



# Investigation of Tin(II)2,3-naphthalocyanine molecule used as near-infrared sensitive layer in organic up-conversion devices



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## ABSTRACT

In this work, a near infrared (NIR) sensitive molecule, Tin(II)2,3-naphthalocyanine (SnNc) was characterized by different techniques. UV–Vis spectroscopy and cyclic voltammetry were performed in order to determine the absorption spectrum, the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) energies of this molecule deposited in form of thin films. We found energies of  $5.0 \pm 0.1$  eV and  $3.7 \pm 0.1$  eV for HOMO and LUMO, respectively. The charge carrier mobility was also investigated by space charge limit current technique showing values of  $\mu_h$   $(8.8 \pm 0.1) \times 10^{-5}$   $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$ . SnNc alone or blended with fullerene was used as efficient NIR sensitive layer due to its absorption around 875 nm. The crystalline structure of SnNc was studied by X-ray powder diffraction, showing a monoclinic system and  $P2_1/c$  space group, with cell parameters  $a = (15.948 \pm 6)$  Å,  $b = (15.818 \pm 2)$  Å,  $c = (14.649 \pm 1)$  Å and  $\beta = (67.096 \pm 6)$  Å. Desorption/Ionization-Time of Flight Mass Spectrometry (LDI-TOFMS) technique was employed to obtain information of molecular structure of the SnNc in thin film, showing that the thin film of the SnNc has no dimer formation. Due to its absorption around 875 nm, SnNc blended with fullerene was used as efficient NIR sensitive layer in the fabrication of an organic up-conversion device. When the device is submitted to IR radiation, a gain of about 133% was observed in the luminous efficiency when compared to values without IR irradiation.

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## 1. Introduction

Organic light-emitting diodes (OLEDs) have been the subject of research since the first discovery of electroluminescence in molecular and polymer thin films [1,2]. However, researchers have also shown interest in another type of organic device: the light converter [3]. Recently [4], an organic near infrared-to-visible up-conversion device using tin phthalocyanine-fullerene (SnPc:C<sub>60</sub>) bulk hetero-structure layer as near infrared (NIR) sensitizer, was reported showing high current efficiency and high up-conversion ratio.

Organic up-conversion devices are able to pick up signals from

the infrared region of the electromagnetic spectrum generating visible radiation. The structure of these devices basically consists of two elements: (i) an organic thin sensitive layer to NIR radiation and (ii) an OLED emitting in the visible region. The OLED emits light only when NIR radiation is absorbed by the NIR sensitive layer, producing a significant increase in charge carriers. Conventional light converter technologies, night vision and medical diagnose [5,6] could be replaced by up-conversion devices, since organic devices are lightweight and highly compatible with flexible substrates. They are also easily processed, have low cost, and allow fabrication of large area devices.

When compared with simple phthalocyanines (PCs) the Naphthalocyanines (Ncs) present important modifications of their optical and electronic properties due to their extended delocalized  $\pi$  electron system. The light harvesting properties of Ncs are also superior when compared to Pcs, which makes them ideal candidates for use in optoelectronic devices [7–9].

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In this paper, we investigate the use of Tin (II) 2,3-naphthalocyanine (SnNc) alone or co-deposited with fullerenes (SnNc:C<sub>60</sub>) as organic NIR sensitive layers for the fabrication of efficient up-conversion devices. SnNcs belong to a class of highly stable organic compounds, which are classified as p-type semiconductors characterized by low mobility and low carrier concentration [10].

## 2. Material and methods

All devices were prepared on glass substrates previously cleaned with liquid detergent, acetone, isopropyl alcohol, and deionized water. Then, the substrates were blown dry with N<sub>2</sub> and treated in UV-O<sub>3</sub> atmosphere at 100 °C during 15 min. All the organic compounds were obtained by Lumtec (Luminescence Technology Corp.) except SnNc purchased from Sigma-Aldrich and they were used without further purification. Thin films of SnNc can be prepared by thermal evaporation technique because of the high stability of this molecule. Organics and metals were evaporated under high vacuum environment with a pressure of  $1 \times 10^{-6}$  Torr and the evaporation rate was 0.1 nms<sup>-1</sup> for organic materials and 0.1 nms<sup>-1</sup> for metals. All the layers were deposited at room temperature. For the fabrication of the OLED, N,N'-Bis(naphthalene-2-yl)-N,N'-bis(phenyl)-benzidine ( $\beta$ -NPB) was used as hole transporting layer, Tris(8-hydroxy quinoline) aluminum (Alq<sub>3</sub>) was used as electron injection layer and emitting layer. As organic NIR sensitive layer a film SnNc codeposited with C<sub>60</sub> was used Indium-tin-oxide (ITO) with a sheet resistance of 15  $\Omega$ /sq and a 100 nm thickness aluminum layer were used as anode and cathode electrodes, respectively. A CW 830 nm diode laser with a power density of 65 mWcm<sup>-2</sup> was used to excite the SnNc:C<sub>60</sub> layer. At this wavelength there are no absorption bands from Alq<sub>3</sub> and NPB, while the tin naphthalocyanine present a good absorption coefficient. The device area was 0.03 cm<sup>2</sup> and all the characterizations were performed at room temperature. The fabricated up-conversion devices were not encapsulated.

UV–VIS and cyclic voltammetry were used to determine the absorption spectrum, the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) of the molecules deposited in form of thin films.

The absorption spectra were acquired using a PerkinElmer Lambda 950 spectrometer. The cyclic voltammetry measures were obtained through a IVIUM Electrochemical Interface & Impedance Analyser with graphite as work electrode, platinum as counter electrode and (Ag/AgCl) as reference electrode. The aqueous base electrolyte was KCl 1 mol/L.

The luminances were measured directly by a calibrated Konica Minolta Luminance Meter LS-100. The current–voltage (I–V) characteristics were measured with a programmable voltage current source (Keithley 2400).

The structural analysis of SnNc was performed by using x-ray powder diffraction technique (XRD). The samples were gently grinded in an agate mortar and the fine powder obtained was deposited in the sample holder equipped with a zero-background silicon plate. The diffraction data were collected by means of overnight scans in the 2 $\theta$  range of 5–105° with steps of 0.02° using a Bruker D8 Advance Da Vinici diffractometer equipped with a Ni-filtered CuK $\alpha$  radiation ( $\lambda = 1.5418$  Å) and a Lynxeye linear position-sensitive detector. The following optics were set up: primary beam Soller slits as 2.94°, fixed divergence slit as 0.3° and receiving slit as 10 nm. The generator was set at 40 kV and 40 mA. Standard peak search was used, followed by indexing through the single-value decomposition approach implemented in TOPAS [11].

In order to confirm non-formation of dimers and trimers in SnNc thin film, Laser Desorption/Ionization-Time of Flight Mass

Spectroscopy (LDI-TOFMS) experiment was performed using a pulsed nitrogen ultraviolet laser ( $\lambda = 337$  nm) of an AXIMA Performance MALDI ToF (Shimadzu Biotech). The experimental setup includes an automatic sample manipulator, where 200 scans were accumulated with 20 repetitions each. The system was operated in high vacuum chamber with a base pressure of about  $3.0 \times 10^{-7}$  Torr. We used positive ion reflectron mode.

## 3. Results and discussion

Carrier mobility is one of the most important parameters in organic electronics. This substantial parameter is used to evaluate the potential of new materials for device applications. Several techniques have been developed to extract the charge mobility, such as: time-of-flight (TOF) [12], dark-injection transient current (DITC) [13], charge extraction by linear increase voltage (CELIV) [14,15], injection limited current (ILC) [16,17], space-charge limited-current (SCLC) [18–20] and others, which leads to a big divergence to the values of mobility of carries in the literature, especially for the case of organic semiconductors [21,22]. Based on the difficulty to find reliable values for carrier mobility of thin films of phthalocyanines, we used the protocol for extracting a space-charge limited mobility benchmark from a single hole-only or electron-

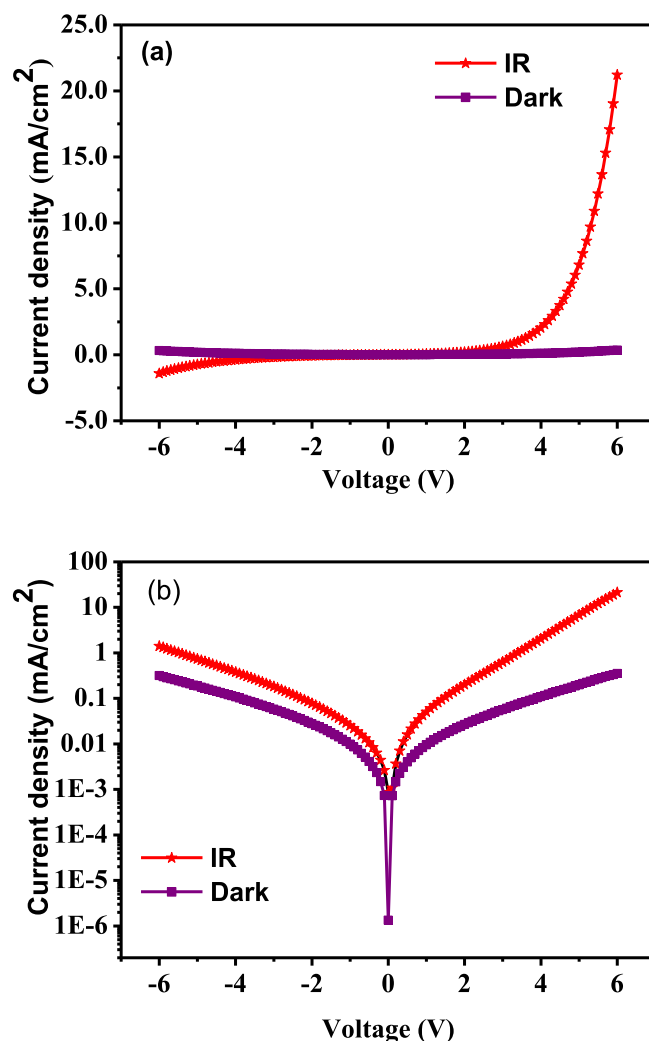


Fig. 1. (a) Current density versus voltage and (b) absolute values of current density in dark and under IR radiation for a 30nm film of SnNc.

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