

All-organic bipolar vertical transistor with sulfonated polyaniline base energy barriers favoring recombination emitter-collector current

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ABSTRACT

We report the development and characterization of an all-organic vertical transistor, with tris(8-hydroxyquinoline) aluminum (Alq₃) as emitter, poly(bithiophene) (PBT), as collector material and a sulfonated polyaniline (SPAN) layer as base material in the structure Au/PBT/SPAN/Alq₃/CsO/Al. A thin cesium oxide (CsO) layer provides improved electron injection from the Al into the Alq₃. The transistor is operated in the usual common-emitter mode where the output characteristic evidences the modulation of the collector current through the applied voltage, showing common-emitter gain of 160 at 4.5 V. The incremented gain is due to an increased recombination current, consequence of the arrangement of the frontier orbitals of PBT, SPAN and Alq₃, which favors the confinement of electrons and holes in the SPAN layer.

1. Introduction

The vertical transistor architecture is an old concept [1] receiving increased attention in recent years, in a tentative to avoid the long pathways found in the usual planar field-effect transistors architecture. The vertical architecture is especially attractive for organic semiconductors, which usually show lower charge carrier mobilities than their inorganic counterparts. Shorter transit paths, of the order of film thickness, help achieving the conditions necessary for device operation at lower voltages.

Several attempts were made by different groups to develop organic transistors in vertical architecture [2–15] but, most of the demonstrated devices operate, essentially, with single majority charge carriers transport through all the involved semiconducting layers. However, devices like VOLETs reported by Xu et al. [7] and others [16–19], and devices like the reported by Tavares et al. [13] and Kashura et al. [20], both charge carriers are required to participate in the charge transport along, or at least in parts of the device. In the device reported by Xu et al. [7] electron-hole recombination was achieved, resulting in observable light emission from the organic semiconductor layer, whereas in the device reported by Kashura et al. [16], light emission could be observed only at high voltages, probably in part due to geometrical constraints for light escape, like light blocking metallic contacts covering the full device active area. The common characteristic, however, is that these two devices allow significant bipolar injection in one of the active layers that originate the emitted light.

Along these lines, we recently proposed an all organic transistor [13], composed of two back-to-back organic Schottky-like junctions with a PEDOT:PSS layer playing the role of the metal layer, like a semiconductor/metal/semiconductor (SMS) metal base transistor (MBT) [21]. However, differently from the metal base transistor, whose structure is based on the bipolar transistor, the device proposed by us has an n-type semiconductor/metal junction as emitter-base junction and a metal/p-type semiconductor junction as base-collector junction arranged in an n-type semiconductor/metal/p-type semiconductor structure. Because of this arrangement, when the device is polarized in the common-emitter mode, both junctions are directly polarized, and both, emitter and collector, depending on the voltage applied to the base (V_{BE}), inject majority charge carriers of opposite signs into the base, where recombination is expected to happen even for $V_{BE} = 0$.

This device is potentially very attractive because the stacked emitter, base and collector layers are all organic and compatible with flexible substrates. The all-organic character also reduces the number of high temperature process steps required for metal evaporation. Additionally, the large diversity of organic semiconductors and physical characteristics also offer plenty of possibilities of adjusting energy levels, injection rates, transport characteristics and recombination rates, which are important factors determining operational characteristics in this type of device.

However, despite the possibility of operation at low voltage presented by this all-organic device [13], the low common-emitter current gain of ~ 7 is a drawback. The recombination in this device occurs at

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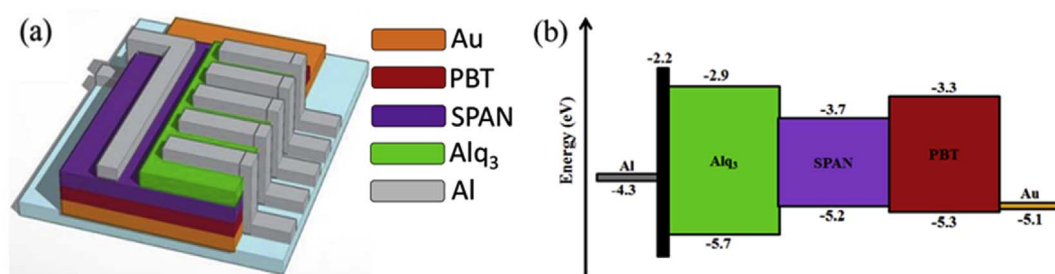


Fig. 1. (a) Device structure and (b) approximate energy levels diagram. In Alq₃, SPAN and PBT, upper and lower values are LUMO and HOMO (Highest Occupied Molecular Orbital) energies, respectively.

the intermediate base layer, constituted by the poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS). PEDOT:PSS, however, as a binary system, is subjected to phase separation and, for this reason, conductivity is not stable at the high temperatures possibly achieved under operation conditions [22]. The low gain can be partially attributed to the frontier orbitals energetic position of the used materials. The PEDOT:PSS base layer has the highest LUMO (Lowest Unoccupied Molecular Orbital) among the active layers, as a consequence reducing the base transit time and base negative charge concentration. These two points reduce the recombination current, which is the contribution responsible for the gain, as discussed further.

In this contribution, we demonstrate that devices prepared using stacked layers of PBT as collector, SPAN as base and Alq₃ as emitter (see Fig. 1) maintain the all-organic characteristic of the mentioned PEDOT:PSS devices, with the advantage of showing high gain.

2. Experimental

The devices were constructed on glass substrates sequentially cleaned in ultra-pure water, acetone and isopropanol (20 min in each solvent) in an ultrasonic bath. Gold films were evaporated to form the collector contact. For improvement of gold adhesion, the substrates were silanized before gold deposition following a procedure reported in Ref. [23]. The devices, that use electrochemically deposited PBT layers as collector, were prepared, with thicknesses varying from 200 to 800 nm thick, using 0.5 M LiClO₄ + 0.1 M 2,2'-bithiophene in propylene carbonate as electrolyte following a previously reported procedure [24]. The SPAN-base was chemically grown directly on the PBT layer from a SPAN solution that was prepared by mixing two solutions [11]. The first was prepared by dissolution of 1.715 g of metanilic acid in 400 mL of deionized followed by ultrasonication for 10 min, after which 455 μ L of aniline and another 100 mL of deionized water were added; the second solution (2) was prepared by dissolving 2.85 g of ammonium persulfate in 62 mL of deionized water. Both solutions were kept at 10 °C for 1 h and then mixed to form p-type SPAN. The PBT-covered substrates were then placed floating on the surface of the solution in a glass beaker to allow the SPAN film growth at 10 °C. After 169 h, which corresponds to a \sim 175 nm thick SPAN film, the substrates were removed. After that, a 120 nm thick Alq₃ layer was deposited, by evaporation, over the SPAN film at a base pressure of 5×10^{-6} Torr. Finally, a \sim 2 nm thick CsO layer was prepared by evaporation of Cs₂CO₃ [25] followed by a 100 nm thick Al layer evaporation, also at 5×10^{-6} Torr. The device active area, controlled by shadow masks and corresponding to the superposition of all device layers, was \sim 2 mm². Besides the steps of Alq₃ and metallic contacts evaporation, taken under vacuum, all other device preparation steps were taken at ambient conditions. Layers thickness were monitored with a quartz balance and measured with a Bruker DektakXT surface profiler. The transistors electrical characterization was performed using a Keithley 2602 dual source meter. All devices were measured in dark, at room temperature, without encapsulation.

3. Results and discussion

Apart from the metal electrode-organic interfaces, two organic-organic junctions compose the vertical transistor, the emitter-base and the base-collector junctions. In the devices reported here, the emitter-base junction is formed between the Alq₃ and SPAN layers and the base-collector junction is formed between the SPAN and PBT layers. The measurements were performed with the devices polarized in the usual common-emitter mode, where the collector and the base are positively biased with respect to emitter. In Fig. 2 the current-voltage $I(V)$ characteristics of the Alq₃/SPAN emitter-base junction (Fig. 2 (a)) and SPAN/PBT base-collector junction (Fig. 2 (b)) are presented.

The hole mobility in PBT, prepared electrochemically under the same conditions as described here, was reported to be $\mu_h \approx 2.6 \times 10^{-5}$ cm²/V.s [26]. Electron mobility information is not available, but in regioregular poly(3-hexylthiophene-2,5-diyl) (P3HT) exposed to air, the hole mobility is reported to be $\mu_h \approx 1.4 \times 10^{-4}$ cm²/V.s and the electron mobility $\mu_e \approx 1.9 \times 10^{-9}$ cm²/V.s [27]. Despite the large availability of P3HT hole mobility information in the literature [28,29], this is not the case for the electron mobility, for which information is scarce. However, some speculation can be made on studies on charge carrier mobility in P3HT blended with electron transporting materials (ETM) films, which discuss the mobility dependence on the ETM fraction present in P3HT [30–32]. These studies show that as the fraction of the ETM in the blend is decreased the electrons mobility decreases and, although no direct measurement of electron mobility at the limit of pure P3HT has been made, by extrapolating the data one can expect mobilities lower than 10^{-8} cm²/V.s, which is consistent with Ref. [22]. Yang et al. [33] suggest that due to the shorter charge carrier recombination time, compared with the transit time of the electrons, the electron transport in the P3HT may be prevented, resulting a very low mobility. Carrier mobility in polythiophenes was shown to decrease with increasing substituted alkyl groups chain length, introduced to increase their solubility [34]. However, for electrochemically grown substituted polythiophenes, it has been observed that the hole mobility does not change strongly with the alkyl chain group length, varying from 1×10^{-4} to 5×10^{-4} cm²/V.s only [35]. Electrochemically grown P3HT [36] has a mobility of $\sim 1 \times 10^{-4}$ cm²/V.s, which is similar to the measured for P3HT deposited by spin coating. Considering the mentioned above and that both PBT and P3HT have the same backbone, even knowing that side chains influence molecular order and packing, it is reasonable to expect at least a similar trend, i. e., electron mobility much lower than hole mobility also in the electrochemically prepared PBT.

The mobility trend in Alq₃ is the opposite, meaning that the electron mobility is higher than the hole mobility. The electric field strength dependent mobility of electrons is larger than the hole mobility by approximately two orders of magnitude over a significant electric field strength range [37].

SPAN is a self-doped conducting polymer showing minority charge carrier mobility $\mu_e \approx 10^{-6}$ - 10^{-7} cm²/V.s [38] and hole field-effect mobility $\mu_h \approx 10^{-2}$ cm²/V.s [39]. The used materials show a

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