

# Absorption coefficient dependent non-linear properties of thin film luminescent solar concentrators

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## ABSTRACT

This research article studied absorption coefficient dependent losses and efficiency of thin film luminescent solar concentrator (LSC). The optimum absorption coefficient was determined by fabricating red dye doped thin film LSCs with varying absorption coefficients of 24–202 cm<sup>-1</sup>. The optical, electrical, and ray-trace model predicted results were compared to establish the optimum absorption coefficient for fabricated thin film LSCs. The optical and electrical efficiencies of thin film LSCs are a function of absorption coefficient; follow linearly at lower values, gradually becoming non-linear at higher values of absorption coefficient which restricts the overall efficiency. The multi layered ray-trace model predicted results deviated from the experimental results at high absorption coefficient. Non-linearity was introduced by losses mechanism of re-absorption, resonant energy transfer (RET), and scattering in thin film LSCs. Re-absorption losses were found to be dependent on optical path-length and absorption coefficient of thin film. They were identified and quantified through locally collected emission and optical path-length in thin film LSCs. Re-absorption losses caused a red shift in emission peak by ≈ 20 nm as the absorption coefficient increased from 24 to 202 cm<sup>-1</sup>. RET decreased fluorescence life time of dye from 5 ns to 2 ns for the absorption coefficient increased from 24–202 cm<sup>-1</sup>. These absorption coefficient dependent losses limit the optical and electrical efficiency of thin film LSC devices, and compared with bulk LSC.

## 1. Introduction

Luminescent solar concentrator (LSCs) technology was proposed in the late 1970s [1–3] as a means to concentrate solar radiation onto a smaller area of solar cell, and hence, enhance the solar cell output. The main objective of this technology is to replace the large area of solar cells in a standard flat-plate photovoltaic (PV) panel by an inexpensive polymer collector, thereby reducing the cost of the module and consequently of solar power generation. LSCs have advantages over alternative concentrating systems: they concentrate both direct and diffuse radiation since they are not subjected to a solar concentration ratio limitation [4]; they have a narrow emission band of luminescent species that can be matched to the spectral response of solar cells [5]; they minimize thermalization losses for PV cells [5]; stacking LSC plates containing luminescent species of different absorption characteristics can separate the solar spectrum, and concentrate different parts simultaneously [6]; and they are well suited for building integrated photovoltaic (BIPV) due to their static nature [7–9].

In LSCs, incident solar radiation is absorbed by luminescent material, and subsequently, light is re-emitted over all solid angles. The re-

emitted light hitting the waveguide at angles larger than the critical angle is guided via total internal reflection (TIR) to the sheet edges where solar cells are attached. Generally, three surfaces are mirrored to ensure that light can only emerge at the fourth edge where PV cells are located, and an air gap between the LSC sheet and the mirror ensures total internal reflection. LSCs are based on the luminescence down-shifting process therefore there is an inherent loss of energy due to internal energy conversion in the luminescent species, leading to a Stokes-shift in the re-emitted radiation [10]. The overall optical and electrical efficiency of LSCs are limited by re-absorption [10–12], escape cone losses [13] scattering [12], narrow absorption range [14,15], photostability of the luminescent material [16].

Doping concentration of luminescent material and absorption coefficient have a strong influence on determining the efficiency of LSC devices [12,17]. The absorption coefficient can be related to the total emission and energy loss processes of re-absorption, scattering and resonance energy transfer (RET) in LSCs. At higher absorption coefficients, quantum yield of the luminescent material is modified through increased non-radiative RET between molecules [18,19], and amplified through re-absorption losses [11,12]. In addition; there is a higher

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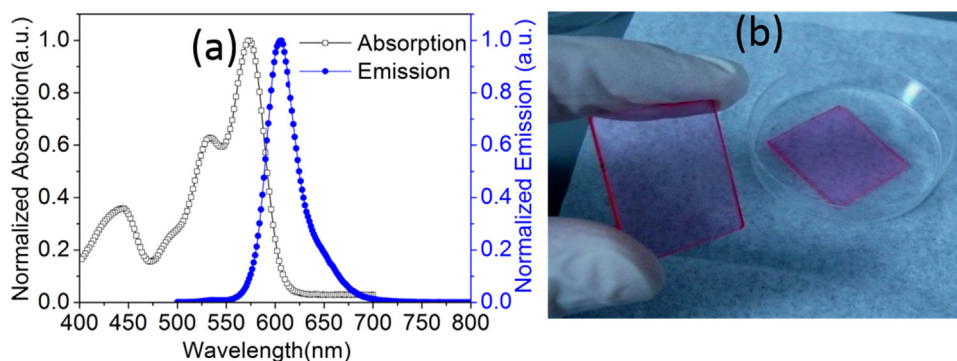


Fig. 1. a) Normalized absorption and fluorescence emission of 0.001 wt% dye in toluene. The peak of absorption and emission are 575 and 606 nm, respectively; b) thin film LSC plate fabricated by spin coating of dye doped PMMA polymer on glass substrate.

probability of forming molecule clusters through aggregation of molecules that can induce scattering losses [20]. All these together adversely affect the efficiency of LSC devices. In order to improve the LSCs efficiency, significant advances have been made in terms of the use of quantum dots [17,21,22], rare earth metals [23], aligning luminophore [24], and stacked structured [25], plasmonic interaction improved fluorescence [26], and thin film LSCs [20,27]. The organic dye was replaced with quantum dots in quantum dot solar concentrator [22]. Quantum dots were expected to have improved photostability being a semiconductor crystalline structure [28], however, have low fluorescent quantum yield of 20–50% [29].

LSC systems are divided into two categories based on their thickness; thin film LSCs and bulk LSCs. Decreasing the thickness of LSCs, increase the optical concentration ratio [15,30]. Where geometric gain,  $G_{\text{geom}}$  is defined as follows

$$G_{\text{geom}} = \frac{A_{\text{surface}}}{A_{\text{edge}}} \quad (1)$$

where  $A_{\text{surface}}$  and  $A_{\text{edge}}$  are the areas of the active collector surface and edge, respectively. The geometric gain in combination with optical efficiency ( $\eta_{\text{opt}}$ ) determines the theoretical limit of the solar concentration ratio  $C_{\text{edge}}$  which is the ratio of irradiance ( $\text{W}\cdot\text{m}^{-2}$ ) received at the LSC edge to the incident absorbed irradiance;

$$C_{\text{edge}} = \frac{\eta_{\text{opt}} A_{\text{surface}}}{A_{\text{edge}}} \quad (2)$$

where  $\eta_{\text{opt}}$ , the optical efficiency, is the ratio of incident radiation delivered to the edge of the LSC. The thin film LSC consists of a thin film of polymer with heavily doped luminescent species (organic dye/quantum dot/rare-earth material) coated on highly transparent thicker substrate of the same refractive index to the thin film [15], and fluorescence emission is primarily trapped in the substrate. The bulk LSC has low doping concentration and fluorescence emission trapped in entire volume of LSC.

The luminescent materials quantity in the thin film LSC is intended to be the same as in a comparable size bulk LSC [31,32]. Concentrated in a smaller volume of thin film only, hence the thin film LSC has higher absorption coefficient. The refractive index matching between the thin film and the transparent substrate ensures efficient coupling of fluorescence emission from thin film to substrate, where it is trapped and wave-guided by total internal reflection to the edge of the plate, with reduced parasitic losses of re-absorption and scattering [31] compared to bulk LSCs. The reduced scattering and re-absorption losses concept for thin film LSCs has been debated [20], where it has been argued that the gain in optical path-length in the substrate is compensated by the losses within the optically dense thin film.

However, thin film LSCs have several advantages: they enable investigation of stacking LSC plates containing different types of luminescent species [33]; reduced fabrication cost since it requires less

doped material [20]; thin films can be coated inside glass and therefore luminescent centres will be protected against UV radiation damage; a very high concentration of luminescent material is easily achieved [8]. This article discusses these arguments through experimentally and predicted results from a ray-trace model.

Monte Carlo ray-trace theoretical model has been applied to optimize bulk LSC device, especially the quantum dot solar concentrator [34–37]. However, this research extends this work and develops a multi-layer ray-tracing model to model thin film LSCs and is validated by comparing predicted and measured results. It provides an accurate tool for quantifying energy loss mechanisms and optimizing thin film LSCs for a given set of material parameters. The ray-trace model predicted results are compared with those experimentally obtained.

## 2. Material and methods

Perylene based Lumogen F Red305 dye (BASF, Germany) was used as a luminescent material (referred as dye) – typical absorption and fluorescence emission spectra are presented in Fig. 1a.

Thin film LSCs were fabricated by spin coating (G3 P-8 series spin coater) dye doped acryl-resin polymer (Plexit 55, Carl Roth GmbH + Co. KG, Germany) on a clean glass substrate of  $25 \times 25 \times 1$  mm. The acryl-resin polymer Plexit 55 (5000 mPa s at 20 °C) which is a pre-polymerized mixture of 65–70% MMA, 30–35% PMMA and 2% photoinitiator. In the typical process, the highly viscous polymer Plexit 55 was diluted to 25 wt% in toluene and doped with varying dye doping concentrations from 0.1 to 0.9 wt%. Subsequently the mixture was spin coated on glass substrate to achieve 10  $\mu\text{m}$  thin film. Thin films were thermally polymerized for 24 h at 60 °C in a vacuum oven to prevent oxygen penetration during polymerization – a fabricated thin film LSC is shown in Fig. 1b. The thickness of the film was 10  $\mu\text{m}$  and measured using an atomic force microscope.

### 2.1. Optical characterization techniques

The absorption and fluorescence emission measurements were carried out using a Perkin Elmer Lambda 900 UV/Vis/NIR spectrometer and Perkin Elmer LS55B Luminescence spectrometer, respectively. An AvaSpec-2048 (Avantes, UK) fiber optic spectrometer was used to record the fluorescence emission at the edge of thin film LSCs; the optical setup is illustrated in Fig. 2a. The detector is close to the edge to stop detection of stray fluorescence emission.

A Zeiss LSM 510 confocal laser scanning microscope was used to measure the local fluorescence emission in thin film LSCs. The confocal microscope fluorescence emission is collected from thin film only rather than combined thin film and glass substrate. It is possible to control the depth of the excitation field and eliminate out-of-focus light, because the confocal arrangement allows manipulation of the focal plane. The focal plane was adjusted at the interface of the thin film and glass

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