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Nanostructured organosilicon luminophores in highly efficient luminescent down-shifting layers for thin film photovoltaics

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ABSTRACT

We use a new type of solution processed luminescent molecules – nanostructured organosilicon luminophores (NOLs) – in printed luminescent down-shifting layers for photovoltaic light harvesting. We show that NOLs exhibit a high luminescent efficiency of 82–90% when added into two chemically different polymer matrices that are commonly used for photovoltaic encapsulation: polyvinyl butyral and ethylene-vinyl acetate. The coated layers are optimized to maximize both UV absorbance and visible light emission and transmittance. Attaching the luminescent layers onto a CIGS solar cell significantly improves the external quantum efficiency in the UV region: from 1% to 55% at 360 nm. As a result, the short circuit current density and power conversion efficiency increase by up to 3.2–4.3%. After experimental verification of our optical simulation model, we employ it to determine the ideal molecular structure of NOLs for luminescent down-shifting applied to CIGS.

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1. Introduction

Single junction solar cells have a maximum theoretical efficiency of 31% [1]. One reason for this low efficiency is lattice thermalization losses in the UV [1]. UV and near-UV photons have energies higher than the band gaps of the semiconductors used in photovoltaics, and so when these photons are absorbed, the excess energy is lost via thermalization. In addition, high energy photons are lost due to front surface recombination and parasitic absorption. Luminescent down-shifting (LDS) layers solve these problems by absorbing UV light and emitting visible light that solar cells can use more efficiently [2]. Thus when applied on top of a solar cell, LDS layers tailor the incident spectrum for more efficient current generation, and in doing so improve solar cell efficiency.

LDS layers have been successfully applied to silicon [1,3–5], CdTe [6–8], CIGS [9–11], dye-sensitized solar cells [12,13], and various other types of photovoltaic technologies. CIGS (copper–indium–gallium–(di)selenide) solar cells are of particular interest here because they perform well in visible wavelengths, but have poor efficiency in UV due to parasitic absorption in the cadmium sulfide buffer layer [9]. Applying LDS layers can be an inexpensive, purely optical means of increasing the efficiency of CIGS and other types of thin film photovoltaics.

Three classes of luminescent materials have been used for LDS: organic dyes, quantum dots, and rare earth-doped inorganic particles (phosphors) [2,14]. Organic dyes are a popular material, as they exhibit photoluminescence quantum yields (PLQY) of ~90–99%, high absorptivities, and are easily embedded in polymer matrices [2,15,16]. However, these properties come at the cost of narrow absorption bands and uncertain UV stability [2,15,17]. Quantum dots (QDs) have been investigated as an alternative

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[18,19]. Although QDs offer broader absorption ranges, tunable emission wavelengths and good UV stability, they also (with a few exceptions [20,21]) have quantum yields of $<60\%$, high re-absorption, and aggregate in polymer matrices [2,14,16,17]. Phosphors, meanwhile, are stable and have high PLQY and broad absorption, but large particle sizes cause scattering and subsequent losses for solar cells [10].

Combining the best properties of QDs and organic dyes while reducing their drawbacks could result in a new, better-performing class of LDS materials. Organometallic complexes, which consist of organic molecules and rare-earth ions, are an example of such an approach [22,23]. Yet energy transfer between the donor and acceptor is often problematic, resulting in low quantum yields. There is therefore still much room for improvement.

Nanostructured organosilicon luminophores (NOLs) are a candidate for overcoming the issues that "traditional" luminescent materials face. NOLs combine the properties of quantum dots and organic luminophores to offer not only high PLQY and absorptivity, but also a broad absorption spectrum, tunable absorption and emission characteristics, and a large pseudo-Stokes shift [24–27]. NOLs have been used for plastic scintillators [24], and similar molecules have found applications in luminescent solar concentrators [28] and photodetectors [29,30]. The optical characteristics of NOLs are potentially ideal for luminescent down-shifting in PV as well. Solution-processable LDS layers are of particular interest because they are easily up-scalable for industrial fabrication. Ideally, NOLs should be embedded into a commonly used encapsulation polymer such as polyvinyl butyral (PVB) or ethylene-vinyl acetate (EVA) with layer thicknesses of $\sim$300–500 μm. Such NOL-based LDS layers would then be suitable for the encapsulation of solar modules.

We therefore address the following question: how well do NOLs perform in combination with polymers as solution-processed LDS layers? We compared the performance of NOLs in polar PVB and less polar EVA layers. We optimized the thicknesses and concentrations of these coatings to achieve the best optical properties for high performance LDS layers. Furthermore, we determined the PLQY of NOLs in both solution and solid state. Next we applied the layers to a CIGS solar cell, which significantly increased the external quantum efficiency (EQE) of the device in the UV region. We also used an optical model to simulate the change in EQE of a CIGS cell with NOL layers. Finally, we computed the optimal absorption spectrum of NOLs, which can serve as a guideline for further synthesis developments. From our work, it is clear that NOLs are a competitive new luminescent down-shifting material.

2. Experimental

2.1. Materials

Nanostructured organosilicon luminophores, type NOL15, (LumInnoTech LLC, Russia) were embedded in a transparent polymer binder of PVB (Butvar B-98, Eastmann, Germany) or EVA (ETIMEX Primary Packaging GmbH, Germany). PVB was dissolved in benzyl alcohol (Sigma Aldrich, Germany), purity $\geq 99\%$, while EVA was dissolved in toluene (Sigma Aldrich, Germany), purity $\geq 99.5\%$.

2.2. Processing of layers

A pure PVB solution was prepared by dissolving polymer flakes in benzyl alcohol at weight percentages of 22.5% and 77.5%, respectively. The flakes were completely dissolved after 12 h of stirring at room temperature, yielding a clear solution with a viscosity of $\sim$12 Pa*s. For the EVA solution, small pieces of solid polymer sheets were dissolved in toluene at 16 and 84 wt%, respectively. After stirring for 24 h at room temperature, the EVA solution was allowed to rest for an additional 24 h, resulting in a clear, homogeneous solution of $\sim$1 Pa*s viscosity. To prepare final solutions for coating, the NOL powder was either (1) dissolved in benzyl alcohol at 150 °C and mixed into the PVB solution at concentrations of 0.014–0.017 wt% NOL; or (2) dissolved in toluene and mixed into the EVA solution at concentrations of 0.015–0.029 wt%. Optimized PVB layers were 0.016 wt% NOL and 300–360 μm thick with an OD of 1.5 at the absorption maximum. Optimized EVA layers were 0.027 wt% NOL and 120–170 μm thick with an OD of 1.4.

Float glass (Weidner Glas GmbH, Germany) with dimensions of $25 \times 25 \times 1$ mm^3 was used as the coating substrate. To prepare the layers, solutions were coated via doctor blading with a coating gap of 450 μm between the blade and the glass substrate at room temperature. Coating speed was kept constant at 2.5 mm s^{-1} to reduce shear stress. Coated PVB samples were dried overnight at 80 °C in a drying oven, while EVA layers were dried for 15 min at 30 °C followed by 30 min at 40 °C and overnight at 80 °C.

2.3. Characterization methods

A FP-8500 Fluorescence Spectrometer (JASCO Inc., United States) was used to obtain absorption, emission, and excitation spectra of the dyes in solution and in layers. The fluorometer was also used to measure the absorption spectra of strongly luminescent samples, running the monochromators synchronously.

For total transmittance, a Perkin Elmer Lambda 950 double beam spectrometer was used. The setup included a 150 mm integrating sphere with a photomultiplier and InGaAs detector. The samples were oriented so that the light beam entered the glass first and then the luminescent layer for all measurements.

The photoluminescence quantum yield of NOLs in solution was determined by measuring the fluorescence of optically dilute solutions [31]. Five standards were used: p-terphenyl, 2,5-diphenyloxazole, anthracene, 1,4-bis-(5-phenyloxazole-2)benzene and rhodamine 6G. The resulting quantum yields are the averages of comparison to all five standards.

To measure PLQY of the layers, a home-built "absorption" setup was used. It was designed after a prototype described in detail by Wilson and Richards [32]. The setup consists of a light source and an integrating sphere coupled with an optical fiber leading to a monochromator. The samples were excited with a standard 5 mm, 16 mW LED (Roithner Lasertechnik GmbH, Austria) with a wavelength of 375 nm (20 mA). A 4-port PTFE-based integrating sphere, 3.3 in. (Newport Corp., USA), with a 0° 1.5 in. port for excitation light was used. The sample holder was designed to hold the sample in the middle of the integrating sphere at an angle of 8°. An iHR 320 with Si CCD-based detector, spectral range 200–1100 nm (Horiba Ltd., Japan) was used to measure the signal. For calibration of the setup, a calibrated Xenon light source L7810–02 (Hamamatsu Photonics K. K., Japan) with WG 295 filter (Schott AG, Germany) was utilized. Fig. 1 shows the final spectrum taken for PLQY determination. Layers of pure PVB and EVA on glass with the same thicknesses as the measured samples were used as references. The resulting PLQY values are the averages of ten samples each.

Layer thicknesses were measured with a Nikon Eclipse L150 confocal microscope. Samples were scratched all the way to the substrate and the difference in levels was analyzed.

The EQE of the solar cells was measured with an Enlitech QE-R Setup (Enli Technology Co. Ltd., Taiwan).

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