

## Ambipolar organic thin film transistors prepared with a one step solution technique



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### ABSTRACT

One of the most challenging problems limiting the development of the technology of organic electronics is a lack of simple, solution based, techniques for producing organic thin film transistors (OTFTs) with ambipolar characteristics. In this work, a simple method of manufacturing ambipolar OTFTs, based on a zone-casting technique, is presented. This technique was used to prepare thin and highly oriented films from a combination of the *p*-type organic semiconductor, TIPS-Pentacene, with one of the two naphthalene bisimide derivatives, both exhibiting electron conductivity. The OTFTs produced with such active layers display ambipolar properties with symmetric characteristics and electron and hole mobility in the best case amounting to:  $\mu_e = 1.5 \times 10^{-2}$  and  $\mu_h = 1.1 \times 10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ , respectively.

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## 1. Introduction

Organic thin film transistors (OTFTs) based on solution processable semiconductor materials are technologically attractive as potential inexpensive active components of such electronic devices as radio frequency identification tags or flexible active matrix displays. In order to construct a basic electronic building block of electronic devices, for instance a low energy consumption CMOS-like complementary circuit, it is necessary to employ a pair of transistors, one exhibiting hole (*p*-channel) and the other electron (*n*-channel) conductivity, both characterized by similar parameters. It is, therefore, necessary to use two types of semiconductors, usually requiring different processing procedures and/or conditions. For this very reason, one of the most challenging goals in the field of printable organic electronics is a development of semiconducting materials characterized by an ambipolar behavior, materials that can be used in a single step assembly to produce a pair of complementary transistors.

Several different methods of preparation of ambipolar transistors have been developed for the last two decades. In one of the first reports in this area, Dodabalapur et al. presented in 1996 an ambipolar transistor based on a heterostructure made of  $\alpha$ -hexathienylene co-sublimated with fullerene C60 [1]. Recently, a pentacene-C60 based ambipolar OTFT with a dual channel was constructed using vacuum deposition techniques [2]. In this device, thin pentacene film was deposited onto dielectric layer with the help of molecular beam deposition, while fullerene layer was placed onto pentacene film by a sublimation technique. Charge-carrier mobilities exhibited by this OTFT amounted to  $2.8 \times 10^{-1} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  and  $1.8 \times 10^{-1} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  for holes and electrons, respectively [2]. As far as one-batch solution approach is concerned, Meijer et al. reported a construction in which the active layer had been prepared by spin-coating of a blend of [6,6]-phenyl C61-butyric acid methyl ester and poly[2-methoxy-5-(3,7-dimethylthioxyloxy)]-*p*-phenylene vinylene [3]. The transistors with such an active layer exhibited charge carrier mobility of  $7 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  for holes and that of  $3 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  for electrons [3]. The highest magnitude of charge carrier mobility for thin film hybrid organic-inorganic transistor, stable in air, was accomplished by Smith et al. [4] The devices prepared by the authors were based on high mobility *p*-type organic semiconductor layer (composed of

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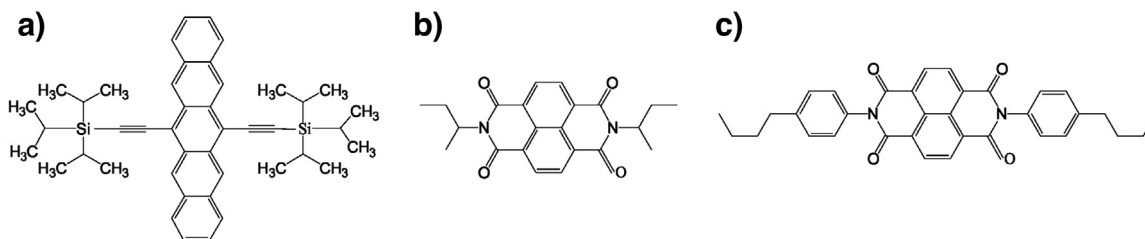


Fig. 1. Chemical formulas of the organic semiconductors: TIPS-Pentacene (a), NBI-s-Bu (b) and NBI-4-*n*-BuPh (c).

a polymer-small molecule blend) deposited on *n*-type metal oxide semiconductor layer and they were equipped with a dual gate system. Charge carrier mobilities, exhibited by these OTFTs, were approaching  $2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ .

In the present work, ambipolar thin film transistors prepared from low molecular weight organic semiconductors with a single step solution procedure and characterized with high charge-carrier mobilities are described. The active layers of these devices are based on solution processed blends of 6,13-bis(triisopropylsilylethynyl)pentacene (TIPS-Pentacene) either with *N,N'*-bis(*sec*-butyl)-1,4,5,8-naphthalenetetracarboxylic-1,4:5,8-bisimide (NBI-s-Bu) or with *N,N'*-bis(4-*n*-butylphenyl)-1,4,5,8-naphthalenetetracarboxylic-1,4:5,8-bisimide (NBI-4-*n*-BuPh). Structural formulas of all three semiconductors used in the work are presented in Fig. 1 below.

One of the crucial factors determining the OTFT performance is morphology of the organic semiconductor layer. It has been known for some time, that with an increase of both short- and long-range order, the charge carrier mobility also increases [5]. In order to assure effective transport and high mobility of both holes and electrons in such blends, each of the two blended semiconductors have to form continuous, crystalline paths connecting source and drain electrodes. For that purpose, the zone casting technique was selected for deposition of thin films composed of the above blends. In this technique, presented schematically in Fig. 2, an appropriate choice of process operational parameters results in a formation of a highly ordered layer of an organic material [6,7]. This method consists in supplying, through a flat nozzle, the solution onto a continuously moving substrate. Concentration gradient, formed in the meniscus due to evaporation of the solvent, constitutes a driving force of unidirectional crystallization of the solute. Zone casting parameters of an essential importance comprise: the concentration and temperature of the solution, solution dosing rate, meniscus height and the advancement velocity as well as temperature of the substrate [8].

It has been demonstrated that this method allows one to fabricate oriented anisotropic layers suitable for high performance organic field effect transistors with both *p*-type [9–12] and *n*-type [13] channels. Bi-functional semiconductor/dielectric composite materials, useful in transistor technology, were also obtained with a help of this technique [14]. In the present work, the zone-casting technique has been adopted for a one-batch, single step production of ambipolar semiconducting layers.

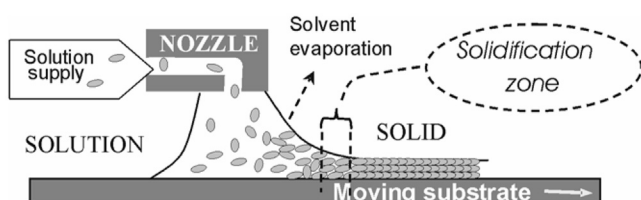


Fig. 2. Schematic view of the zone-casting technique.

Apart from the method of deposition of the ambipolar layer of blends of organic semiconductors with desired morphology, another crucial step in a construction of the OTFT device is an appropriate selection of the dielectric material. As far as the system of gate dielectric and gate electrode is concerned, a combination of silicon dioxide and silicon is usually used in the case of either inorganic or hybrid organic/inorganic transistors [4]. In organic devices, poly(methyl methacrylate) (PMMA) combined with metallic electrodes serve the same purpose [15,16]. However, there is a class of dielectric materials becoming increasingly useful in the field of organic electronics, and namely vacuum deposited/polymerized films of a poly(*para*-xylylene) family, known under their trade name of Parylenes [17,18]. A schematic representation of a deposition process of a chlorine substituted derivative of poly(*p*-xylylene) (Parylene C) is given below in Fig. 3, together with the accompanying chemical reactions.

Compared to PMMA and other conventional plastics, xylylene polymers are characterized by an extremely high purity. This purity, resulting from the polymerization process itself wherein no trace of external initiator is used, is a reason of a relatively low density of localized states present on an interface between dielectric and semiconducting layers of a transistor [18]. In the present work, a combination of vacuum deposited/polymerized Parylene C with thermally evaporated silver was selected as an optimum solution for gate dielectric and gate electrode, and it was done on a very basis of the authors' long and successful experience with these materials [13,19–21]. Apart from providing a clear semiconductor/dielectric interface, the bilayer system of silver/Parylene C acts as an effective protection coating for the organic semiconductors, what is particularly desired in the case of *n*-type organic semiconductors [13].

## 2. Experimental details

### 2.1. Materials

6,13-Bis(triisopropylsilylethynyl)pentacene (TIPS-Pentacene) was purchased from Sigma Aldrich ( $\geq 99\%$  (HPLC)); both naphthalene bisimide derivatives, namely *N,N'*-bis(*sec*-butyl)-1,4,5,8-naphthalenetetracarboxylic-1,4:5,8-bisimide (NBI-s-Bu) and *N,N'*-bis(4-*n*-butylphenyl)-1,4,5,8-naphthalenetetracarboxylic-1,4:5,8-bisimide (NBI-4-*n*-BuPh) were synthesized according to the procedure described elsewhere [22].

### 2.2. Transistors

Thin film transistors were made in a top gate bottom contact (TGBC) configuration presented in Fig. 4. Semiconducting blends: TIPS-Pentacene/NBI-s-Bu of a weight ratio 1:1 (blend 1) and TIPS-Pentacene/NBI-4-*n*-BuPh of a weight ratio 3:4 (blend 2) were deposited by the zone-casting technique onto precleaned glass substrates with previously evaporated gold source and drain electrodes (50 nm thick). For the purpose of evaporation of the source and drain electrodes, a commercial shadow mask was used,

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