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Enhancement of outdoor stability on dye-sensitized solar cells by spectrum conversion layer with 1,8-naphthalimide derivatives

Dae Man Han,[ad] Hye-Jin Song,[b] Chi-Hwan Han *[b] and Youn Sang Kim *[ac] ,

As DSSCs are vulnerable to continuous irradiation of Ultra Violet (UV) light, for outdoor stability, UV cut-off filter is vital to shield the UV light at outdoor conditions. Unfortunately, the large drop of photo-conversion efficiency by the UV cut-off is inevitable to maintain the outdoor stability. Herein, we propose a novel UV conversion layer using unique combination of spectrum conversion materials with UV absorbing 1,8-naphthalimide derivatives in poly (ethylene-co-vinyl acetate) on the photo-anode. This functional layer has shown unique characteristics which form exciton complex molecules at high concentration under UV absorption. As a result of converting absorbed UV light to visible light source for the sensitizing dye, the relative efficiency of the proposed DSSCs have shown comparable initial photo-conversion efficiency to bare DSSC and maintained 18% higher relative photo-conversion efficiency after 48 days outdoor condition than DSSC using commercial UV cut-off filter.

Introduction

Dye-sensitized solar cells (DSSCs) are mainly consisted of photo-anode, sensitizing dye, electrolyte and counter electrode. In general, the photo-anode composed with titanium dioxide nanoparticulate film on the transparent conductive glass. Titanium dioxide (TiO_2) nanoparticles provide the support for sensitizing dye binding site with a wide specific surface area and work as a wide band gap n-type semiconductor layer for electron collection. However, in case the TiO_2 nanoparticles coated on the photo-anode absorb a UV light, they act as a photocatalyst inducing the degradation of sensitizing dye.[1-3] Also, the tri-iodides (I_3^-) in liquid electrolyte, which act as a redox couple with iodide (I^-), are reduced irreversibly on the TiO_2 surface to colorless iodide ions by the photocatalyst effect, followed by inducing a lack of reversible oxidation-reduction reaction of the electrolyte.[4, 5] In this reason, DSSCs needs a UV cut-off filter for the protection of degradation by UV light. DSSCs without UV cut-off filter show higher photo-conversion efficiencies but lower stability at outdoor conditions. On the contrary, DSSCs with UV cut-off filter have higher stability at outdoor conditions but lower photo-conversion efficiency due to large amount of incident light loss.[6, 7]

UV cut-off materials are selected among UV absorbing materials like as silica, alumina, zirconia, titania and ceria to cut off wavelength under 387 nm selectively, which is the maxima absorption peak of anatase type TiO_2. However, there has been limit to mitigate the relative drop of photo-conversion efficiency using conventional UV cut-off filter only, because the filtered amount of UV light cannot be re-used for DSSCs operation. There have been advanced researches to enhance the photo-conversion efficiency of DSSCs using (i) quantum dots (ii) organic dyes and (iii) rare-earth ions/complexes as spectrum conversion materials by (i) coating on the front or rear glass (ii) doping into the TiO_2 nanoparticles and (iii) dispersing in the liquid electrolyte.[8-11] However, there was little improvement of photo-conversion efficiency through coating spectrum conversion materials on the front glass against previous theoretical expectations.[12] Meanwhile, applying the spectrum conversion materials by dispersing in the liquid electrolyte or by coating on the rear glass were more effective to improve the photo-conversion efficiency.[13, 14] This result arises from the large loss of incident light and deficient utilization of absorbed light, because the emitted wavelength from spectrum conversion layer (SCL) is too short to be used by the sensitizing dye.[15] The spectrum range needed to shield for outdoor stability is under 387 nm and the spectrum range needed to enhance the photo-conversion efficiency is a visible light between 400 and 530 nm. Thus, the challenge of replacing UV cut-off filter with SCL for outdoor stability is to increasing the stoke shift large enough for effective utilization by sensitizing dye.[16]

To address this issue, we propose a novel system using unique combination of SCL materials with UV absorbing 1,8-naphtalimide derivatives on the photo-anode. To increase the stoke shift by the formation of exciton complex molecule upon UV absorption, we used a heteromolecular compound of 1,8-naphthalimide derivatives. A compound of 1,8-naphthalimide derivatives as a guest material and poly (ethylene-co-vinyl acetate) as a host material was dissolved in toluene solvent to form a functional layer on the photo-anode of the DSSC. (**Scheme 1**) Prepared 1,8-naphthalimide derivatives are generally synthesized from the corresponding anhydrides by reaction with alkyl amine. Each alkyl amine with different alkyl chain produces different family of derivatives showing unique fluorescent properties. General synthetic route for 1,8-naphthalimide derivatives, numbering and representative formula are shown in **Figure S1**.[17]

To demonstrate the effect of SCL with heteromolecular fluorescent compound, we conducted several analyses as follows. First, we have shown that the stoke shift could be increased by the formation of exciton complex at higher concentration of the 1,8-naphthalimide derivatives, by measuring the transmittance, absorbance, and fluorescence of the SCL on the FTO glass. Second, the evidence of converting absorbed UV light for power generation in DSSC by SCL was shown by measuring the IPCE especially under 370 nm and photo-conversion efficiency of the prepared DSSCs. Finally, we conducted outdoor stability test using prepared DSSCs for 48 days at outdoor field. As a result, DSSC with SCL has shown comparable initial photo-conversion efficiency to bare DSSC and maintained 18% higher relative photo-conversion efficiency after 48 days outdoor condition than DSSC using commercial UV cut-off filter.

Experimental

(1) Preparation of Spectrum Conversion Material

A compound of 1,8-naphthalimide derivatives as a spectrum conversion material was prepared from a company (Lumicolor-Violet, ICB Korea). Poly (ethylene-co-vinyl acetate) (EVA, vinyl acetate 40 wt.%, Aldrich) bead 5 wt.% was dissolved in toluene as a host material stock solution. Different concentrations of spectrum conversion materials were prepared by dissolving 1,8-naphthalimide derivatives in host material stock solution with the ratios of 0.5, 1.0, 1.5, 2.0, 2.5 and 3.0 wt.% respectively. TiO_2 sol (solution of TiO_2 nanoparticle diameter 15nm, ENB Korea) was prepared as a reference UV absorbing material. Commercial UV cut-off film (PR70, 3M) was prepared as a reference UV cut-off filter.

(2) Fabrication of the DSSC

Photo-anode films were prepared by screen printing TiO_2 pastes on the FTO glass (TEC 8/2.3mm, 8 Ω/sq, Pilkington) with aperture area of 0.2 cm^2 and sintered at 550 ^oC for 30 min.[18] Dye adsorption was carried out by dipping the TiO_2 photo-anode films into a 4×10^{-4} M t-butanol/acetonitrile (Merck, 1:1) solution of the ruthenium dye (N719, Solaronix) for 48 hour at 25 ^oC. Nano-gel type electrolyte which was prepared according to the previously reported method adding silica nanoparticle was coated on the TiO_2 photo-anode film.[19] Counter electrodes were prepared by dropping of 10 mM hydrogen hexachloroplatinate (IV) hydrate (99.9%, Aldrich) in a 2-propanol solution onto the transparent FTO glass and after treatment at 400 ^oC for 30 min. The two electrodes were assembled and separated using thermal adhesive polymer film (Surlyn, thickness 60 μm).

(3) Formation of Spectrum Conversion Layer

FTO glasses (25 mm x 40 mm) were prepared by washing and sonication. Each side of the FTO glasses or DSSCs was masked using polyimide tape before coating 1,8-naphthalimide derivatives on the FTO glass using dip coater, to open the active area of front side only. Prepared FTO glasses or fabricated DCCSs were dipped in 1,8-naphthalimide derivatives solution to form exciton complex forming SCL on the substrate using table-top dip coater (e-flex, EF-4200). The raising speed of the dip coater was controlled between 1 and 10 mm/sec after dipping 5 seconds in solution to keep the optimal thickness for anti-reflection.

(4) Characterization of the SCL and DSSC

In order to investigate the optical properties of SCL, transmittance and diffuse reflectance of the FTO glasses with SCL were measured using UV-vis spectrophotometer (CM-5 Konica Minolta, Japan) and analyzed with color data software (SpectraMagic NX Ver.2.1 Konica Minolta, Japan). The absorbance of the SCL was calculated from the obtained transmittance and reflectance data. The fluorescence of the SCL was evaluated by measuring the emission spectrum of the FTO glasses with SCL using Fluorescence Spectrometer (FS-2, scinco) and analyzing with spectrum data software (FluoroMaster Plus Software, Scinco). Emitted photon numbers between the wavelength of 300 and 700 nanometers by the excitation at 370 nanometer UV light were counted by the probe under fixed condition of scan speed 300 nm/min and PMT voltage 500 volt followed by spectrum correction.

(5) Characterization of the photovoltaic properties

The photovoltaic properties of the prepared DSSCs were measured using a 1600W Ozone free lamp with a 'Class A' Air Mass 1.5G filter as a light source in a solar simulator (Newport-94083A) and with an exposure control instrument (Newport-68951). The light intensity was adjusted with a reference Si cell (Fraunhofer Institute for Solar Energy System). Photovoltaic performance was characterized by V_{oc}, J_{sc}, and FF (fill factor), and the overall photo-to-current conversion efficiency was characterized using the current density-voltage (J-V) curve.

Incident photon-to-current efficiencies (IPCE) were measured by the illumination of prepared DSSC with a 200W Xenon Arc light source through a filter monochromator using optical chopper rotating at 2 Hz under 1 sun bias light (Newport-CS260).

For the test of outdoor stability under continuous UV light irradiation, the prepared DSSCs were arrayed on an outdoor field which was set up custom-built for this study. Then, the photo-to-current conversion efficiencies were characterized weekly.

Results and discussion

Transmittance of the FTO glass coated with TiO_2 sol was measured as a reference UV absorbing material. **Figure S2** shows that the transmittance of the FTO glass coated with TiO_2 sol was cut off below 370 nm compared to that of bare FTO glass. UV cut-off peak was slightly red shifted according to the number of dip coating, though the change was very little. This UV cut-off is needed because the degradation of DSSC by UV light occurs as a result of photocatalytic reaction from the direct excitation of the TiO_2 band gap. Anatase phase TiO_2 is photoactive up to 387 nm with band gap of 3.2 eV, while rutile phase TiO_2 is photoactive up to 411 nm with band gap of 3.02 eV. If the photo-anode is made with TiO_2 nanoparticle starting from a mixture of anatase and rutile phase (for example, Degussa P25), the UV cut-off filter must shield up to 411 nm. However, if the whole incident UV light under 411 nm were cut-off without re-using, relative drop of photo-conversion efficiency would be large compared to the bare DSSC without UV cut-off filter.[20]

Transmittance of the FTO glass coated with 1,8-naphthalimide derivatives for SCL was measured according to the concentrations to show the possibility as a substitute for UV cut-off filter. **Figure 1** shows that the transmittance of the SCL below 370 nm falls down according to the concentration of the 1,8-naphthalimide derivatives. Additionally, the absorption edge was red shifted gradually up to 400 nanometers. However, the visible light transmittance decreased slightly according to the concentration of the 1,8-naphthalimide derivatives. The concentration of 1,8-naphthalimide derivatives over 2 wt.% was favorable for higher UV light stability, but, further gain of UV light stability according to the concentration was very small while the loss of light transmission increased. To maintain a balance between UV light stability and photo-conversion efficiency, higher outdoor performance was expected at the concentration of 2 wt.% of 1,8-naphthalimide derivatives, showing high absorbance at UV absorption peak and wide range of absorption edge to cover the absorption range of TiO_2 film on the photo-anode, previously.

The absorbance of the coating layers were calculated from the measured transmittance and reflectance data. (The reflectance curve is shown in **Figure S3**.) **Figure S4** shows that the absorption peak is at around 370 nm and absorption edge is at around 414 nm for SCL. The absorption peak of TiO_2 sol coated FTO glass as a reference material for UV absorbing property of the photo-anode shows the similar shaped graph with SCL coated FTO glass. This result confirms that absorption peak of SCL is adequate to cover the absorption peak of photo-anode and the absorption band is adequate to block the required range of UV light to replace the UV cut-off filter, which shields the UV light on the TiO_2 nanoparticulate photo-anode film layer.

Fluorescence of the SCL by the excitation at 370 nm was measured with prepared SCL FTO glasses using 1,8-naphthalimide derivatives with concentration of 0, 0.5, 1.0, 1.5, 2.0, 2.5 and 3.0 wt.%, respectively. **Figure 2** shows that the emission peak of SCL remained at 424 nm under low concentration, however, the peak was gradually red shifted up to 476 nm by increasing the concentration of 1,8-naphthalimide derivatives to 2 wt.%, thereafter, it was saturated at further increase of concentrations. This phenomenon can be explained by the formation of exciton complex molecules at high concentration in the excited state. Because, exciton complex molecule formation is dependent on a bimolecular interaction, it is promoted by high monomer density. At low density, excited monomers decay to the ground state before they interact with an unexcited monomer to form an exciton complex molecule. However, at high density, formation of dimeric molecule is possible very shortly on the order of nanoseconds in an electronic excited state. Once one of the heterodimer components is excited by the absorption of UV light, the charge is transferred to the LUMO (Lowest Unoccupied Molecular Orbital) of unexcited component monomer followed by emitting longer wavelength light with smaller energy.[21, 22]

Traditional N719 dye has shown high photo-conversion efficiency exceeding 11 % when employing I^-/I_3^- liquid electrolyte, with relatively intense absorptions between 400 and 535 nm ($\varepsilon \approx 14{,}000\ M^{-1}cm^{-1}$), exhibiting high open circuit potential and ultrafast electron injection after light absorption. The absorption maximum for N719 dye bound to TiO_2 is 531 nm (2.34 eV), having a lowest optically active excited state from the dye π^* located at 0.3 eV above the lowest transition localized in the TiO_2. By the formation of exciton complex molecule, the emission spectrum of SCL could reach to the lowest optically active excited state of N719 dye bound to TiO_2 to produce higher energy for the minimum driving force needed for electron injection.[23]

The thickness of SCL after dip coating is a very important figure for optic and economic valuation of proposed system. The thickness of SCL with 0 wt.% of 1,8-naphthalimide derivatives was identified to be between 253 ~ 263 nanometers using scanning electron microscope (SEM). The thicknesses of SCL containing 1,8-naphthalimide derivatives were kept to be same with SCL with 0 wt.% to keep optimal optic property. **Figure S5** shows the cross-section picture of SCL with 0 wt.% 1,8-naphthalimide derivatives on FTO glass taken by SEM after pretreatment with liquid nitrogen for rapid cooling. However, further cross-section measurement was difficult by the elasticity of the SCL.

To investigate the initial photo-conversion efficiency drop by the additional layer on the DSSC, the DSSCs were prepared using TiO_2 nanoparticulate film of anatase/rutile phase mixture, N719 sensitizing dye and nano-gel type electrolyte with I^-/I_3^-

redox couple. **Figure 3(a)** shows the photo-conversion efficiency result of the DSSCs having bare, SCL and UV cut-off filter coated FTO glasses, respectively. Relative photo-conversion efficiency of DSSC using commercial UV cut-off filter dropped 22.6 % compared to the bare DSSC. On the contrary, the relative photo-conversion efficiency DSSC using SCL was 0.07 % higher compared to the bare DSSC by the effect of anti-reflection effect and conversion of absorbed UV light, despite the large loss of visible light source due to longer wavelength absorption edge. **Figure S6** shows the transmittance and reflectance of commercial UV cut-off filter. Total transmittance was 69.5 % and UV cut-off edge was 400 nm. **Figure S7** shows the difference of photo-conversion efficiency of DSSCs with SCL 2 wt.% and bare DSSC. Number of sample was 37 and the maximum deference was below 0.3 %.

To demonstrate more concrete evidence to prove the enlarged usage of incident UV light, Incident Photon-to-Current efficiencies (IPCE) of the prepared DSSCs were compared. **Figure 3(b)** shows the IPCE result of the DSSCs having bare, SCL and UV cut-off filter coated FTO glass, respectively. There was a very interesting finding in the IPCE result of DSSC using SCL at UV range below 370 nm. The IPCE value of DSSC using SCL was 19 % at 370 nm, this efficiency is extraordinarily high. The IPCE value of DSSC using bare FTO glass was 36 % from the transmittance of 61 % at the same wavelength. The expected IPCE value from the transmittance of 21% is only 12 %, in case of DSSC using SCL. Therefore, there was an extra gain of 7 % IPCE value at 370 nm. This is a compelling evidence of re-using absorbed UV light to achieve prominent photo-conversion efficiency enhancement by the spectrum conversion.[14]

As an ultimate objective of this study, outdoor photo-conversion efficiencies of the prepared DSSCs were compared. **Figure 4** shows the relative outdoor photo-conversion efficiency result of the DSSC using bare, SCL and UV cut-off filter, respectively. Relative photo-conversion efficiency of the DSSC using a compound of 1,8-naphthalimide derivatives for SCL was 18% higher than the DSSC using commercial UV cut-off filter, after 48 days outdoor conditions, which confirms that the higher performance was achieved at outdoor condition by the additional exciton complex forming SCL coated on the bare DSSC.

Conclusions

In summary, the potential of unique combination of SCL materials with UV absorbing 1,8-naphthalimide derivatives as a substitute to the UV cut-off filter on the DSSC was studied to enhance the outdoor stability. The measurement of transmittance and absorbance of SCL coated FTO glass showed that the UV light below 414 nm could be largely absorbed. The measurement of Stokes fluorescence of SCL coated FTO glass showed that the emission peak could be red shifted from 424 nm to 476 nm at 2 wt.% concentration of a compound of 1,8-naphthalimide derivatives according to the formation of exciton complex molecules upon UV absorption. The Incident Photon-to-Current efficiency result provided concrete evidence that the absorbed UV light below 370 nm was converted into a longer wavelength and used to enhance the photo-conversion efficiency. The actual outdoor field test was conducted to evaluate the ultimate objective of the DSSC using proposed spectrum conversion layer with a compound of 1,8-naphthaimide derivatives. Outdoor stability test for 48 days demonstrated that the functional layer coated on the DSSC with concentration of 1,8-naphthalimide derivatives 2 wt.% helped to maintain the comparable initial photo-conversion and 18% higher relative photo-conversion efficiency after 48 days than DSSC using commercial UV cut-off filter. We believe that our new finding would provide inspiration to enhance outdoor stability of DSSCs especially for building and automobile integrated photovoltaic devices.

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Notes and references

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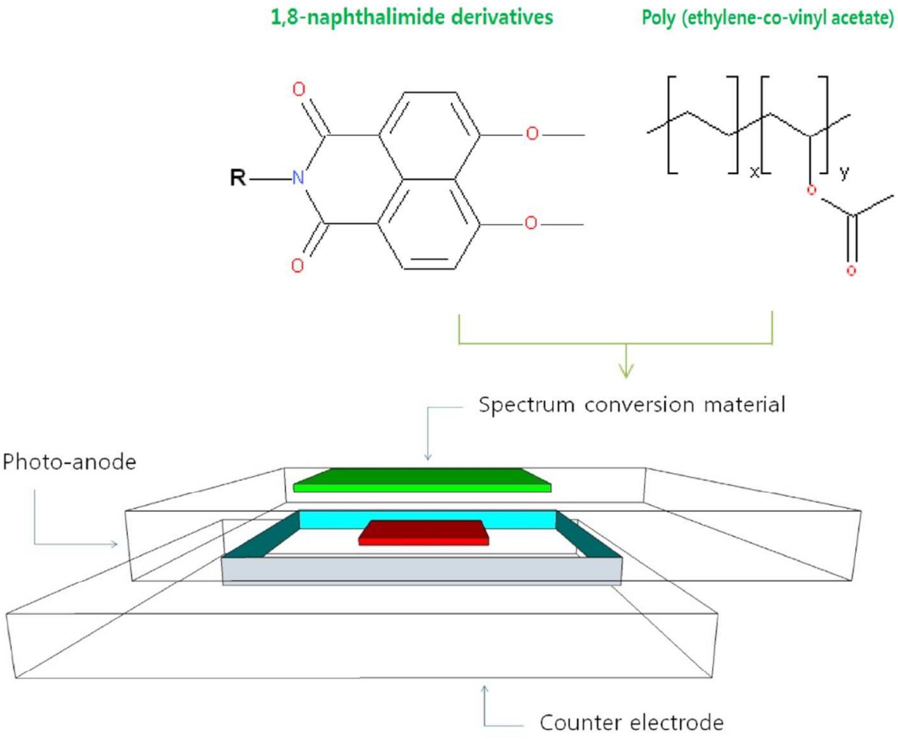


Scheme 1 Exciton complex forming spectrum conversion material with a compound of 1,8-naphthalimide derivatives in EVA (poly (ethylene-co-vinyl acetate)) is coated on the photo-anode of the DSSC.

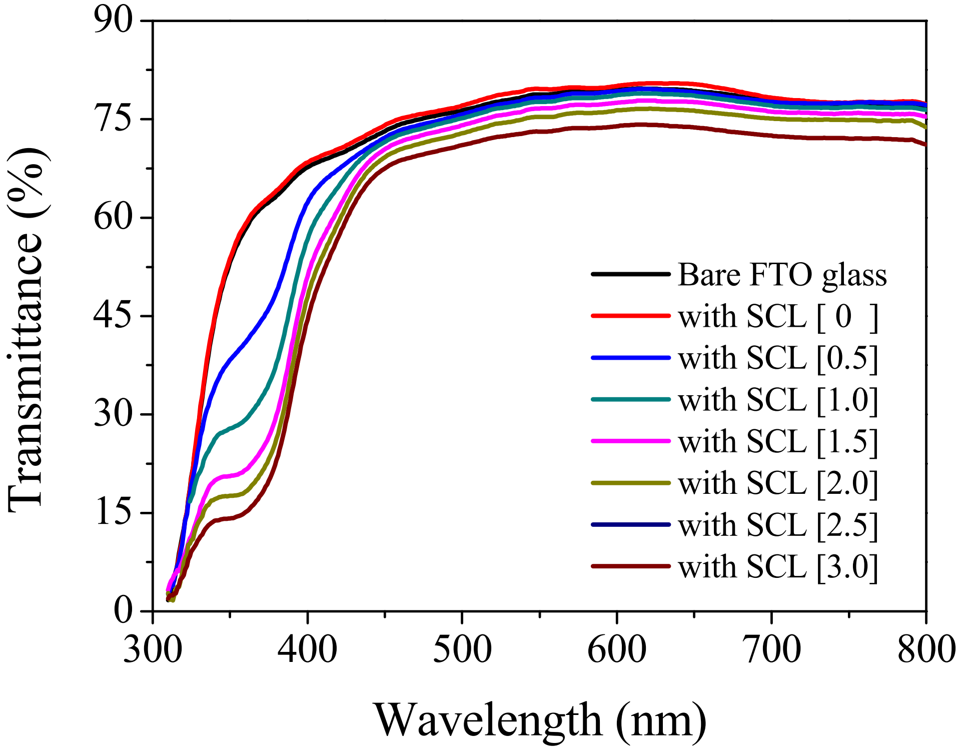


Fig. 1 Transmittances of FTO glasses with SCL of 0, 0.5, 1.0, 1.5, 2.0, 2.5 and 3.0 wt.% 1,8-naphthalimide derivatives, respectively.

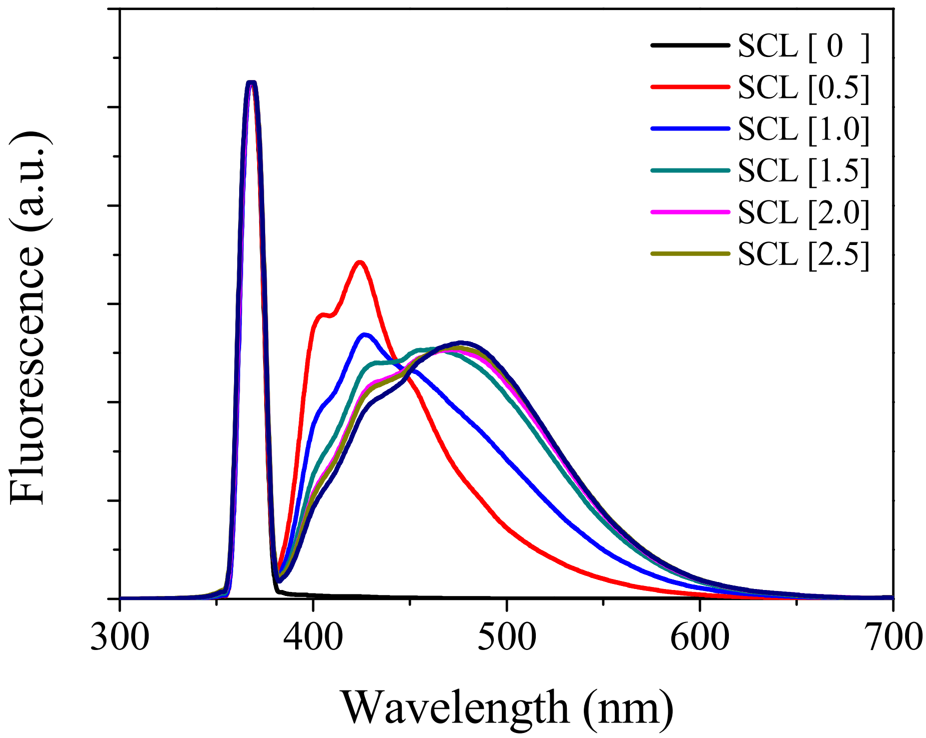
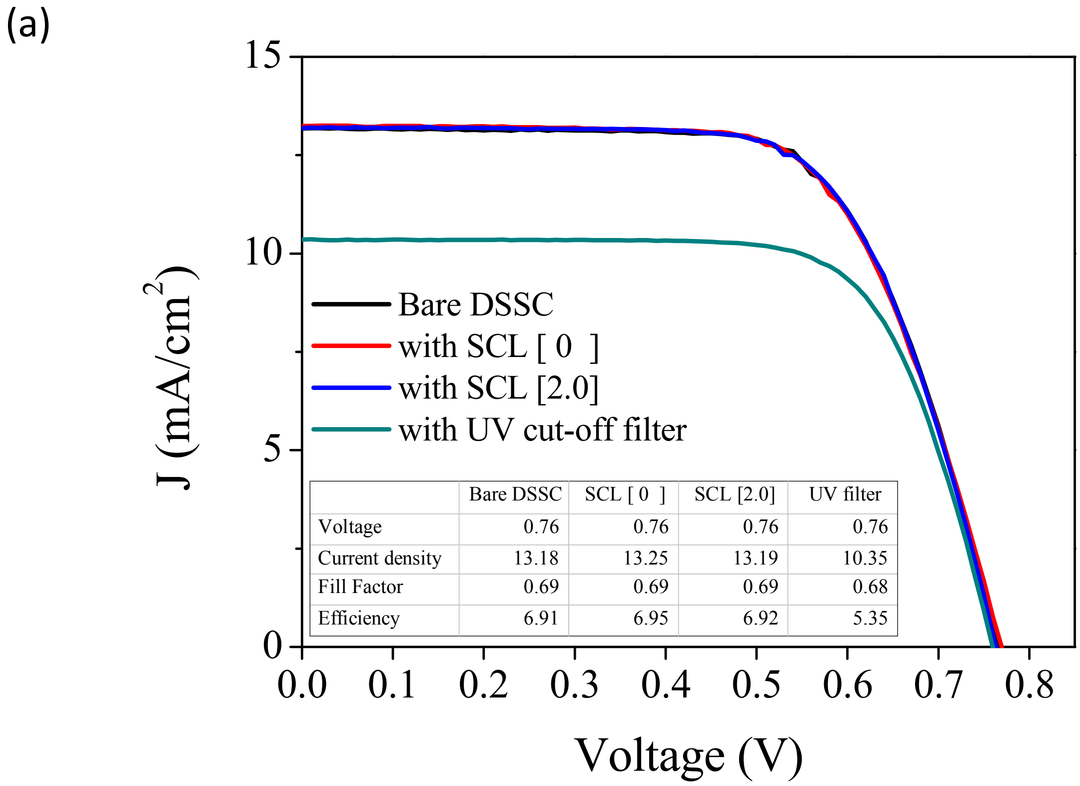
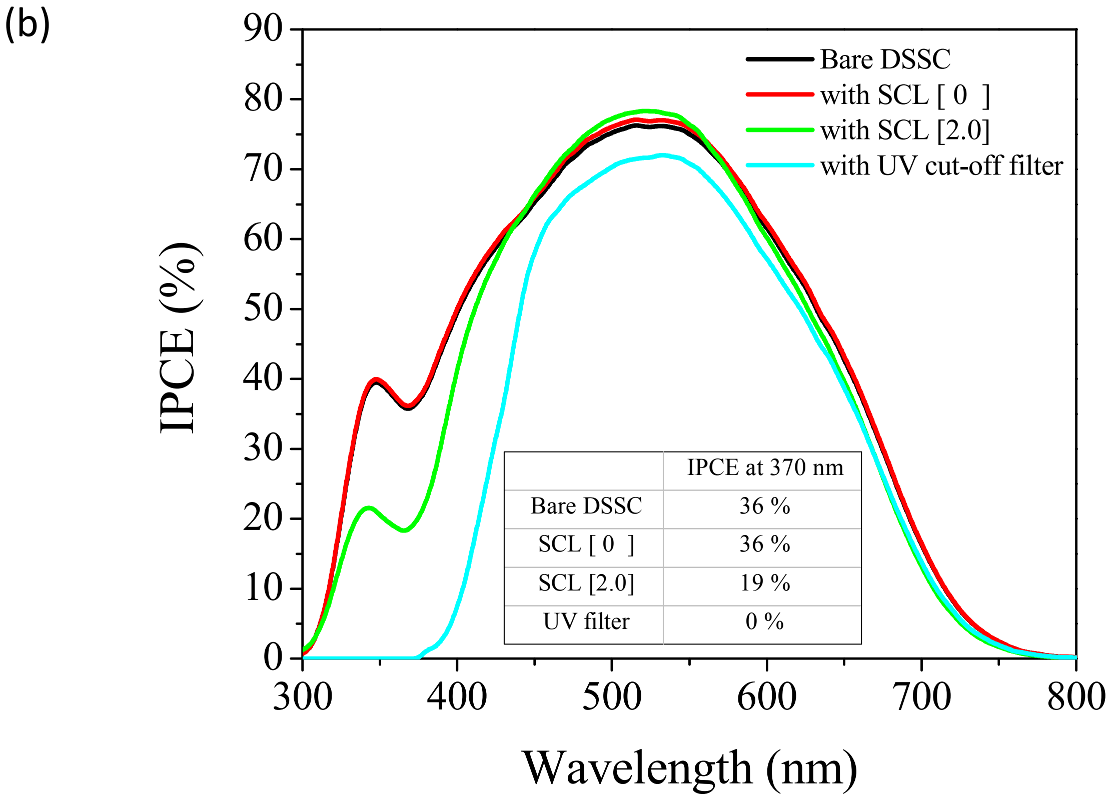


Fig. 2 Fluorescences of FTO glasses with SCL of 0, 0.5, 1.0, 1.5, 2.0, 2.5 and 3.0 wt.% 1,8-naphthalimide derivatives, respectively.



	Bare DSSC	SCL [0]	SCL [2.0]	UV filter
Voltage	0.76	0.76	0.76	0.76
Current density	13.18	13.25	13.19	10.35
Fill Factor	0.69	0.69	0.69	0.68
Efficiency	6.91	6.95	6.92	5.35

Fig. 3 (a) Initial Photo-to-Current conversion efficiencies and (b) Incident Photon-to-Current efficiencies (IPCE) of the prepared DSSCs using (i) bare FTO glass without any optical filter, (ii) 1,8-naphthaimide derivatives 0 and 2 wt.% as SCL and (iii) commercial UV cut-off filter, respectively.



	IPCE at 370 nm
Bare DSSC	36 %
SCL [0]	36 %
SCL [2.0]	19 %
UV filter	0 %

Fig. 3 (a) Initial Photo-to-Current conversion efficiencies and (b) Incident Photon-to-Current efficiencies (IPCE) of the prepared DSSCs using (i) bare FTO glass without any optical filter, (ii) 1,8-naphthaimide derivatives 0 and 2 wt.% as SCL and (iii) commercial UV cut-off filter, respectively.

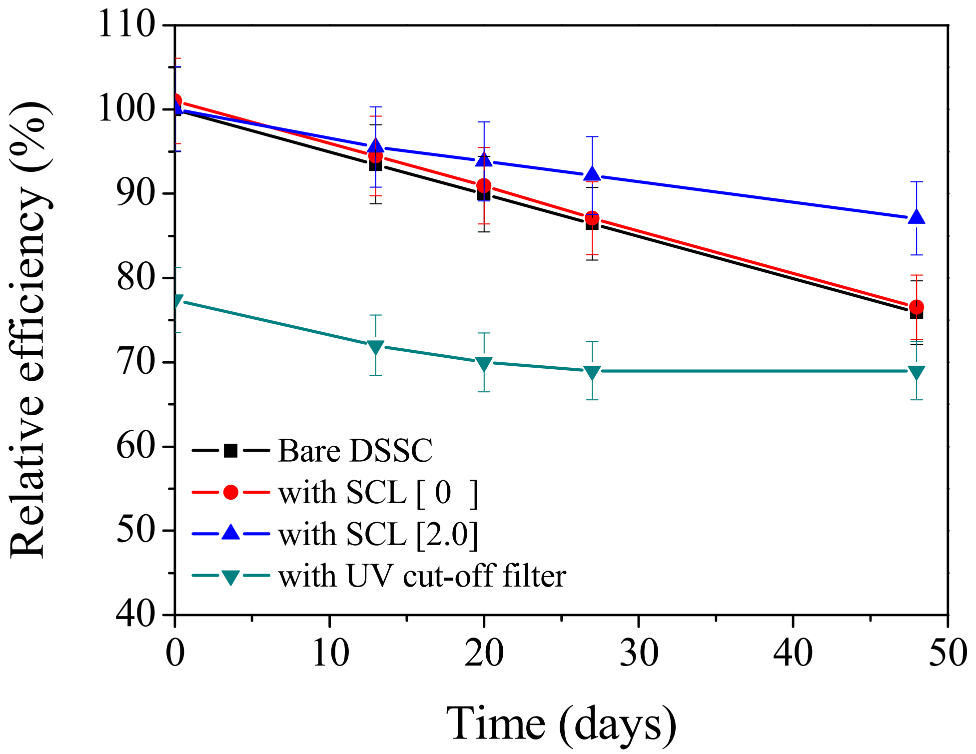


Fig. 4 Relative outdoor Photo-to-Current conversion efficiencies of the prepared DSSCs using (i) bare FTO glass without any optical filter, (ii) 1,8-naphthaimide derivatives 0 and 2 wt.% as SCL and (iii) commercial UV cut-off filter, respectively.

Enhancing outdoor stability of dye-sensitized solar cells by the spectrum conversion layer with 1,8-naphthalimide derivatives

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Abstract

We propose novel UV conversion layer with a compound of 1,8-naphthalimide derivatives, which absorb damaging UV light in advance and emit longer wavelength light by Stokes fluorescence. By converting absorbed UV light more effectively, the outdoor stability maintained higher level than bare DSSC without spectrum conversion layer.