

Influence of poly (3,4-ethylenedioxythiophene)-poly (styrenesulfonate) in polyfluorene-based light-emitting diodes: Evidence of charge trapping at the organic interface

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ABSTRACT

Although the improvement of the indium tin oxide (ITO) anode by PEDOT:PSS has often been reported in literature, the origin of it is not well known. In this paper, we investigate polyfluorene-based devices with a relatively high work function (Al) cathode by electroabsorption (EA) spectroscopy and thermally stimulated current (TSC) techniques. A 0.9 V increase of built-in voltage evidenced by EA in PFV-based devices was similar to that obtained previously in PFO-based devices. It was inconsistent with the work function difference between ITO (4.9 eV) and PEDOT:PSS (5.2 eV). The formation of new types of traps related to the presence of the PEDOT:PSS layer was detected by TSC in PF-N-Ph based devices, confirming that the PEDOT:PSS/PF contact layer acts as an electron trapping surface. We conclude that the TSC technique should allow a deeper characterization of charge traps created at the electrodes and a better modelling of charge injection in future studies.

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1. Introduction

Organic semiconductors and the related electronic components have attracted an increasing interest both from academic researchers and industry during last decades [1]. The main advantages of organic technology compared with the inorganic one are the use of low-temperature, large surface and potentially low cost printing techniques, the possibility to tailor-make the electronic properties of materials in an almost infinite number of ways by chemical engineering, and the possible use of flexible substrates. The commercialization of electronic components has long been hindered by stability issues. Today, however, organic light-emitting diodes (OLEDs) are used in small displays (telephones, mp3 players...) and have been integrated in TV

screens since 2007. Despite these achievements, the comprehension of fundamental processes governing device operation is still incomplete. At the same time, a total control of various parameters, particularly those concerning interfaces, has not yet been achieved.

Poly (3,4-ethylenedioxythiophene)-poly (styrenesulfonate) (PEDOT:PSS) is a conducting polymer that is widely used as anode buffer layer in polymer-based OLEDs (or PLEDs). Indeed, unlike devices based on low-molecular weight materials, the use of multilayer structures for PLED optimization is limited by problems of redissolution of the existing layer(s): the optimization of injecting contacts is thus crucial. Although the improvement of the indium tin oxide (ITO) anode by PEDOT:PSS has often been reported in literature, the origin of it is not well known. Smoothing of the anode surface by this material, as well as improvement of hole injection due to higher work function are current explanations [2]. Electron trapping at the PEDOT:PSS/emitter interface has been suggested in poly (9,9-dioctyl-

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fluorene) (PFO) based devices, relying on the evolution of electromodulation spectrum with the applied dc voltage. Screening of the applied electric field was observed in the presence of a low work function (Ba) cathode, which was supposed to be due to strong electron injection and accumulation at the anode interface [3]. In this paper, we investigate poly (9,9-di-(2-ethylhexyl)-9H-fluorene-2,7-vinylene) (PFV) based devices with a relatively high work function (Al) cathode by electroabsorption (EA) spectroscopy to probe the effect of PEDOT:PSS on built-in voltage, and compare the experimental results with those obtained previously on PFO. In a second part, the thermally stimulated current (TSC) technique was used to probe the trapped charge density in poly (9,9-dihexylfluorene-co-N,N-di(9,9-dihexyl-2-fluorenyl)-N-phenylamine) (PF-N-Ph) based devices. First results show that this technique is adequate to characterize electron or hole trap levels that could be created deliberately close to the anode, or the cathode, respectively.

2. Experimental overview

Devices with an active area of 10 mm² were fabricated by successively spin coating an ITO anode with a 40–50 nm thick layer (2000 rpm, 60 s) of PEDOT:PSS (Baytron P VP Al4083, H.C. Starck), followed by a layer of polyfluorene. ITO coated glass substrates (VisionTek Systems Ltd.) were previously cleaned in successive ultrasonic baths of distilled water, ethanol and acetone. After drying in nitrogen atmosphere, they were treated by UV ozone before use. A 100 nm thick layer of poly (9,9-di-(2-ethylhexyl)-9H-fluorene-2,7-vinylene) (PFV, Sigma–Aldrich) or a 70–80 nm thick layer of poly (9,9-dihexylfluorene-co-N,N-di(9,9-dihexyl-2-fluorenyl)-N-phenylamine) (PF-N-Ph) was deposited by spin coating. Their chemical structures are shown in Fig. 1. Finally, a 100 nm aluminium cathode was evaporated under high vacuum conditions (<10⁻⁴ Pa) and the devices were encapsulated with a glass coverlid using a UV-cured epoxy resin.

Electromodulation spectroscopy was proposed [4] to probe internal electric field in organic components due to the inadequacy of conventional techniques for thicknesses lower than 5 μm [5]. This consists in applying to the device a combined ac modulation and dc operating voltage and measuring changes in the transmission (*T*) of a probe beam (150 W xenon lamp, Oriel) through the active layer(s)

using phase sensitive lock-in detection (SR830, Stanford Research). The light beam entered the sample through the glass substrate with a 45° angle of incidence, and was detected after double transmission and reflection on the back aluminium electrode. Provided that the origin of the electromodulation signal is Stark effect (it is then called electroabsorption), theory predicts that the fractional change in transmission $-\Delta T/T$ is proportional to the third order dc Kerr nonlinear susceptibility and the square of the electric field. One can deduce that the differential transmission is then modulated at both the 1st and 2nd harmonic frequencies in accordance with the following equations:

$$-\frac{\Delta T}{T}(1\omega) \propto \text{Im}\chi^3(\lambda) E_{dc} E_{ac} \cos(\omega t) \quad \text{1st harmonic}$$

$$-\frac{\Delta T}{T}(2\omega) \propto \text{Im}\chi^3(\lambda) E_{ac}^2 \cos(2\omega t) \quad \text{2nd harmonic}$$

Under conditions of low carrier injection, the bulk electric field E_{dc} is related to the dc component of the applied voltage V_{dc} by $E_{dc} = (V_{dc} - V_{Bi})/d$, where V_{Bi} is the built-in voltage and d is the thickness of the layer. V_{Bi} is thus the applied voltage at which the first harmonic (1ω) EA spectrum is cancelled.

To examine the change induced by the presence of interface states, which may be the origin of electron trapping at the anode side, TSC experiments were used to determine the traps in devices [6–8]. The measurements were carried out using a cryostat whose temperature was controlled by an Oxford ITC 503 unit. A Keithley 230 programmable voltage source coupled with a Keithley 617 electrometer was used for monitoring the TSC spectra.

Two similar types of structures have been fabricated which differ by the presence of PEDOT:PSS layer. ITO/PEDOT:PSS/PFV/Al and ITO/PFV/Al structures have been investigated by EA spectroscopy. ITO/PEDOT:PSS/PF-N-Ph/Al and ITO/PF-N-Ph/Al structures have been investigated by TSC technique.

3. Results and discussion

Fig. 2 shows the first harmonic electromodulation spectrum of an ITO/PFV/Al device for different applied dc voltages ranging from –3 to 1 V. The signal is composed of two broad peaks, centred at approximately 385 and 465 nm, respectively, and with opposite signs. The increase of the

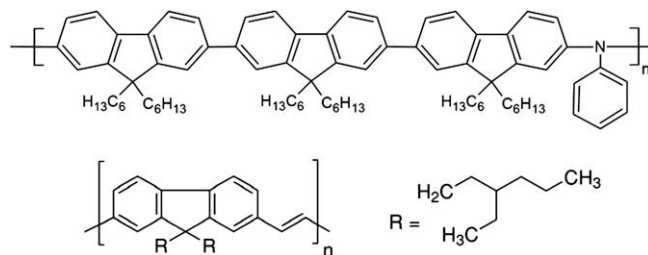


Fig. 1. Chemical structures of polyfluorene derivatives used in this study: poly (9,9-dihexylfluorene-co-N,N-di (9,9-dihexyl-2-fluorenyl)-N-phenylamine) and poly (9,9-di-(2-ethylhexyl)-9H-fluorene-2,7-vinylene), respectively.

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