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Printable luminescent down shifter for enhancing efficiency and stability of organic photovoltaics



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ABSTRACT

The proof of concept of using luminescent down shifting (LDS) layers as alternative UV filters for P3HT:PCBM OPVs is demonstrated using a lanthanide-based metal complex. The results are verified using a combination of indoor light soaking, with single cell devices, and outdoor performance monitoring, using a 16-cell monolithically connected OPV module. By applying the LDS layer, a ~5% relative enhancement in photocurrent is observed for both sets of devices. More significantly, indoor light soaking tests on single cell devices without encapsulation showed an 850% enhancement in the measured half-life ($T_{50\%}$). The OPV modules were encapsulated and tested for outdoor stability over a 70 day period in the Negev desert, Israel. The modules made with the LDS filter are shown to match the stability of those made with a commercial UV filter and outperform the modules with no filter applied, with a 51% enhancement in the measured stability ($T_{75\%}$). Significantly, the work provides clear experimental evidence that the LDS layer can act as a UV filter in OPVs without compromising the efficiency of the solar cell, thus providing an added benefit over commercial UV filters.

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1. Introduction

Organic photovoltaics (OPVs) based on solution processable polymers and fullerenes have attracted remarkable interest during the last decade because of their potential for low cost, printability and flexibility. Recent research in this area has led to the report of a power conversion efficiency (PCE) of over 10% [1]. However, there are still challenges that the technologists developing OPVs need to overcome before it can become a mainstream solar technology, in particular combining high efficiency and long term operational stability in outdoor environments [2]. The causes of instability are numerous and complex and are induced or accelerated by a range of environmental effects such as temperature, water and ultraviolet (UV) light exposure [3], with the latter leading to photo-oxidation of active layer components or polymer chain scission [4,5]. As a result, the technology is unsuitable for outdoor applications unless a UV filter is applied. One major

disadvantage of incorporating a UV filter into the module is that the application is normally accompanied by a reduction in efficiency, due to increased optical losses [6].

In recent years, luminescent down-shifting (LDS) materials have been widely studied and applied to photovoltaics to improve the PCE [7,8]. These materials absorb photons at wavelengths (λ) where the PV responds poorly ($\lambda < 400$ nm) and re-emit photons in the visible spectrum, where the spectral response of the PV is much greater. For this reason, the application of an LDS onto an OPV could actually serve two purposes: (1) to filter the incident UV light and to suppress OPV degradation and (2) to improve the response of the OPV at short-wavelength.

Whilst the potential for efficiency and stability enhancements via LDS layers have been reported in a wide range of PV technologies, including Dye Sensitised Solar Cells (DSSCs) [9], there are only a limited number of studies that have been carried out on OPV materials [10] and devices [11–13]. In particular, Sloff et al. and Engmann et al. have reported promising results that show a photocurrent improvement by applying LDS layers in OPVs, but the stability issue was not addressed. This paper reports the proof

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of concept of improvement of both the lifetime and efficiency of OPV cells by applying LDS layers. The LDS layer can replace conventional UV filters, which are known to reduce the solar cell performance after application. The results are verified using a combination of indoor light soaking and outdoor performance monitoring, which was conducted over a 70 day period in the Negev desert in Israel.

2. Experimental

2.1. Single cell fabrication and indoor stability measurements

OPV cells were initially prepared in a clean room environment using indium tin oxide (ITO) coated glass substrates ($R_s = 16 \Omega^{-2}$) that were cleaned using solvents, then treated in a UV-ozone reactor with oxygen plasma for 10 min. A layer of zinc oxide (ZnO) film was deposited on ITO/glass substrate in a sputtering system at ambient temperature. Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) was spin coated on at 5000 rpm for 30 s and baked on a hotplate at 120 °C, before moving the samples into a nitrogen glovebox. Active layer blends using P3HT and [6,6]-phenyl-C₆₁-butyric acid methyl ester (C₆₁-PCBM), supplied by Nano-C Inc. USA, with weight ratios 1:0.8 were prepared and mixed with chlorobenzene solvent with a concentration of 30 mg/mL. Prior to coating, the blend was allowed to dissolve for 24 h on a hot plate stirrer and filtered using a 0.45 μm PTFE filter. Samples were transferred into a nitrogen atmosphere glovebox ($[O_2], [H_2O] < 1$ ppm), where the active layer was applied by spin-casting from a 60 °C solution (1500 rpm for 60 s). The active layer was annealed at 140 °C for 1 h. Finally, thermal evaporation of the anode was performed through a shadow mask to define device area and consisted of 10 nm of molybdenum trioxide (MoO₃) and 100 nm of silver (Ag).

The LDS layer was dissolved in PMMA (20 mg/mL). The films were formed by doctor blading, with a final thickness measured at ~1 μm. The doctor bladed layers were dried at room temperature for one hour. The LDS used for these tests was Tris(hexafluoroacetylacetonate) mono(1,10-phenanthroline)europium(III) [sym.: Eu(hfac)(phen)], which was purchased from Lumtec, Taiwan and used as received. Overall, a schematic of the cell is shown in Fig. 1(a) and an image of the LDS under UV excitation is shown in Fig. 1(b). Absorption and transmission were measured with UV-vis-NIR (UV-3600 SHIMADZU). Photoluminescence was measured with a Fluoromax-4 spectrophotometer.

OPVs were checked for initial performance prior to lifetime testing using a Newport solar simulator with 100 mW cm⁻² AM1.5G output (calibrated using a silicon reference cell from RERA in the Netherlands) and a Source Measurement Unit (SMU) for taking current density-voltage (*J-V*) measurements. For stability measurements, the cells were tested in accordance with ISOS-L-2 standards [14]. The cells were placed under the solar simulator for light soaking and were kept at open circuit in between measurements, with *J-V* measurements were made every 30 min for 550 h. Whilst the temperature of the cells was not controlled, it was measured and stayed relatively constant during the testing at 40 ± 5 °C.

2.2. Large area LDS printing for outdoor testing

In order to apply the LDS coatings over larger areas for outdoor performance monitoring, a DEK 248 screen printer was used for printing the Eu(hfac)(phen):PMMA (abbreviated to 'Eu:PMMA') inks. For screen printable inks, the LDS layer was dissolved in PMMA (10 mg/mL) with 4 wt% polyethylene glycol 400 (PEG 400) to act as a binder. The LDS coatings were printed onto a

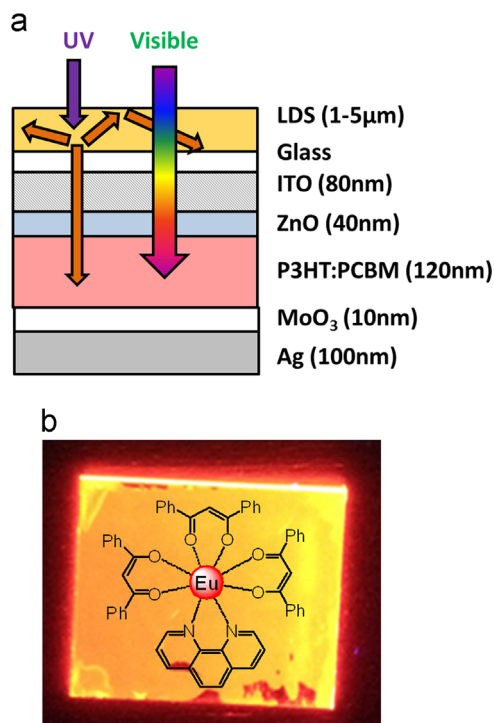


Fig. 1. (a) Schematic of the device used for single cell and 16-cell OPVs, with the LDS applied to the top surface and (b) the Europium complex and photoluminescent response from above using UV excitation.

polyethylene terephthalate (PET) carrier substrate of thickness = 125 μm, supplied by DuPont Teijin Films, UK. Trials were performed to optimise the ink transfer onto the substrate by studying the level of the snap-off gap and squeegee load that produced acceptable prints for the Eu:PMMA inks. After optimisation, it was discovered that optimal flood and print speed was 70 mm/min and Squeegee load was 10 kg. The PMMA inks were dried at 90 °C for 3–5 min on a belt dryer. The LDS layers were printed into rectangular patterns with dimensions of 8.5 × 10 cm². As the film thickness of the screen-printed LDS layer was 5 μm, wave-guiding losses to the edge of the substrate were minimised and are estimated to contribute also to < 0.5% of optical losses. As the printed LDS was slightly thicker than the layers described in Section 2.1, the concentration of LDS material was reduced to compensate. Measurements of the surface roughness of the printed LDS layer show the formed layer is very smooth ($R_A = 3$ nm). Fig. 2(a) shows a photograph of the PET substrate before and after Fig. 2(b) coating of the LDS layer, with no obvious change in visible appearance. The photoluminescence from UV excitation of the uncoated and coated LDS samples are shown in Fig. 7(c) and (d), respectively, confirming the uniformity of the LDS material dispersed in PMMA after screen printing.

2.3. Module (16-cell) fabrication and outdoor stability measurements

Roll-to-roll (R2R) coated OPV modules were produced without UV-filter and used for the outdoor performance tests. The fabrication of these modules followed the literature reports with the exception that the barrier material employed did not include a UV-filter, as part of the 'free OPV programme' at www.plasticphoto-voltaics.org [15,16]. The devices had an ITO free structure of Carbon/PEDOT:PSS/ZnO/P3HT:PCBM/PEDOT:PSS/Carbon/PET-substrate (Fig. S1 in Supporting information). Outdoor performance monitoring of these type of R2R coated OPVs which possess only a PET barrier layer is known to lead to rapid degradation in less than

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