



Letter

Nano graphite platelets enhanced blue emission in alternating current field induced polymer based electroluminescence devices using Poly (9,9-dioctylfluorenyl-2,7-diyl) as the emitter



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ABSTRACT

We report a simple way of enhancing blue emission in Poly (9,9-dioctylfluorenyl-2,7-diyl) (PFO) based alternating current field driven electroluminescence devices by mixing graphite nano platelets in the emission layer. Significantly strong and well resolved blue peaks at 437 nm and 470 nm are observed both in the devices' electroluminescence and photoluminescence spectra with the presence of graphite nano platelets. The origin of this strong blue emission has been identified as the PFO's beta-phase formation confirmed by transmission electron microscopy images and UV–Vis absorption spectra. The nano platelets loading in PFO solution can be controlled by the dispersion time in organic solvents. When the loading increases, the green emission in the electroluminescence spectrum can be reduced. This technique has been found useful in the generation of bright white light when illuminating through down conversion phosphor.

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1. Introduction

Polymer-based light emitting devices (PLED) have been well developed in recent years due to the great potential for cost effective production on a large scale. Alternating current (AC) field induced electroluminescence devices using polymers as emitters are drawing great attention lately because of their potential long life time and convenient lighting applications, wiring directly to the household [1–3]. The emission layer (EML) is located within a capacitor and can be driven by an electric field without a large current flowing through [4]. It is easy to control the emission colors using organic materials and thus realize either pure color or natural white for household lighting purposes [5–8]. Different device configuration/materials have been demonstrated and achieved great progress in AC EL devices [1,3,4,9].

Polyfluorenes (PFs) are excellent candidates as blue emitters due to their high photoluminescence (PL)

quantum efficiency [10], high charge mobility [11], and potential pure color emission [12]. Other than the material chemical properties, the emission peak intensity and positions are also correlated with the polymer film morphology. Poly (9,9-di-*n*-octyl-2,7-fluorene) (PFO), for example, has been proven to have various crystal forms [13–15], in which the beta phase exhibits strong and narrow singlet 0-0 and 0-1 emission corresponding to ~430 nm and ~460 nm. It is important to achieve such a phase since ordered polymer chains can provide an effective path for charge transfer and confines the excited state away from the quenching site, therefore, resulting in higher luminescence efficiency [16]. Various approaches have been proven to be effective treatments to achieve beta phase in PFO based organic light emitting diodes (OLED), such as cyclic heating and cooling [17], solvent vapor exposure [18], side chain control [19,20], etc.

However, when using PFO as a blue emitter, it is difficult to maintain such performance in AC-driven devices under this high voltage. An undesirable low energy green emission could appear and dominate the electroluminescence (EL) spectrum which degrades the color purity

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[21]. A number of studies have explored the various origins of the green emission, such as thermal or photo oxidation degradation [22], polymer chain reorganization, on chain defects [20,23] or aggregation during processing, and inter-chain/segment excimer emission during device operation [24].

In this letter, we report a novel way of controlling the PFO film morphology for strong blue electroluminescence emission in this new type of AC driven field induced electroluminescence devices by doping nano graphite platelets (GNPs), or thick (many layered) graphene platelets, in the polymer matrix. The film morphology change by adding GNPs was observed by transmission electronic microscopy (TEM) and confirmed by photoluminescence and absorption spectrum on the film. A potential application is in white light generation when illuminating through a layer of Ce^{3+} doped Yttrium Aluminum Garnet (Ce:YAG) [25,26], which has been proven to be an effective down conversion phosphor for UV or blue light to white light conversion.

2. Experiment

GNPs are used as purchased (Angstrom Materials Inc, 99.0%) with an average size smaller than 40 μm and thickness between 50 and 100 nm. They were pre-dispersed in 1,2-dichlorobenzene (oDCB) by probe sonication and the upper clear solution was removed after centrifuge at 5000 rpm for 10 min. The stock solution was aged for a few days and remained stable for weeks. 200 μl graphite dispersion was then mixed with 1 ml of a solution containing 12 mg PFO in chlorobenzene (CB) that had been stirring for over 10 h. The device fabrication process has been reported in our previous work using the asymmetric device structure of ITO/dielectrics/emitter/Aluminum15 using the stock solution. The control group was also fabricated with PFO dissolved in CB with the same amount of pristine oDCB for comparison.

AC sinusoidal voltages were applied from a 200 MHz function/arbitrary waveform generator (Agilent 33220A) connected to a Model PZD700A M/S amplifier (Trek), while the current and voltage across the device was measured on an oscilloscope (Tektronix). The EL spectra were collected with an ILT 950 spectroradiometer. The film morphology was observed with a JEOL 1200 transmission electron microscope (TEM) with an acceleration voltage of 80 kV.

3. Results and discussion

The asymmetric device configuration is shown in Fig. 1a, where only ITO and aluminum connected with an alternating current (AC) power supply were used as contacts for simplicity. The charges were injected through the aluminum side while the ITO side was blocked by the poly(vinylidene fluoride-trifluoroethylene) [P(VDF-TrFE)] di-block copolymer dielectric layer ($\sim 2.4 \mu\text{m}$). We have shown two types of AC-EL devices [4] in our previous study. One of them had both of the contacts been isolated from the emission layer by insulator so the charge can only be generated in the active layer. Another structure had one

contact insulated and charge can be injected alternatively through the other contact. Similar device operation has been demonstrated by Sung et al. [1]. In this study, we employed the device configuration as: the ITO is insulated by thick PVDF-TrFE layer ($\sim 2.4 \mu\text{m}$) and the emission layer is making contact with Aluminum. Therefore the hole cannot be injected through the ITO side. We thus propose that both electron and holes are injected through the aluminum side. Although such device configuration is not the best approach for electron/hole injection due to the work function mismatch between aluminum and PFO's HOMO and LUMO levels, we use such configuration for simple film morphology study. According to the similar behavior of the current–voltage curve for the non-doped and doped devices (data in supporting information), the graphite doping did not change the impedance of the whole device so we will focus on the film morphology change induced by graphite doping and its influence towards device performance.

We note our devices are driven under high voltage. We attribute this for using very thick dielectric layer ($\sim 2.4 \mu\text{m}$) and a thin emission layer ($\sim 100 \text{ nm}$). So if we consider the two layers as two capacitors connected in series, together with the dielectric constant (PVDF ~ 9 , conjugated polymer ~ 3) and the thickness of each layer, one can estimate the voltage drop on each of the layers roughly by Eqs. (1)–(3):

$$C = \varepsilon \frac{A}{d} \quad (1)$$

$$C_1 V_1 = C_2 V_2 \quad (2)$$

$$V = V_1 + V_2 \quad (3)$$

where ε is the dielectric constant, A is the plate area and d is the layer thickness; C_1 , V_1 and C_2 , V_2 refer to the capacitance and voltage on dielectric layer and emission layer respectively. By careful calculation, the peak to peak voltage drop on emission layer is 10–16 V, and in each AC period, the peak voltage should be 5–8 V.

It is interesting to note the doped film in Fig. 1c forms ordered parallel chains (bright fibers) radiating away from GNPs (dark rods) compared to the pristine PFO film in Fig. 1b that shows randomly distributed domains. According to the selective area diffraction pattern (SAD) in the inset of Fig. 1b and c, the pristine film showed a diffused and dim ring while the graphite nanoplatelets doped film showed a strong and sharp ring, which indicate a more ordered polymer structure in the bulk film with GNPs. This proves that the polymer chains are more ordered in the doped film compare to the pristine film. When the graphite doped PFO films are fabricated into the AC-driven devices, this morphological difference induces a noticeable change in the EL spectrum shown in Fig. 1d. The non-doped device has a sharp blue peak at 438 nm but a broadened 470 nm peak that extends to 550–600 nm that could induce the undesirable green emission during device operation. However, GNPs doped device has 5 times higher EL intensity at the 438 nm peak compared to the non-doped device and well resolved vibronic peaks at 439 nm, 465 nm, 491 nm. There is a small shoulder at 534 nm when driven under

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