



# Performance and stability improvements for dye-sensitized solar cells in the presence of luminescent coatings



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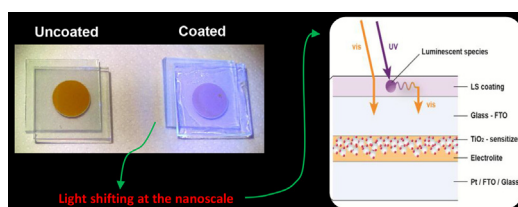
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## HIGHLIGHTS

- A multifunctional photocurable fluoropolymeric light-shifting coating is proposed.
- A luminescent agent in the coating shifts near-UV photons to VIS wavelengths.
- A 60% improvement of the DSSC efficiency was measured.
- An aging test was carried out outdoor for more than 2000 h.
- Light-shifting coating allows both efficiency and stability improvements.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Here we present how the sunlight radiation incident on a dye-sensitized solar cell (DSSC) can be shifted of a few tens of nanometers by means of an economical, easy to prepare and multifunctional photocurable fluoropolymeric light-shifting (LS) coating, to achieve both improved efficiency and device stability. By the introduction of a very small amount of a luminescent agent in the LS coating, the downshifting of near-UV photons to higher wavelengths easily harvestable by the organic dye of a DSSC is successfully demonstrated. This optical effect not only results in an over 60% improvement of the power conversion efficiency of DSSC devices, but the UV light filtering action promoted by the luminescent agent also provides protection to the photosensitive DSSC components. This aspect, combined with a potential thermal shielding effect and the easy-cleaning behavior imparted to the coating by its fluorinated nature, leads to excellent device stability as evidenced from an aging test performed outdoors under real operating conditions for more than 2000 h. Our study demonstrates that the use of light-cured multifunctional coatings with light management characteristics at the nanometer scale represents a new promising strategy to simultaneously increase the performance and durability of DSSC devices.

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## 1. Introduction

Dye-sensitized solar cells (DSSCs) have proven to be one of the most valuable third generation photovoltaic (PV) technology for the

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conversion of solar energy into electricity during the last twenty years [1]. The seminal work of O'Regan and Grätzel is the most cited article in the whole energy field [2], and more than 13,000 papers have been written to propose new materials, characterization techniques and large-scale implementation of DSSCs [3]. At present, efficiencies higher than 13% have been obtained on a laboratory scale [4], but also the manufacture of flexible devices [5], the design of unconventional electrodes [6], and the fabrication of hybrid cells [7] are research topics of great interest.

However, a few important aspects still need to be addressed in order to make DSSCs even more attractive and reliable on the global energy market. First of all, investigations have been carried out mostly on organometallic dyes, but metal-free organic dyes are getting increasing attention in view of obtaining market-competitive devices [8]. Organic dyes do not contain any rare or noble metal, thus generating fewer concerns about resource limits, and have recently demonstrated to be able to produce a 12.5% efficiency [9]. The second topic concerns the outdoor use of DSSCs, and therefore their resistance to meteorological phenomena, pollution and undesired UV radiation. Quite surprisingly, coatings have been developed almost exclusively for antireflective purposes, especially for silicon- and CdTe-based solar cells [10,11]. During the last two years, multifunctional coatings have also been designed for combined antireflective and self-cleaning purposes, and have been applied onto silicon and polymeric solar cells [12,13]. Even if polymers have been chosen as basic matter for coating preparation, also inorganic matrices (pure or mixed metal oxides) have been investigated [14]. However, despite the great academic and industrial efforts spent on DSSCs components, the development of functional coatings for improving the outdoor stability and performance of this type of third generation devices has been surprisingly neglected. Most of the research groups overlook this aspect, and only a small fraction of them apply a commercial antireflective coating on the fabricated PV cells [15]. Very recently, a couple of interesting works has been proposed on this matter. Park et al. prepared a superhydrophilic nanoparticle-based coating to be applied as anti-fogging layer [16], while Heo et al. fabricated a hydrophobic nanopatterned coating with antireflective and self-cleaning properties [17]. We think that the impact of these seminal works may trigger the interest of the scientific community on a very far ignored aspect of DSSC technology. In particular, the great potential of fluorinated polymer matrices has yet to be exploited in the DSSC field. Indeed, fluorinated polymers are a class of high-performance materials that rely on the superior strength (higher dissociation energy) of the carbon–fluorine bond compared to the carbon–hydrogen bond to achieve excellent durability, weatherability, chemical and photochemical resistance [18–20]. The use of fluorinated polymer coatings for outdoor applications represents a consolidated way to achieve high weathering resistance and long-term durability in a variety of technological fields (e.g., architectural, nautical), and only very recently their application to the field of energy storage and conversion has also been demonstrated [21–23].

Our recent experimental research activities have been separately focused on the preparation of light-managing coatings for PV applications [23,24] and on the development of new materials for different PV technologies [25–27]. A very ambitious goal would be that of combining the ability of a coating to be easily-cleanable with a light-shifting (LS) effect promoted by the addition of an appropriate luminophore to the coating system. Considering this latter point, a few recent articles reported on the use of rare earths as UV or visible light shifters [28–32]. However, such systems have two major flaws: 1) The luminophores ( $Y_2WO_6:Ln^{3+}$ ,  $NaYF_4:Yb^{3+}/Er^{3+}$ ,  $CaZnOS:Eu^{2+}$ ,  $Y_2O_3:Er^{3+}$ ) are composed of rare and expensive inorganic elements; 2) Aging studies on devices including these LS

systems are not presented, presumably because these inorganic materials are water soluble or suspendable, thus real outdoor applications are not conceivable. A first example of a stable luminescent system for DSSCs has been very recently proposed by our group, and consisted of a polymer coating containing a Europium complex as downshifter [33]. However, the presence of rare and expensive element such as Europium does not make this technology suitable for a large-scale trade.

In this work, we propose a multifunctional coating which can simultaneously be easy-cleaning and LS for application in organic DSSCs. In detail, the easy-cleaning ability is ascribed to the use of a fluoropolymeric matrix system that can be photocured in a few seconds, thus potentially enabling its use in large-scale production volumes. By the addition of a rare elements-free organic fluorophore, this coating is also able to serve as LS system by absorbing light in the UV portion of the solar spectrum and re-emitting it in the spectral region where the DSSC sensitizer shows a maximum absorption (Fig. 1). Since organic DSSC dyes often present a relatively narrow spectral breadth [34,35], our proof of concept shows the possibility to partly overcome this problem, by shifting otherwise-wasted and potentially harmful UV-photons to wavelengths suitable for absorption by the DSSC device. The optimization of the LS-DSSC system allowed us to improve by 62% the efficiency of a DSSC prepared with the organic dye D131. Moreover, thanks to the UV-screening action of the rare elements-free fluorophore used in the LS-coating presented in this work, the often detected photo-oxidative degradation of the organic dye [36] could be prevented. Accordingly, an aging test conducted for more than 2000 h in real outdoor conditions revealed the excellent stability of the new LS-DSSC system presented in this work compared to control devices.

## 2. Experimental

### 2.1. Materials

The chloro-tri-fluoro-ethylene vinyl-ether (CTFE-VE) polymeric binder (Lumiflon LF-910LM) was obtained from Asahi Glass Company Ltd., while 2-isocyanatoethyl methacrylate (IEM) was obtained from Showa Denko K.K. Both were used as received. The luminescent species employed in this work was Lumogen F Violet 570, purchased from BTC Europe.

As regards DSSC components, conducting glass plates (FTO glass, Fluorine doped Tin Oxide over-layer, sheet resistance  $7 \Omega \text{ sq}^{-1}$ , purchased from Solaronix) were cut into  $2 \text{ cm} \times 2 \text{ cm}$  sheets and used as substrates for both the deposition of a  $\text{TiO}_2$  porous film from a paste (DSL 18NR-AO, Dyesol) and the fabrication of platinized counter-electrodes. Sensitizing dye 2-[[4-(2,2-diphenylethenyl)phenyl]-1,2,3,3a,4,8b-hexahydrocyclopenta[b]indole-7-yl]methylidene]-cyanoacetic acid (D131) was purchased from Inabata Europe S.A.

All other reagents were purchased from Sigma Aldrich, unless otherwise stated.

### 2.2. Synthetic method

The CTFE-VE polymer was allowed to react with IEM to form the photocrosslinkable polyurethane precursor. In a typical synthesis, 40 g of CTFE-VE and 7.3 g of IEM were poured in stoichiometric ratio ( $\text{OH}/\text{NCO} = 1$ ) into a three-necked round-bottomed flask equipped with a bubble condenser. Di-*n*-butyltin dilaurate (0.3 wt.%) was added successively to act as catalyst. The reaction was conducted in a nitrogen atmosphere and under vigorous magnetic stirring at  $75 \text{ }^\circ\text{C}$ . The extent of reaction was controlled by monitoring the disappearance of the  $\text{N}=\text{C}=\text{O}$  stretching signal ( $2270 \text{ cm}^{-1}$ ) by

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