increase is related to stronger scattered light in the active layer and near-field enhancement around the nanoparticle. When the nanoparticle gets closer to aluminum, absorption enhancement decreases. The decrease is related to less

scattering around the nanoparticle in the active layer and a smaller fraction of field localization near the nanoparticle's surface. This is more obvious when particle size is larger than 35 nm. At a position larger than +14 nm, Ag:SiO<sub>2</sub> nanoparticles penetrate partially into the aluminum surface. Particle penetration reduces the area in contact with the active layer, reducing light absorption enhancement.

The thickness of the SiO<sub>2</sub> enclosing the silver nanoparticle determines the decay of the evanescent field from the surface of the silver nanoparticle into the active layer. A thicker shell reduces the electric field penetrating the active layer, reducing the light absorption enhancement. The highest light absorption enhancement is obtained for the thinnest silica thickness, which is 1 nm, independent of particle position and silver core diameter. The distribution of the electric field around the nanoparticle is shown in Figure 4. The electric field distributed around Ag:SiO<sub>2</sub> nanoparticle is much higher when the SiO<sub>2</sub> layer is 1 nm, as shown as a denser red-colored field, and decreases when the thickness is increased to 4 nm.



**Figure 5.** scattering cross-section of 40 nm silver nanoparticles at the optimum position  $h = +14$  nm for different thicknesses of SiO<sub>2</sub> shell: (a) 1 nm, (b) 2 nm, (c) 3 nm, and (d) 4 nm

The scattering cross-section of Ag:SiO<sub>2</sub> for different silver particle diameters and silica shell thicknesses is shown in Figure 5. The absorption peaks are in the violet range of visible light. A red shift of the scattering peaks with diameter is expected. The absorption enhancement is proportional to the area below the scattering curve.

The highest scattering occurred for each nanoparticle size when the thickness of the SiO<sub>2</sub> shell was 1 nm. The scattering cross-section decreases with the shell thickness. An unusually high scattering of 50 nm silver particle at a shell thickness of 1 nm is caused by partial penetration of the particle into the aluminum layer.

### Conclusion

Embedding metal nanoparticles into the active layer of an OSC increases light absorption. The diameter of the silver nanoparticle, the thickness of the SiO<sub>2</sub> shell, and the position of the Ag:SiO<sub>2</sub> core-shell nanoparticle in the organic semiconductor layer influence light absorption significantly. The optimum diameter of the silver nanoparticle is 40 nm corresponding to large scattering, less metal absorption, low light blocking, and significant field enhancement around the nanoparticle. Maximum absorption enhancement is obtained for a thin SiO<sub>2</sub> shell that is 1 nm. Thicker shell results in a faster decay of the electric field into the active layer of OSC. The decay causes a lower absorption enhancement. The Ag:SiO<sub>2</sub> must be positioned deeper in the P3HT:PCBM active layer to obtain the highest absorption enhancement and not too close to the aluminum layer. The optimum position is +14 nm from the center of organic semiconductor layers. Under the optimum condition and absorption enhancement, up to 210% can be achieved. High absorption enhancement OSC will directly correspond to a higher conversion efficiency of solar radiation.

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