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# Efficiency enhancement of silicon solar cells through a downshifting and antireflective oxysulfide phosphor layer



Solar Energy Material

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## ABSTRACT

A simple, rapid, and low-cost anti-reflection coating (ARC) for polycrystalline silicon (pc-Si) solar cells has been developed by spin coating a solution of submicron spherical  $Gd_2O_2S:Eu^{3+}$  phosphor impregnated in a polyvinylpyrrolidone (PVP) matrix on the textured surface of the solar cell. Under a simulated one-sun illumination, this predesigned device achieved an enhancement of 6.74 mA/cm<sup>2</sup> in short-circuit current density and 2.76% in power conversion efficiency relative to those for a pristine cell. Based on photoluminescence, reflectance, and optical spectral studies, the underlying mechanism for these improvements can be attributed to the luminescent down-shifting (LDS) capability in the ultraviolet range and additional promotion of light absorption within the measured spectral region. Our results indicate that a suitable inorganic phosphor impregnated in a polymer matrix may potentially serve as a feasible solution to compensate for the spectral mismatch and Fresnel reflection loss of silicon solar cells.

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#### 1. Introduction

During the past few decades, there has been a gradual realization that fossil energy resources on earth are not infinite, and this has led to a search for clean and renewable sources of energy. Solar power, the flow of radiation from the sun, is a green and virtually sustainable energy source. Currently, the most widely used commercial photovoltaic cells are fabricated with silicon, but this does not guarantee that silicon is an ideal material for optoelectronic devices. In fact, silicon has two major intrinsic drawbacks: (i) the spectral mismatch loss, i.e., the non-absorption photons with energy below the band gap ( $E_g = 1.12 \text{ eV}$ ) and the strong thermalization of charge carriers generated by absorption of photons with energy above the bandgap [1]; (ii) the high Fresnel reflection loss due to the high contrast of refractive indices between air and silicon [2]. Typically, coating an anti-reflective layer of  $SiN_x$  is one of the most common methods to suppress Fresnel reflection, yet this layer requires expensive chemical precursors and fabrication apparatus with high-vacuum capability. Hence, using silicon nitride to serve as an ARC is not a cost-effective solution and the problematic spectral mismatch loss cannot be resolved.

Recently, several innovative anti-reflection techniques have been developed, such as surface texturing [3,4], decoration with metal nanoparticles [5–7], and induction of a nanowire or nanotip

\* Corresponding author. E-mail address: tmchen@mail.nctu.edu.tw (T.-M. Chen). structure [8-10]. Surface texturing involves the introduction of regular or random features by dry or wet etching processes. This texturing, can be successfully used to trap light and leads to an elongation in optical path length, but the low reproducibility and high cost result in such solar cells being barely competitive with fossil fuels. Metal nanoparticles have large scattering cross section to enhance light absorption in the absorbing layer of the solar cells because of their unique localized surface plasmon resonances. However, Ag and Au nanoparticles are the most widely used materials, which make this modification impractical for largescale solar cells due to the scarcity and high price of Ag and Au. Furthermore, metal nanoparticles always show extraordinary absorption in their wavelength range of surface plasmon resonances, which decreases the overall performance of solar cells. Nanowire or nanotip structures provide excellent broad band antireflection characteristics but also greatly increase the probability of surface recombination in parallel.

Lanthanide oxysulfides are well-known phosphor hosts and have been extensively investigated because of their excellent features, including low toxicity, non-deliquescence, easy fabrication, and high thermal stability [11,12]. Europium-doped gadolinium oxysulfide ( $Gd_2O_2S:Eu^{3+}$ ) is our material of interest owing to its broad absorption band in the UV region and visible emission in response to these stimuli. According to the literature [12], upon UV excitation, the  $Gd_2O_2S:Eu^{3+}$  phosphor emits strong red light in the wavelength range where silicon solar cells exhibit a robust spectral response. This suggests that  $Gd_2O_2S:Eu^{3+}$  may be a promising candidate for use as a spectral converter in silicon solar cells. In addition, to deposit the Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> phosphor on the textured surface of silicon solar cells and eliminate surface reflection of silicon over a broad spectral wavelength, polyvinylpyrrolidone (PVP), with a proper refractive index ( $n \approx 1.54$ ) and widely used as a stabilizer for improving the stability of a suspension and the homogeneity of deposited films [13,14], was chosen as the adhesive and matrix for Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup>. Therefore, we could expect such inorganic-in-organic composite layer to be a suitable solution that is capable of resolving the spectral mismatch and Fresnel reflection of silicon solar cells.

In this research, we prepared submicrometer Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> spheres through a simple urea-based homogeneous precipitation and subsequent annealing process. Then the particle size of the  $Gd_2O_2S:Eu^{3+}$  can be finely tuned from 125 to 419 nm by adjusting the conditions of nucleation. The ARC of PVP/Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> was formed by dispersing the phosphor particles in the PVP aqueous solution and spraying the mixture onto the textured surface of pc-Si solar cell through a spin-coating technique. Finally, we investigated the photovoltaic characteristics of the devices with and without a PVP/Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> coating and found a significant enhancement in photocurrent and power conversion efficiency. Based on reflectance and external quantum efficiency measurements, we showed that these enhancements can be ascribed to photon down-shifting and promotion of light absorption. This work establishes the possibility of a lower cost ARC composed of the combination of an inorganic phosphor and a polymer matrix to overcome the poor efficiency of silicon solar cells. Moreover, this ARC can be applied to other related solar photovoltaic devices.

### 2. Experimental

The solar cells used in current work were commercially available pc-Si solar cell wafers without the Si<sub>3</sub>N<sub>4</sub> anti-reflection layer, and for convenience the cell was then cut into smaller pieces (26 mm × 52 mm) by high-power pulsed laser. Gadolinium nitrate hexahydrate Gd(NO<sub>3</sub>)<sub>3</sub>·6H<sub>2</sub>O, europium nitrate pentahydrate Eu (NO<sub>3</sub>)<sub>3</sub>·5H<sub>2</sub>O, polyvinylpyrrolidone K30 (PVP-K30) and urea (NH<sub>2</sub>)<sub>2</sub>CO were purchased from Sigma-Aldrich. All reagents were used without further purification.

In a typical procedure, the synthesis of  $Gd_2O_2S:xEu^{3+}$ ( $1 \le x \le 9\%$ ) phosphor was carried out in two steps: the first step was the preparation of monodisperse  $Gd(OH)CO_3:Eu^{3+}$  colloid spheres via a urea-based homogeneous precipitation method described by Matijević and Hsu [15] with some modifications, the second step was the sulfurization of hydroxycarbonate particles. For the synthesis of Gd(OH)CO<sub>3</sub>:Eu<sup>3+</sup>, stoichiometric amounts of Gd(NO<sub>3</sub>)<sub>3</sub>· 6H<sub>2</sub>O and Eu(NO<sub>3</sub>)<sub>3</sub>· 5H<sub>2</sub>O were dissolved in deionized water. Urea was added into the above solution under magnetic stirring until a transparent solution was formed. The total volume of the solution varied from 70 to 210 ml, while the molar ratio of urea/cations was kept at 50. The mixture was then heated at 80 °C and aged for 80 min in the water bath. The obtained suspension was collected by centrifugation, and the supernatant solution was discarded. The solid phase was washed with ethanol and deionized water several times and then dried in an oven at 80 °C overnight. For sulfurization of the precursor particles, the dried Gd(OH)CO<sub>3</sub>:Eu<sup>3+</sup> powder was placed into a quartz tube and annealed at 900 °C for 8 h in a Ar–H<sub>2</sub>S atmosphere (85–15 vol%). Finally, the Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> with white appearance was obtained.

To determine the best combination between pc-Si solar cell and Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> phosphor and to investigate the influence of particle size of  $Gd_2O_2S:Eu^{3+}$  on the solar cell performance, we therefore first dispersed the photoluminescence (PL) intensity-optimized phosphor of various particle sizes in the aqueous PVP solution, followed by ultrasonication, and vigorous stirring at room temperature for 48 h. The particle density of Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> phosphor was fixed at 5 mg/ml, while the concentration of PVP was kept at 0.06 g/ml. The ARC coated solar cells were fabricated by spin coating the as-prepared mixture solution on the cell textured surface under appropriately controlled spin-coating rate. In addition, the solar cells with solely PVP coating were also fabricated for comparison. In addition to the size optimization of Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> sample, the particle density of Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> is another important factor in this research. Hence, a series of mixture solution of the size-optimized Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> with 2.5, 5, 7.5, and 10 mg/ml particle densities was prepared and then span coated on the solar cells. The spin-coating fabrication process of PVP/Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> modified pc-Si solar cell is summarized in Fig. 1.

All solar cell devices with and without PVP/Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> coating were evaluated at room temperature on the illuminated current density versus voltage characteristics, the reflectance, and the external quantum efficiency (EQE). The photocurrent was measured by a solar simulator (Newport-91190) under the Air Mass 1.5 Global (AM 1.5G) illumination condition (100 mW/ cm<sup>2</sup>, 25 °C). The reflectance spectra were recorded with a double-beam ultraviolet–visible (UV–vis) spectrometer (Hitachi-U3010). The EQE was analyzed using a monochromator-based spectral response system calibrated with a reference cell (Titan Electro-Optics, QE-3000). The phase purity of the reaction product was identified by powder X-ray diffraction (XRD) analysis using a

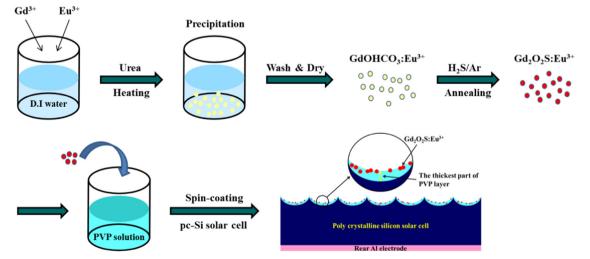


Fig. 1. Flow diagram describing the preparation of Gd<sub>2</sub>O<sub>2</sub>S:Eu<sup>3+</sup> phosphor and its integration on the surface of pc-Si solar cell.

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