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Efficiency improvement of Si solar cells using metal-enhanced nanophosphor fluorescence

J.-Y. Chen, C.K. Huang, W.B. Hung, K.W. Sun*, T.M. Chen

Department of Applied Chemistry, National Chiao Tung University, Hsinchu 30010, Taiwan

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ABSTRACT

In this study, Eu^{2+} -doped barium silicate ($\text{Ba}_2\text{SiO}_4:\text{Eu}^{2+}$) nanophosphors dispersed in a surfactant solution were spin-coated on commercially available silicon solar cells to form colloidal crystals on the surface. The crystals then act as luminescence down-shifting centers to generate low-energy photons for incident ultraviolet light. The fluorescence from the $\text{Ba}_2\text{SiO}_4:\text{Eu}^{2+}$ nanophosphors was further enhanced by coating a metal-enhanced layer composed of Ag nanoparticles and a SiO_2 spacer. The solar cells showed an enhancement of 0.86 mA/cm^2 in short-circuit current density and approximately 0.64% increase in power conversion efficiency when coated with nanophosphors, SiO_2 spacers, and Ag nanoparticles. The properties of cells integrated with the metal-enhanced layer were characterized to identify the roles of nanophosphors and Ag nanoparticles in improved light harvesting. These experiments demonstrated that the colloids of $\text{Ba}_2\text{SiO}_4:\text{Eu}^{2+}$ acted as luminescence down-shifting centers in the ultraviolet region and the metallic nanoparticles also helped to enhance fluorescence in the visible region to increase light absorption within the measured spectral regime.

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1. Introduction

The maximum efficiency of a single-junction solar cell is constrained by the Shockley–Queisser limit [1], which defines the maximal output power as a function of the bandgap in a solar cell. Limitations set on the maximum efficiency of the cells are due to the loss mechanisms in solar cells, such as sub-bandgap-energy photon loss [2] and thermalization of charge carriers caused by the absorption of high-energy photons with energies larger than the bandgap of the solar cell. These fundamental losses directly lead to an efficiency limit of approximately 30% for a material with a bandgap of 1.1–1.3 eV under non-concentrated Air Mass 1.5 (AM 1.5) illumination [1].

Various schemes have been proposed to overcome this fundamental efficiency limit for single-junction solar cells [3–5]. However, these steps are either difficult to implement or expensive to mass produce. One way to surpass this limit is using a luminescence down-shifting (LDS) process to absorb high-energy photons and re-emit them at longer wavelengths such that the photovoltaic device exhibits a significantly better response. The application of an LDS layer was first demonstrated in the late 1970s to improve the poor spectral response of solar cells to short-wavelength light. The LDS layer can help harvest full solar energy

by expanding the operating spectral range toward the ultraviolet (UV) range. A recent theory demonstrated that a conversion efficiency of 38.6% can be achieved under unconcentrated sunlight using a silicon solar cell with an ideal LDS material [6]. Detailed reviews of the progress in this area can be found in Refs. [7–14].

An increase of 0.3% (absolute) was reported when an organic luminescent dye is included in the ethylene vinyl acetate (EVA) encapsulation layer of multicrystalline silicon photovoltaic modules [15]. Thermodynamic evaluation and analysis involving both lossless and lossy theoretical limits have further demonstrated that 7% efficiency improvement in efficiency can be achieved using a perfectly down-converting material [16]. The down-conversion process increases the short-circuit current while retaining the open-circuit voltage. For semiconductor nanoparticles, the high surface-to-volume ratio may possibly create a high concentration of mid-level trap states and coalesce into a band that function in doubly radiative emission processes in an efficiency manner [5,16,17]. Therefore, the increase in surface traps could be advantageous for the LDS process for the nanophosphors as well. Furthermore, if nanoparticles are embedded within the top layer of the solar cells, then the scattering becomes inconsequential and the inherent scattering property in a down-converting layer could be solved. This becomes an advantage for any three-dimensional configuration of the down-conversion layer [16]. For example, YVO_4 nanophosphors were directly integrated into the textured solar cell surface without host materials. The proposed hybrid system showed a 4% enhancement in short-circuit current density

* Corresponding author. Tel.: +886 3 5712121.

E-mail address: kwsun@mail.nctu.edu.tw (K.W. Sun).

and approximately 0.7% in power conversion efficiency using AM 1.5 illumination [18].

At the other end of the spectrum, interactions of metallic surface or particles are attracting increased interest. Near-field interactions are those occurring within a wavelength distance of an excited phosphors/fluorophores [19,20]. The spectral properties of fluorescence materials can be altered by near-field interactions with the electron plasma present in metals by several methods that have not been used in classical fluorescence experiments. Experimental results showing the effects of fluorophore-to-silver distance on the emission of cyanine-dye-labeled oligonucleotides suggest that silver island films yield improved fluorescence measurements and maximum fluorescence enhancements occur at approximately 9 nm from the metal surface [21]. The effects of metals on fluorescence are explained using a concept based on radiating plasmons [22]. Wavenumber matching considerations can be used to predict whether fluorophores at a given distance from a continuous planar are emitted or quenched. A twofold enhancement in structured fluorescence is also observed in perylene in close proximity to silver island films [23]. The emission of cerium-doped yttrium aluminum garnet (YAG:Ce) phosphor thin films can be enhanced by surface plasmon coupling to a suitably nanostructured silver-island layer. The photoluminescence intensity is significantly enhanced by capping Ag islands with a 10 nm-thick SiO₂ layer as a dielectric medium [24].

The current study demonstrates the increase in conversion efficiency of Si solar cells by combining rare-earth element-doped barium silicate (Ba₂SiO₄:Eu²⁺) nanophosphors, Ag NPs, and SiO₂ dielectric spacer directly onto the textured surface of the devices. Ba₂SiO₄:Eu²⁺ is a widely used optical material with many good features, including excellent thermal efficiency and high luminescent quantum efficiency [25,26]. UV or visible excitation for Ba₂SiO₄:Eu²⁺ nanophosphors produces broadband luminescence with peak wavelengths of 505 nm. Therefore, Ba₂SiO₄:Eu²⁺ phosphors are promising UV-absorbing spectral converters for solar cells because they possess broadband absorption throughout the entire UV region (300–450 nm) and they enable the emission of intense visible lights. Thus, these materials are well suited for spectral conversion in solar cells. The current study aims to investigate the quantum efficiency enhancement associated with metal-enhanced light harvesting and energy transfer properties of Ba₂SiO₄:Eu²⁺ nanophosphors.

2. Experimental

Fig. 1 shows a schematic diagram of the synthesis of a micro-textured cell surface covered with nanophosphors, SiO₂ dielectric layer, and Ag NPs (Ba₂SiO₄:Eu²⁺/SiO₂/Ag NPs). The crystalline Si (c-Si) solar cell device used was manufactured following the

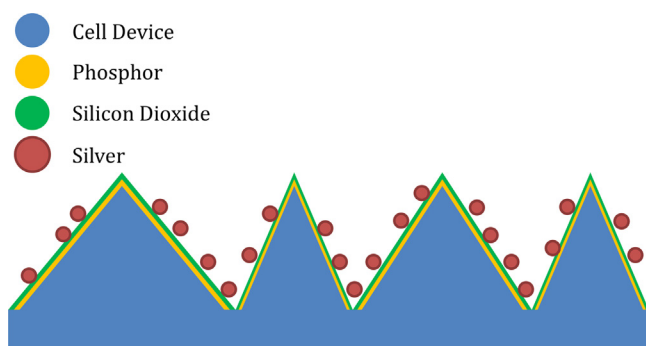


Fig. 1. Schematic of textured cell structures covered with Ba₂SiO₄:7% Eu²⁺ nanophosphors, SiO₂ layer, and Ag NPs.

procedures reported in Ref. [27]. Given that incident UV radiations mostly produce electron–hole pairs near the device surface, photo-generated carriers easily disappear by recombining with surface defects, which can lead to inferior cell efficiency. However, in the presence of the Ba₂SiO₄ nanophosphors on the front side, more photons can be absorbed closer to the depletion region when the UV photons are luminescence down-shifted to the visible region. The built-in electric field automatically separates the photo-generated electron–hole pairs and enhances the photovoltaic effect.

Barium nitrate (Ba(NO₃)₂), europium nitrate pentahydrate (Eu(NO₃)₃·5H₂O), tetraethyl orthosilicate (TEOS), and ethylene glycol (EG) were purchased from Sigma-Aldrich. Citric acid (CA) and 28% ammonium hydroxide (NH₄OH) were supplied by Showa (Chemical Co., Ltd., Tokyo, Japan). Ag NPs dissolved in deionized water at a concentration of 100 ppm and average diameters of 10–20 nm were purchased from Particular GmbH. All reagents were used as received.

(Ba_{1-x}Eu_x)₂SiO₄ (0 ≤ x ≤ 0.09) compounds were synthesized by the typical Pechini method. Stoichiometric amounts of Ba(NO₃)₂, Eu(NO₃)₃·5H₂O, and CA were dissolved in deionized water. Subsequently, TEOS mixed in ethanol and EG were added to the mixture solution under vigorous stirring, and the pH was adjusted to 2 with NH₄OH. The molar ratio of CA to cations and to EG was 4:1:2. The solution was heated on a hot plate at 80 °C for 3 h to yield a transparent solution, and then the temperature was increased to 150 °C for water evaporation. When the solution turned into a viscous brown gel, it was dried at 100 °C in air overnight. A fine precursor powder was produced by calcining the dry gel in air at various temperatures (250–500 °C) for a few hours. After eliminating organic content, the porous powder was ground and annealed at 900 °C for 6 h in reducing atmosphere (5% H₂+95% N₂). Then, Ba₂SiO₄:7% Eu²⁺ phosphor with pale-green appearance was obtained.

To determine the optimum SiO₂ film thicknesses and nanophosphor concentration for maximizing the fluorescence intensity in the visible region, the Ba₂SiO₄:Eu²⁺/SiO₂/Ag NPs layers were first fabricated on a planar Si surface. The Ba₂SiO₄ nanophosphors were dispersed in ethanol to obtain a 1, 2, and 3 mg/ml solutions, respectively. Spin-coating was used for nanophosphor deposition. Using 1, 2, and 3 mg/ml of the Ba₂SiO₄ nanophosphors, uniform and perfect-light-scattering thin films with a thickness of approximately 100 nm were obtained under properly controlled spin-coating rates. Six samples with SiO₂ spacer thicknesses ranging from 5 nm to 100 nm were deposited onto the nanophosphor layer using a plasma-enhanced chemical vapor deposition (PECVD) system followed by spin-coating of the Ag NPs. The decay times of the metal-enhanced and down-shifted photoluminescence were measured using a time-correlated single photon counting (TCSPC) system with spectral resolution and system response of 0.1 meV and 300 ps, respectively. To determine the optimal particle density of the nanophosphors, electrical characterizations on solar cells coated with nanophosphors of different densities were performed by varying the concentrations of the nanophosphors solution from 1 mg/ml to 3 mg/ml.

After optimal parameters were determined, the integrated Ba₂SiO₄:Eu²⁺/SiO₂/Ag NPs layers were fabricated on textured c-Si solar cells. The c-Si cells with and without Ba₂SiO₄:Eu²⁺/SiO₂/Ag NPs layers were evaluated at room temperature based on the illuminated current density versus voltage (*J*-*V*) characteristics, external quantum efficiency (EQE), and reflectance. Photocurrent was analyzed using a solar simulator under the Air Mass 1.5 Global (AM 1.5G) illumination condition (100 mW/cm², 25 °C). The EQE was measured using an AM 1.5G standard spectrum and an Optosolar simulator (SR-150). The reflectance spectra of the samples were recorded using a UV–vis–NIR spectrophotometer

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