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Light- and bias-induced effects in pentacene-based thin film phototransistors with a photocurable polymer dielectric

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ABSTRACT

In this work, pentacene-based thin film phototransistors were fabricated with a photocurable polymer insulator and their electrical stability was monitored when the devices were exposed to light sources at different wavelengths. The magnitude of the photocurrent induced by illumination was found to be the result of two distinct factors: a direct photocurrent, related to electron–hole pair generation, and a current enhancement caused by a threshold voltage shift. The direction of threshold translation is attributed to the nature of trap states, specifically those located in the pentacene film near the interface with the polymer, and is affected by a measurement-induced effect, so that the photosensitivity can be modulated by a persistent gate bias during illumination. The equations for these two contributions were developed to study the light effects on material structure, the trapping process of electrons at the insulator–semiconductor interface and the photoconductive efficiency in the organic semiconductor.

1. Introduction

Organic thin film transistors (OTFTs) have gained considerable importance in recent years for cost-effective, large area and flexible electronic device production, encouraging the development of new applications [1–4]. Researchers have investigated different methods to enhance transistor performance; some examples are the improvement of the semiconductor–insulator interface [5,6], the introduction of additional organic layers [7,8], the use of efficient contact materials [9,10] and the fabrication process optimization [11,12]. However, the theoretical interpretation of their electronic and optical properties is not always straightforward. The large variety of organic semiconductors and their particular sensitivity to various elements and composites make OTFTs a good candidate for low-cost and selective chemical and physical sensors [13,14]. Conversely, for applications when enduring stability is required, it is fundamental to reduce the suffering due to the exposure to external factors, such as humidity, electric field and light, which could induce an unwanted chemical degradation and a reduction of the device performance [15].

Since OTFTs can be integrated in optoelectronic applications, as part of the driving circuitry of displays, or used as photo-detectors or light-activated memory devices, it is important to understand the effect of illumination on their electrical performance [16–18]. Recently, organic phototransistors (OPTs), where the control of channel conductance can be enabled by both the magnitude of gate voltage and the absorption of light, have attracted considerable attention thanks to the good response time and the higher responsivity with respect to organic photodiodes [19]. The combination of the organic semiconductor and the gate dielectric plays a key role to determine the performance of OPTs. In particular, it is essential to employ organic semiconductors with both high mobility and excellent light sensitivity and to exploit polymer dielectrics that yield low gate leakage currents and the proper surface for the semiconductor growing.

Pentacene, which is one of the most studied p-type organic semiconductor, has found application in OPTs thanks to its high electrical performance and absorption properties with a great generation efficiency in a wide wavelength region [20–22]. A pentacene-based transistor subjected to light excitation exhibits a behavior depending on the photon energy. Under white or low energy ultraviolet (UV) light, an enhancement of the saturation current, a positive shift of the threshold voltage and a reduction of the total trap density with increasing illumination intensity were reported [23]. In particular, in TFTs with pentacene deposited on

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polymer dielectrics, permanent effects were observed after a low energy UV treatment, which was therefore proposed as a simple way of enhancing the general performance of transistors and controlling their threshold voltage [24]. The photosensing properties of a pentacene OPT are mainly affected by the dielectric surface, which impacts the interface trap density [25], and it was seen that a thinner dielectric layer with a smooth surface is essential for an efficient transport in the channel and enhances photosensitivity [26]. Higher performance under illumination was also observed by reducing the thickness of the pentacene active layer [27] or by introducing a fullerene buffer layer under the electrodes [28].

The present study is devoted to the investigation of photocurrent in OTFTs fabricated with pentacene deposited on a photocurable polymer. The comparison between the electrical characterizations in dark and the results obtained under light conditions has yielded the development of equations useful to describe the photocurrent evolution and to distinguish the effects of photogenerated electrons and holes. Upon light absorption, a high concentration of free charges is induced by exciton generation. In particular, photoinduced holes flow to electrodes, giving a direct contribution to the photocurrent. Conversely, electrons escaped from exciton recombination near the insulator interface, due to the lower mobility, become trapped inducing a positive shift in the threshold voltage. Thus, thanks to the large concentration of both charge carriers generated by light, the measurements performed under photoirradiation allow the identification of trap states affecting charge transport. At the same time, it is possible to evaluate the stability of the electrical performance of the organic transistors under steady-state illumination and to assess the applicability of such devices as photodetectors. However, the measurements under illumination depend on the testing conditions. In our case, a measurement-induced effect has been found as a result of the prolonged application of a gate bias under photoirradiation.

2. Materials and methods

Our organic thin film transistors were fabricated in the top-contact bottom-gate configuration, starting from a glass substrate. A transparent gate electrode of ITO (Indium–Tin-Oxide) was deposited with a thickness of 125 nm. A photocurable polymer by Polyera Corporation (ActivInk D0150) was used as gate insulator. The polymeric solution was spin-coated to afford a film thickness of about 600 nm. The film was cured under ultraviolet irradiation at 300 nm wavelength to activate a cycloaddition process (Fig. 1). The degree of bulk crosslinking was monitored by the optical absorption spectroscopy of the corresponding film on a quartz substrate, in particular through the evaluation of the progressive absorbance decrease of a peak at 270 nm, assigned to the cynnamoyl fragment [29]. The film was irradiated for 30 min and then baked at 120 °C for 2 min. Subsequently, thermal evaporation of a 50 nm thick pentacene active layer was conducted with a base pressure of $2 \cdot 10^{-7}$ mbar. The deposition rate of the organic molecule was maintained at 0.5 Å/s while the substrate was kept at room temperature. After semiconductor film deposition, the transistors were completed by evaporating a 50 nm thick layer of gold through a shadow mask to realize source and drain electrodes.

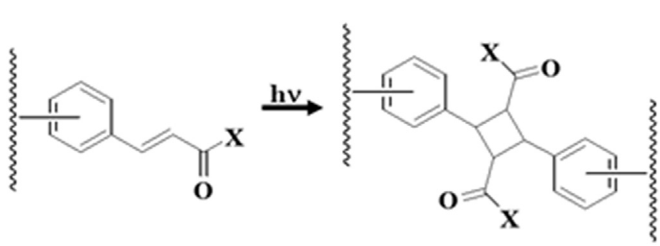

Fig. 1. Schematic of the cycloaddition reaction occurring in ActivInk D0150 film via photocrosslinking.

AFM images and height profiles of the polymer surface (Fig. 2) were recorded, showing that the resulting films are very smooth with a root-mean-square roughness less than 0.3 nm. Several samples of ITO/D0150/Au capacitors were fabricated in order to evaluate the leakage current density as a function of the electric field. The *J*–*E* curves (Fig. 3) reveal very low leakage current densities across the insulator, with values smaller than 10^{-8} A/cm^2 at 3 MV/cm in most cases. Thanks to the capacitors, the dielectric constant of polymer was determined to be 3.3.

Transistors were characterized at room temperature, both in dark and under light conditions, and fundamental parameters were estimated. Many devices were considered; since they showed similar features, typical results are reported. In order to investigate the effects of the incident light on the OTFT performance, the devices were illuminated from the top side, by using LEDs located at a distance of 5 mm from the substrate. Wavelengths covering the visible and ultraviolet range between 285 nm and 630 nm and various irradiances were used. Light with energy lower than HOMO-LUMO gap of pentacene (1.97 eV) did not affect the devices significantly and their results are not reported [19,21,30].

It was seen that both the measuring method and the bias application time affect the performance parameters. In particular, after each measurement, the device seemed to switch on with less negative threshold voltage. This behavior can be attributed to the exposure to ambient air; indeed, by keeping the devices in vacuum for at least 12 h it was possible to partially regain the original features, reasonably due to the release of oxygen, which had been absorbed from the atmosphere enhancing film conductivity by acting as dopant for pentacene [31,32]. After about four months, the repeated bias stress, as well as the inherent degradation process essentially due to moisture absorption, tended to modify permanently the transistor behavior (Fig. 4) [33]. Because of bias stress, also an increased leakage current through the insulator (by a factor of 3) was detected.

The exposure to irradiation, especially to ultraviolet light, altered the electrical characteristics even more incisively. The results were a transconductance decrease and a threshold voltage shift toward larger positive values, so that the saturation regime was no longer observed within the bias range considered during the first electrical characterization, as it is evident in the transfer characteristics of Fig. 4(a). On the contrary, the exposure to a UV light had a positive effect on the insulator; indeed, an irradiation centered at a wavelength (285 nm) close to the one used for the dielectric curing (300 nm) was revealed to restore the pristine values of the gate leakage.

On the time scale of these experiments, it seems that in dark molecular oxygen does not react irreversibly with pentacene and that also a visible light excitation is not sufficient to promote a reaction, so that oxygen, diffused into pentacene at atmospheric pressure, can evacuate rapidly under vacuum, according to experimental results found in literature [34]. In contrast, the exposure of pentacene to air in presence of UV light most probably yields an oxidation by singlet oxygen and/or ozone produced by UV light [24,35].

3. Experimental results

The electrical characteristics collected in the dark and under light conditions are compared. An example is reported in Fig. 5, where the transfer curves of an OTFT working in linear region are