Enhancing Optical Efficiency of Thin-Film Luminescent Solar Concentrators by Combining Energy Transfer and Stacked Design

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Abstract

We report on a new approach aimed at enhancing the optical efficiency of luminescent solar concentrators (LSCs) based on poly(methyl methacrylate) (PMMA) thin-films doped with highly emissive fluorophores. In detail, a series of high-energy absorbing fluorophores (HEF) and perylene bisimides (PBI) red-emitting dyes were utilized in order to favor the collection of a large number of incident photons by the thin-film LSCs. The use of two fluorescent PMMA layers coated on the opposite surfaces of a glass slab was found to effectively increase the light concentration. The worthwhile combination of the appropriate HEF and PBI fluorophores within the secondary (bottom) PMMA layer assured optically efficiencies 10–14% greater than the maximum value of single-dye LSCs.

1. Introduction

Luminescent solar concentrators (LSCs) have been widely studied as a viable alternative to decrease the price of photovoltaic energy.[1] LSCs show advantages like the ability to work with diffuse light,[2, 3] light weight, reduced costs, and transparency, which makes them available to be implemented in architectural solutions.[4] LSCs consist in a slab of transparent material doped with a fluorophore able to absorb the solar spectrum.[4-6] The higher refractive index of the host compared to the environment makes it possible to trap a fraction of the emitted photons by means of total internal reflection.[7] Photons are then collected at the edges of the device to produce electric power by means of photovoltaic (PV) cells. In recent years, the research on PV devices based on LSC technology has been focusing on obtaining high power conversion efficiencies.[3, 8-17] Still, for prototype single-dye LSCs coupled to commercial Si cells efficiencies higher than 3-4% are rarely met.[4] This is due to the many losses of such devices, both related to the physics of the phenomena involved and to a not-yet-optimized fluorescent system.[18] A simple approach to obtain higher concentrations is to enhance the spectral window of absorption of the LSC. For example, multiple dyes have been proposed to cope for the narrow absorption characteristic of organic dyes as well as new design solutions.[2, 13, 19, 20] Noble metals nanoparticles[21, 22] and quantum dots have also been investigated for their broad absorption features although compatibility problems with host matrices seldom arise.[9, 23-25] Among the proposed designs, the stacking of two or more differently dyed slabs and the mixing of multiple fluorophores in the same bulk are the more often encountered in the literature. In the stacked geometry,[15-18, 26] slabs doped with different dyes that use different portions of the solar spectrum are piled up on top of the other and are connected to different PV cells. Sloff et al.[17] described a stacked device with a power conversion efficiency of 7.1%, which is, at best of our knowledge, the highest efficiency ever reported for LSC-PV systems. Conversely,

a dye mixture in a single slab offers the possibility of cascading of emission via non-radiative processes such as the fluorescence resonance energy transfer (FRET).[2, 27]

In striving to contribute towards improved LSC outcomes, we report on the preparation of LSCs based on fluorophore-doped poly(methyl methacrylate) (PMMA) thin-films coated onto high optical quality glass slab. A series of high-energy absorbing fluorophores (HEF), such as 2,5-bis(5-tert-butyl-benzoxazol-2-yl)thiophene (BTBBT), perylene (Per), and two different perylene bisimide (PBI) red-emitting dyes such as N,N'-bis-(1'-phenylethyl)-perylene-3,4,9,10-tetracarboxydiimide (PE-Pery)[28] and Lumogen F 350 (LR) were utilized (Figure 1).
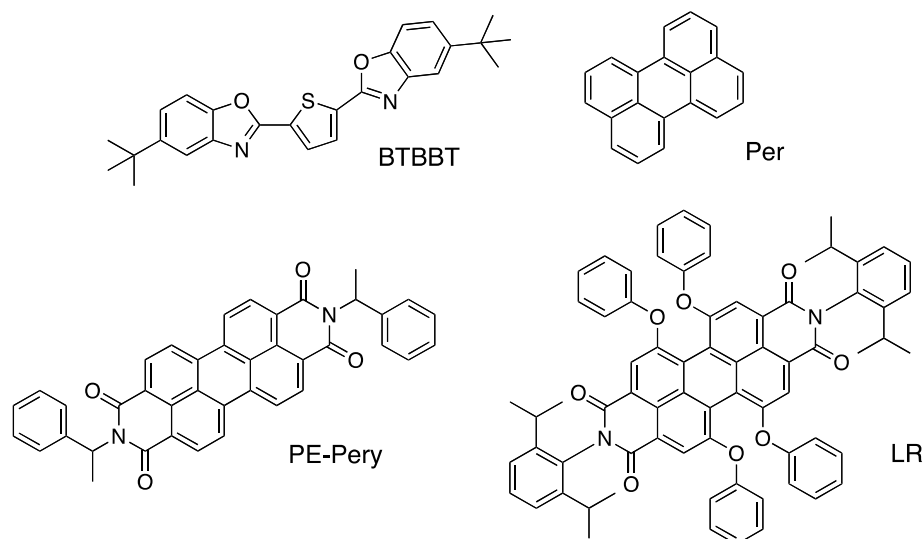

Figure 1. Chemical structures of the investigated fluorophores

The LSCs were prepared taking inspiration from the previously mentioned geometries for bulk-dyed LSCs, namely a stacked geometry that was also flanked with a cascading of emission *via* FRET. In this last design, named "*sandwich*", two distinct polymer layers containing different dyes were coated on the opposite faces of the glass slab. The top (primary) layer of the glass, i.e. that exposed to the light source, was coated by the polymer film containing the PBI dyes, that are responsible of the highest LSC optical efficiency.

Conversely, the bottom (secondary) layer of the glass was coated by the polymer film containing a selected mixture of the HEF and PBI (Scheme 1).

Moreover, compared to the ordinary stacked design, the *sandwich* LSC does not possess any air gap between the slabs. To the best of our knowledge, no study is reported on the proposed design for thin-layer LSCs.

Experimental

Materials

Poly(methyl methacrylate) (PMMA, Aldrich, M_w = 350,000 g/mol), Lumogen Red F350 (LR, BASF), 2,5-bis(5-tert-butyl-benzoxazol-2-yl)thiophene (BTBBT, Aldrich) and perylene (Aldrich) were used as received. N,N'-Bis-(1'-phenylethyl)-perylene-3,4,9,10-tetracarboxidiimide (PE-Pery) was synthetized according to the literature.[28, 29]

Preparation of polymer films for optical studies

Dye/PMMA thin films were prepared by drop casting, i.e. pouring 0.8 mL chloroform solution containing 30 mg of the polymer and the proper amount of dye to obtain concentrations in the range 0.05–2 wt.% over a 76x25x0.8 mm microscope slide. The glass slides were previously cleaned with chloroform and immerged in 6 M HCl for at least 12 h, then they were rinsed with water, acetone and isopropanol and dried for 8 h at 120 °C. Solvent evaporation was performed on a warm hot plate (about 30 °C) and in a closed environment. The thickness of the films was measured to be 25±5 µm.

Preparation of LSC samples for concentration efficiency measurements

Dye/PMMA thin films were prepared by drop casting, i.e. pouring 0.9 mL chloroform solution containing 40 mg of the polymer and a proper quantity of dye to obtain concentrations in the range 0.05–2 wt.% over a 50x50x3 mm optically pure glass substrate (Edmund Optics Ltd BOROFLOAT window 50x50 TS). Solvent evaporation was performed

on a warm hot plate (about 30 °C) and in a closed environment. Prior deposition the glass slides were cleaned as described before. The films thickness was measured to be 25±5 µm. "*Sandwich*" samples were prepared by successive depositions on the opposite faces of the glass substrates.

Apparatus and Methods

Absorption spectra were recorded at r.t. on a Perkin-Elmer Lambda 650 spectrometer. Fluorescence spectra were measured at r.t. on a Horiba Jobin-Yvon Fluorolog®-3 spectrofluorometer and equipped with a 450 W xenon arc lamp, double-grating excitation and single-grating emission monochromators. The thickness of the films was measured with a Starrett micrometer.

Photocurrent measurements[30]

A proper apparatus was build and composed by a plywood wooden box 15x15x30 cm with walls 1.5 cm thick. During the measurement a solar lamp TRUE-LIGHT® ESl E27 20W was used. The spectrum of the lamp is reported in Figure S1. Two 50x3 mm slits were carved out at 5 cm from the bottom of the box to exactly fit the LSC systems. On the outer side of the slit, a set of three 1x1 cm photodiodes (THORLABS FDS1010 Si photodiode, with an active area of 9.7 x 9.7 mm and high responsivity (A/W) in the spectral range of 400–1100 nm (Figure S2)) connected in parallel fashion was placed and coupled to a multimeter (KEITHLEY Mod. 2700).

Efficiency measurement using a PV-cell

A different set of LSC samples was prepared to measure the concentration efficiency attaching a Si-PV cell (IXYS SLMD121H08L mono solar cell 86x14 mm, with a solar cell efficiency of 14.7% and a fill factor > 70%, Figure S3) to one edge of the sample using silicone grease, while the remaining edges were covered with an aluminum tape. These devices where then placed over a white poly(ethylene terephthalate) scattering sheet

(Microcellular® MCPET reflective sheet, ERGA TAPES Srl) and placed about 20 cm under a solar lamp (TRUELIGHT® ESL E27 20W, with a correlated color temperature of 5500 K).

Results and discussion

BTBBT and Per, were selected as high-energy absorbing fluorophores (HEF), whereas PE-Pery and LR, were tested as highly fluorescent red-emitting dyes (Figure 1).

BTBBT displays blue fluorescence and shows excellent heat stability. It has found several optical applications such as optical brightener in many polymers and textiles and as scintillation dye.[31, 32] Per emits blue light as well, and it is used as a dopant material in OLEDs and as organic photoconductor.[33] PE-Pery and LR belong to a well-known class of organic dyes which present many appealing properties for LSC application[34] such as a broad absorption in the most intense part of the solar spectrum, high QY,[12][3] and good photostability.[35] Notably, LR is considered the state-of-the-art in dyes for LSC applications[4] and its use has been reported in many papers.[36-39] LR shows a quantum yield (QY) of about 1 even at high concentration (>1 mmol) in polymers and in the solid state[40] and a good photostability.[41]

When dispersed in polymer matrices such as PMMA, PE-Pery experienced dye aggregation as it was observed by the change in the absorption profile and by the appearance of a new red-shifted aggregation band in the fluorescence spectra, flanked by a progressive quenching of the dye monomer emission at around 530 nm (Figure S4).[29, 42, 43] This phenomenon moved up the main emission feature to λ higher than 600 nm, with a Stokes shift of more than 80 nm (2738 cm^{-1}). The characteristic red-shifted emission of dye aggregates was used to improve LSC performances by means of reducing the self-absorption.[44] Differently from dye aggregates with negligible fluorescence, PE-Pery aggregates in PMMA actually revealed to be strongly emissive, with QY values of about 30-40% even at the highest dye

concentration. LR in PMMA showed maximum absorption and emission bands at 573 nm and 609 nm, respectively, with a Stokes shift of 36 nm (1032 cm^{-1}) (Figure S5). LR absorbs light down to 410 nm as well, thanks to a second structureless band centered at 440 nm.

In order to assess the performances as LSC, an optically pure 50x50x3 mm glass was coated with the selected PBI dye-doped PMMA films with a thickness of 25±5 μm. Photocurrent measurements were accomplished with a home-built apparatus by using a set of three 1x1 cm photodiodes assembled in parallel fashion. This approach was used to study the best working conditions for different dye/polymer LSC systems since the response curves of the photodiodes and the utilized PV module do not differ significantly. The photocurrents measured for a set of samples based on PE-Pery/PMMA and LR/PMMA thin films are reported in figure 2.
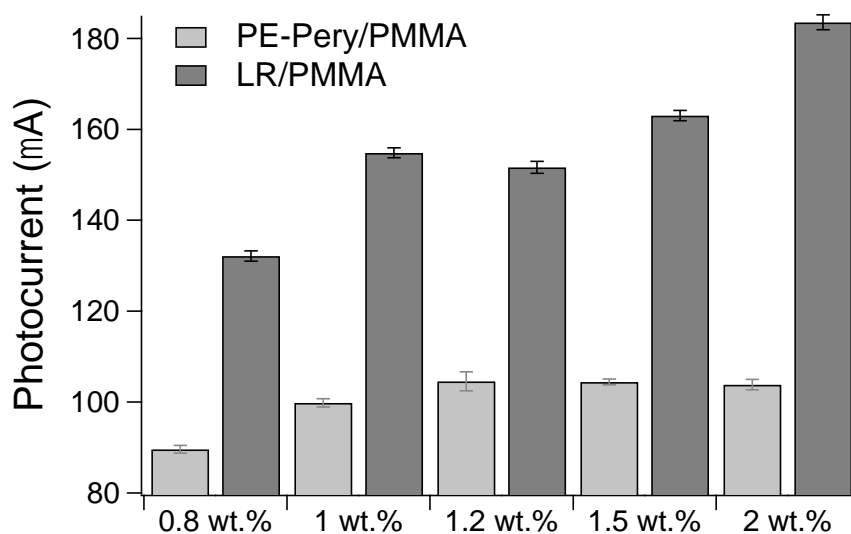

Figure 2. Photocurrent measurements for PE-Pery/PMMA and LR/PMMA films of a thickness of 25 ± 5 μm and different dye content (wt.%).

The data follow a peculiar trend, *i.e.* photocurrent increasing with fluorophore content and, for PE-Pery, a levelling off at high concentration. The higher photocurrents gathered from LR are due to the near unity QY of the dye, which is one of the fundamental features that prevents LSC efficiency losses. Notably, the lesser attitude of LR in generating self-

absorption and efficiency losses due to concentration, allowed to increase the dye content up to 2 wt.%. A complete study on the effect of fluorophore characteristics on the optical efficiencies of thin-film LSC has been recently published by our group.[30]

The LSC with the highest photocurrent were then analyzed by using a Si-based PV cell attached to one edge of the concentrator. The optical efficiency η_{opt} was evaluated from the concentration factor C, which is the ratio between the short circuit current measured in the case of the cell over the LSC edge (I_{LSC}) and short circuit current of the bare cell put perpendicular to the light source (I_{SC}) (eq. 1):

$$\eta_{opt} = \frac{I_{LSC}}{I_{SC} \cdot G} \qquad (eq. 1)$$

where G is the geometrical factor (in our case, G = 16.7), which is the ratio between the area exposed to the light source and the collecting area.

The highest optical efficiency of 7.8 % was recorded for 2 wt.% LR/PMMA LSC systems compared to the 7.0 % of the 1 wt.% PE-Pery/PMMA LSC systems (Table 1), which correspond to that gathered from LSC based on PBI in the same range of fluorophore concentration and geometrical factor.[10, 38] The different η_{opt} was addressed to the near-unity QY of LR in PMMA that render almost negligible the influence of all the fluorescence quenching mechanisms due to the dye, such as the smaller Stokes shift compared to RS-Pery.

One of the biggest challenges faced by LSCs based on the thin-layer design is the possibility to collect a large number of incident photons. Notably, a second thin layer placed at the bottom of the glass slab could collect the light transmitted through the LSC. Therefore, we realized new LSC systems with different design: the top (primary) layer of the slab, i.e. that exposed to the incident radiation, was coated with the polymer film containing the PBI dyes, whereas the bottom (secondary) layer was coated by the polymer film containing HEF or PBI fluorophores. The fluorophores are able to collect the transmitted light thanks to their main

absorption bands (HEF) and to the single chromophore $\pi-\pi^*$ transitions (PBI). The optical efficiencies of these new LSC were reported in Table 1.

Table 1. Effect of the LSC design on the optical efficiencies. The film thickness is 25±5 μm and the LSC geometry is 50x50x3 mm, where not specified. C is the ratio between I_{LSC} and I_{SC}, whereas η_{opt} is the optical efficiency

LSC design		C	η_{opt}
top layer	bottom layer		
1 wt.% PE-Pery	-	1.176 ± 0.005	7.0±0.1
1 wt.% PE-Pery (50±5 μm)	-	1.216 ± 0.005	7.3±0.1
1 wt.% PE-Pery	0.25 wt.% BTBBT	1.178 ± 0.006	7.1±0.1
1 wt.% PE-Pery	0.50 wt.% BTBBT	1.188 ± 0.007	7.1±0.1
1 wt.% PE-Pery	0.50 wt.% Per	1.191 ± 0.005	7.2±0.1
1 wt.% PE-Pery	1 wt.% PE-Pery	1.244 ± 0.010	7.5±0.1
2 wt.% LR	-	1.294 ± 0.013	7.8±0.2
2 wt.% LR	2 wt.% LR	1.352 ± 0.005	8.1±0.1

This new design appeared only substantial when PBI/PMMA thin-films were utilized as bottom layers. The light collected by HEF and emitted to the edge of the LSC is not effective for the electric current amplification possibly due to the wavelength gap between fluorophore emission range (400–500 nm, Figures 3 and 5) and the spectral quantum efficiency of the PV cell (Figure S5). Conversely, when 1 wt.% PE-Pery/PMMA or 2 wt.% LR/PMMA was employed as bottom layer, the optical efficiency increased of 7% or 4%, respectively. These performances were also compared to those of a LSC composed by a single layer of PMMA but with higher thickness (50 µm, compared to the two 25 µm thick film in the double-layer). Remarkably, the double-layer performed better than the single-layer with double thickness notwithstanding the same amount of material used (both polymer and dye). The two thin-layer and the thick one therefore absorb the same amount of radiation but the formers work with an higher efficiency.[14] This is not surprising since when thick films are used at a fixed dye concentration, a larger number of fluorescent molecules is included per unit area, thus increasing the light absorption. Nevertheless, the relation between the thickness and the

output current is not linear since the increase in the amount of polymer is flanked by optical efficiency losses caused by more probable dye-dye interactions due to the larger mean path length of the emitted radiation.

Considering these assumptions we developed a new LSC design aimed at increasing the optical efficiency. As reported in the earlier configurations, two distinct polymer layers containing different dyes were coated on the opposite faces of the glass slab (Scheme 1). The top (primary) layer of the glass, i.e. that exposed to the light source, was coated by the polymer film containing the PBI dyes, whereas the bottom (secondary) layer of the glass, was coated by the polymer film containing a mixture of the HEF and the red emitting PBI. Notably, the light transmitted by the top layer is composed by high energy photons that once collected in the bottom layer, are absorbed by the HEF and possibly re-emitted by the PBI molecule *via* FRET (Scheme 1).
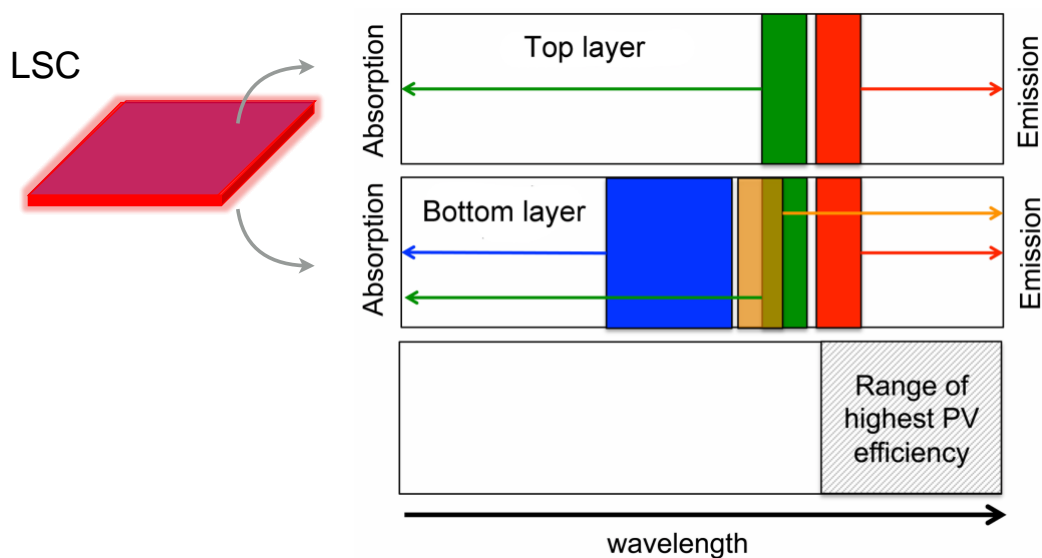
Scheme 1. Scheme of a LSC with the *sandwich* design with ideal absorption and emission ranges of fluorophores embedded in the top and bottom polymer layers coated on glass

Our first investigations were therefore focused on the study of the optical characteristics of the HEF/PBI mixture in the PMMA films. In PMMA, BTBBT absorbs in the near UV (λ_{max} =

376 nm) and its emission ($\lambda_{max} = 433$ nm) partially overlaps with the absorption ($\lambda_{max} = 493$ nm) of PE-Pery (Figure 3).

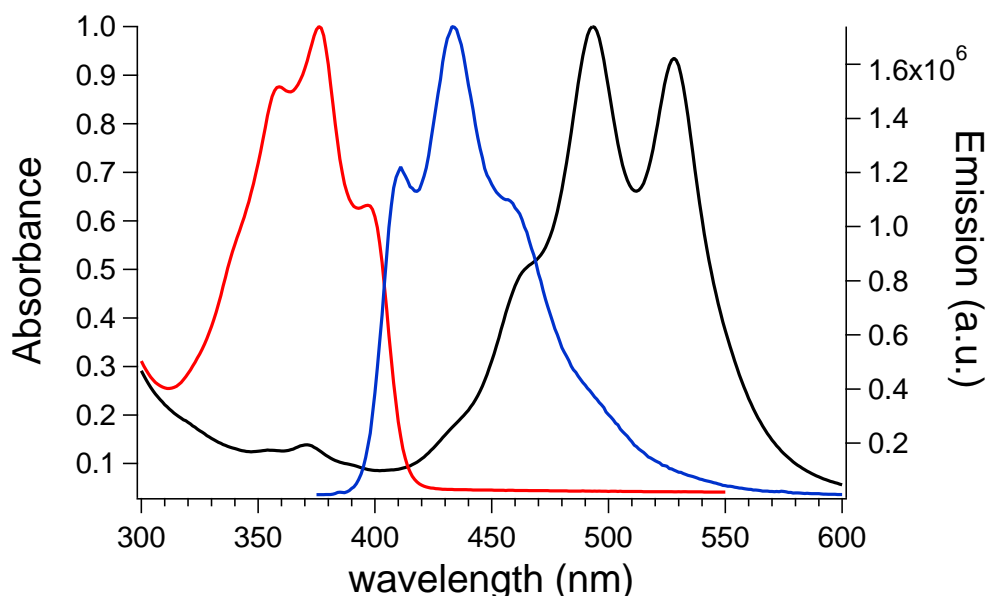**Figure 3**. Comparison between the absorption (red curve) and emission (blue curve, $\lambda_{exc.} = 350$ nm) spectra of a 0.5 wt.% BTBBT/PMMA film and the absorption spectrum (black curve) of a 1 wt.% PE-Pery/PMMA film

The BTBBT/PE-Pery mixture in PMMA containing the 0.5 wt.% of BTBBT and the 1 wt.% of PE-Pery showed almost overlapping band with the absorption features of the 1 wt.% PE-Pery/PMMA films (Figure 4a), thus indicating that adverse electronic interaction between the two dyes were negligible. Conversely, differences arise in the fluorescence spectra (Figure 4b). When excited at 350 nm, i.e. with a radiation that is almost absorbed by BTBBT only, PE-Pery experienced an evident increase of its fluorescence band upon the addition of progressive amounts of BTBBT (0.25 and 0.5 wt.%). Assuming an isotropic distribution of the fluorophores within the PMMA film (thickness = 25±5 μm, d = 0.88±0.07 g/cm^3), the mean distance between BTBBT and PE-Pery molecules in the film was estimated to be about 10 nm, thus suggesting FRET as a viable hypothesis of PE-Pery fluorescence enhancement.[45] Being such fluorophore distance just below the limit imposed by FRET mechanism, the energy transfer was considered only partial (Figure 4b).
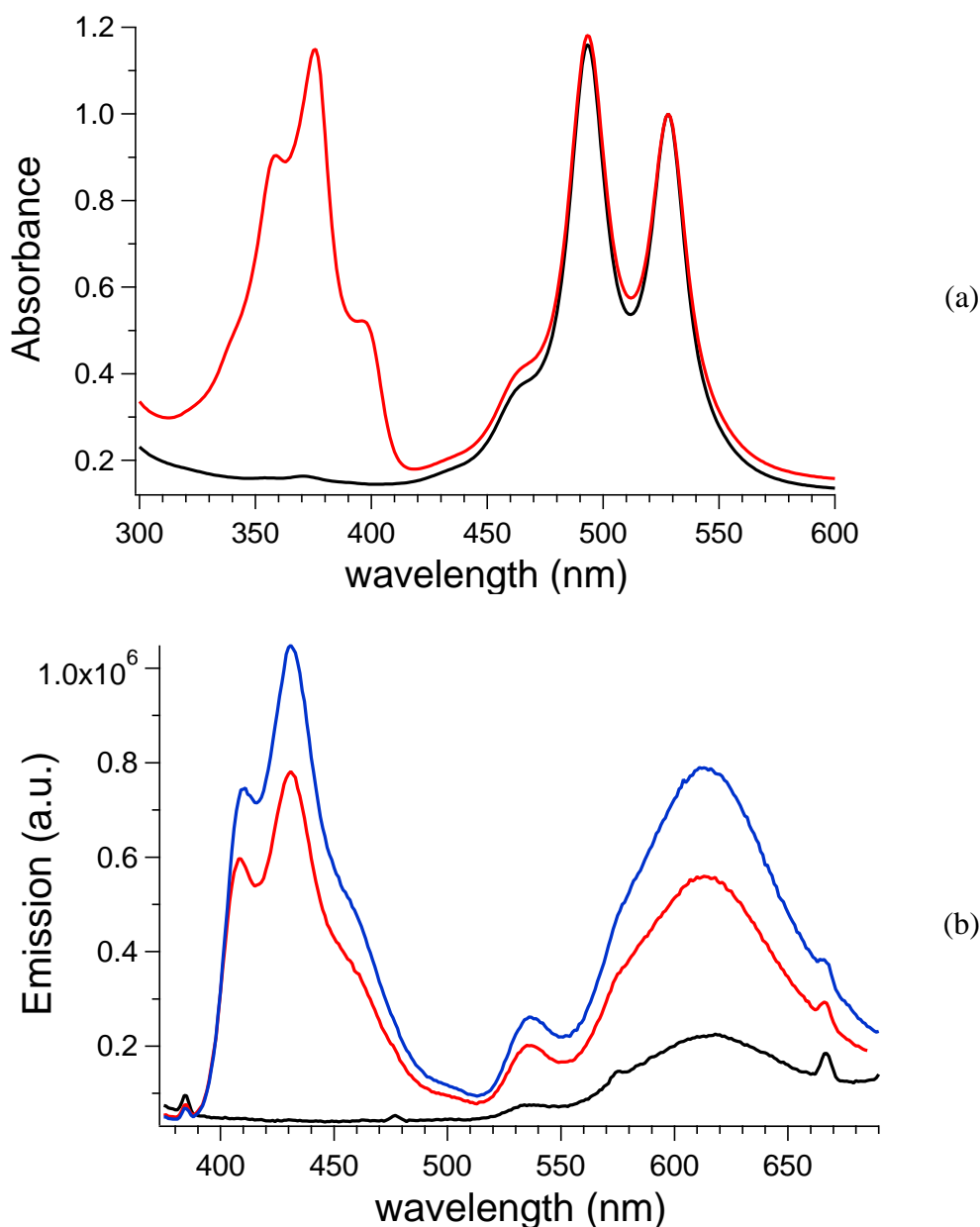
Figure 4. (a) Absorption spectra of 1 wt.% PE-Pery/PMMA film (black curve) and of BTBBT+PE-Pery/PMMA film containing the 0.5 wt.% of BTBBT and the 1 wt.% of PE-Pery (red curve); (b) Emission spectra ($\lambda_{exc.}$ = 350 nm) of 1 wt.% PE-Pery/PMMA film (black curve), of BTBBT+PE-Pery/PMMA film containing the 0.25 wt.% of BTBBT and the 1 wt.% of PE-Pery (red curve) and of BTBBT+PE-Pery/PMMA film containing the 0.5 wt.% of BTBBT and the 1 wt.% of PE-Pery (blue curve)

Compared to BTBBT, Per absorption and emission are red-shifted of about 40 nm and therefore Per overlaps better with PE-Pery absorption (Figure 5).
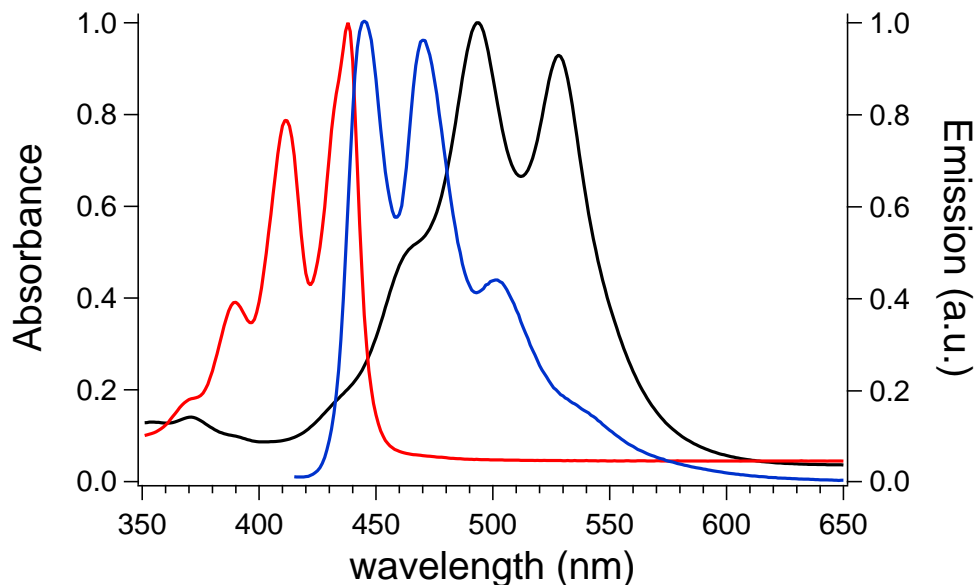
Figure 5. Comparison between the absorption (red curve) and emission (blue curve, $\lambda_{exc.}$ = 400 nm) spectra of a 1 wt.% Per/PMMA film and the absorption spectrum (black curve) of a 1 wt.% PE-Pery/PMMA film

Apparent differences are however observed when comparing the optical features of 1 wt.% PE-Pery/PMMA film with those of PER/PE-Pery mixture in PMMA, i.e. containing the 0.3 wt.% of Per and the 1 wt.% of PE-Pery (Figure 6a). The spectra differ significantly in these two cases, both concerning the line shape and the maxima positions. This phenomenon is possibly addressed to the strong $\pi-\pi$ interactions between Per and PE-Pery aromatic backbones which alter the supramolecular structure of PE-Pery and therefore its absorption features.[43, 46, 47] These variations were also noticed in fluorescence (Figure 6b) by studying various PMMA films containing a fixed amount of PE-Pery (1 wt.%) and a progressive content of Per (from 0 to 2 wt.%).
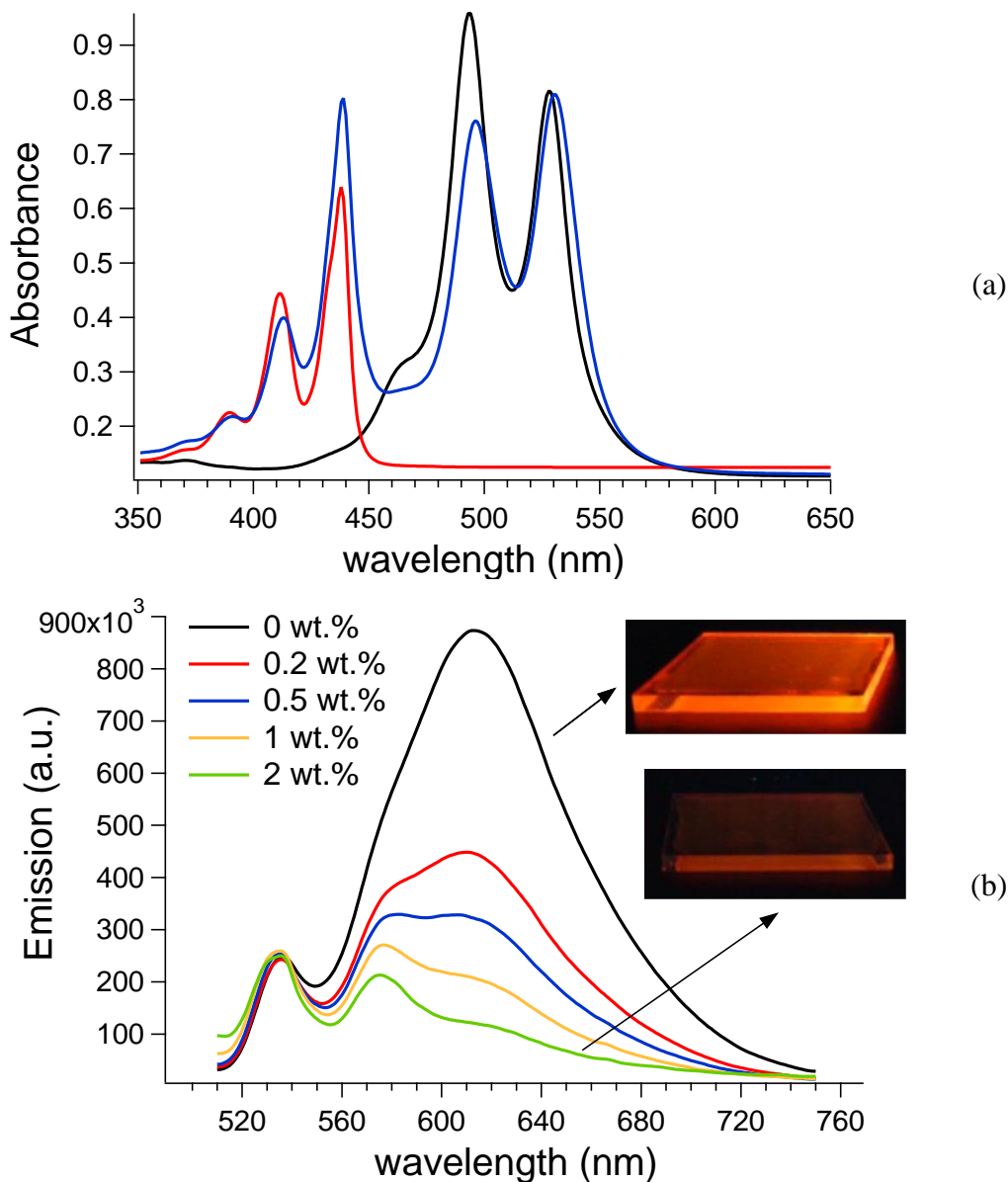
Figure 6. (a) Absorption spectra of 1 wt.% PE-Pery/PMMA film (black curve), of 0.3 wt.% Per/PMMA film (red curve) and of Per+PE-Pery/PMMA film containing the 0.3 wt.% of Per and the 1 wt.% of PE-Pery (blue curve); (b) Emission spectra ($\lambda_{exc.}$ = 450 nm) of 1 wt.% PE-Pery/PMMA film (black curve) and of 1 wt.% PE-Pery/PMMA films containing progressive amounts of PER. The pictures of the films coated over glass were taken under the excitation at 366 nm and reported as insets.

The resulting spectra show that increasing Per concentration, PE-Pery fluorescence at about 620 nm attributed to aggregate emission is rapidly quenched compared to the emission of isolated PE-Pery fluorophores at about 540 nm. The addition of Per molecules limits therefore the formation of PE-Pery aggregates, thus preventing the development of the red-emitting

band, as well evidenced by the LSC pictures taken at 366 nm (inset of figure 6b). Therefore, considering this adverse effect on the PE-Pery emission, Per was not considered for the determination of LSC performances.

The new *sandwich* designs were then tested and the relative η_{opt} reported in Table 2 and compared with the original LSC in figure 7.

Table 2. Effect of the LSC *sandwich* design on the optical efficiencies. All the polymer layers have a thickness of 25±5 μm and the LSC geometry is 50x50x3 mm. C is the ratio between I_{LSC} and I_{SC}, whereas η_{opt} is the optical efficiency

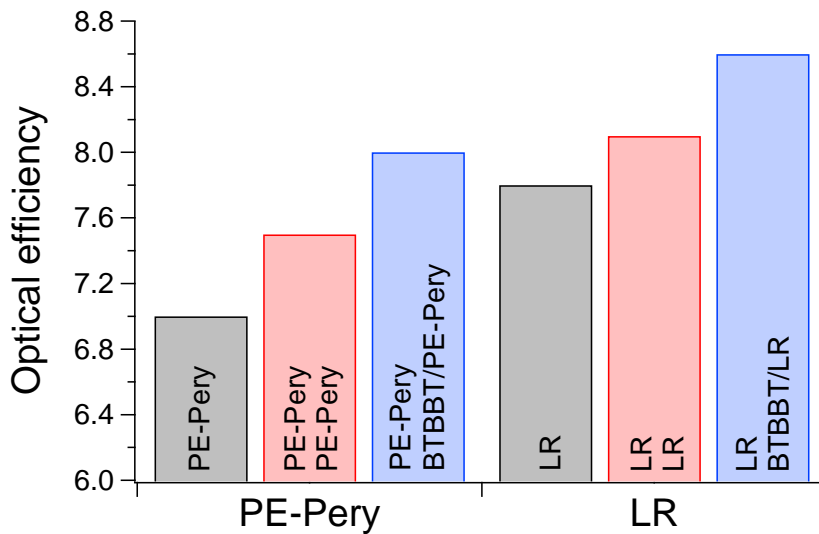| Top layer	Bottom layer	C	η_{opt}
1 wt.% PE-Pery	1 wt.% PE-Pery + 1 wt.% BTBBT	1.269 ± 0.008	8.0±0.1
2 wt.% LR	2 wt.% LR + 1 wt.% BTBBT	1.370 ± 0.025	8.6±0.2

Figure 7. Optical efficiencies calculated for the different 25±5 μm PMMA based on single-layer (gray bar), double layer (red bar) and *sandwich* LSC.

The *sandwich* LSCs showed a distinct increase of η_{opt} with respect to both single and double layer designs. Notably, the addition of the 0.5 wt.% of BTBBT in the bottom 1 wt.% PE-Pery/PMMA layer (blue bar) increased η_{opt} of 14% and 6.7% with respect to the single layer (gray bar) and to the double layer (red bar) LSCs, respectively. This gain in optical efficiency

agrees well with the spectroscopic investigations, that is, the energy hopping from the short-wavelength absorber to the red-emitting fluorophore appears extremely effective, notwithstanding the partial overlap between BTBBT emission and PE-Pery absorption. Furthermore, an improvement in the optical efficiency was also reported for LR/PMMA films, i.e. 10.7% and 6.2% with respect to single (gray bar) and double layer (red bar) designs, respectively, thus reaching the maximum absolute η_{opt} of 8.6.
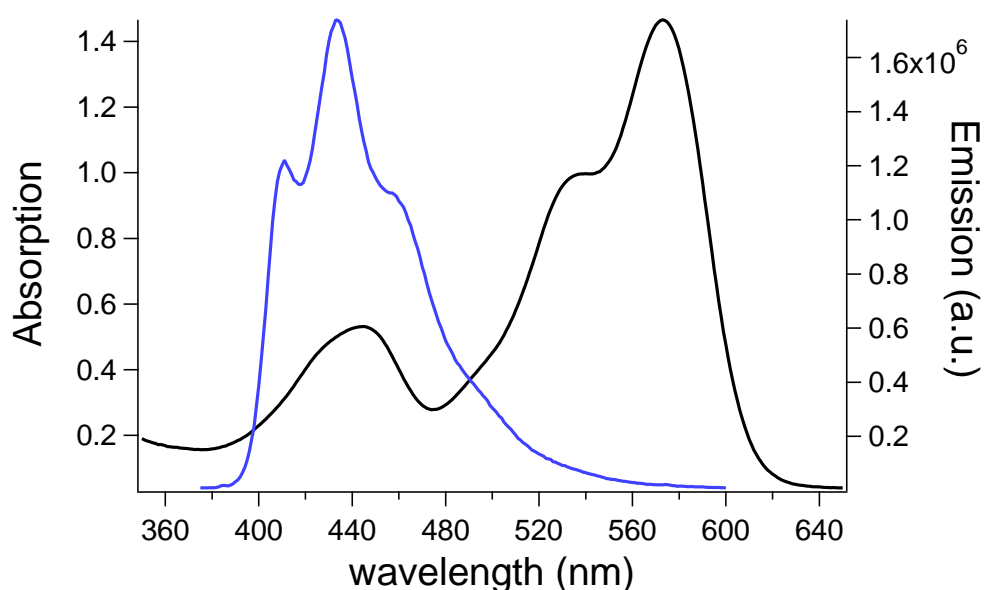

Figure 8. Comparison between the emission spectrum (blue curve, $\lambda_{exc.}$ = 350 nm) a 0.5 wt.% BTBBT/PMMA film and absorption (black curve) spectrum of a 2 wt.% LR/PMMA film

The lower gain in optical efficiency in the case of LR can be attributed to a smaller overlap (~8%) between BTBBT emission and LR absorption compared to that of PE-Pery (Figure 8).

Conclusions

We have demonstrated that multi-layer stacked LSCs based on PMMA thin-films doped with HEF fluorophores and PBI red-emitting dyes are able to increase the optical efficiencies of a maximum of 14% with respect to conventional single-dye LSCs. Different strategies were proposed aimed at increasing the light collection by the LSC. LSCs were prepared taking

inspiration from the stacked geometry, i.e. combining two fluorescent PMMA layers coated on the different surfaces of a glass slab. In the most effective design (*sandwich*), the top layer of the glass was coated by the polymer film containing the PBI molecules that are responsible of the highest LSC optical efficiency. Conversely, the bottom layer was coated with the polymer film containing an appropriate mixture of HEF and red emitting PBI. In this configuration, the light transmitted through the slab is absorbed by HEF and efficiently re-emitted by the PBI molecule *via* FRET, thus amplifying the electric current production.

In light of these peculiar features, the *sandwich* LSC design showed η_{opt} 10–14% greater than the maximum values gathered from LSC based on single PBI fluorophores.

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