

Research paper

Patterned water dispersible conducting polymer electrode in organic thin film transistor through a parylene lift-off process



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ABSTRACT

Availability of non-destructive micro-patterning techniques is essential for the realization of organic electronic technologies that are compatible with chemically sensitive materials. Organic thin film transistors (OTFTs) with conductive polymer composite polyaniline:polystyrene sulphonic acid (PANI-PSS) electrodes that are fabricated by a simple modified Parylene lift-off process is reported. The functionality of the conducting and semi-conducting material is not affected by this technique and it is fully amenable for patterning on a flexible substrate. This patterning technique for conducting polymer electrode, without exposure to harmful solvents, results in a resolution of 5 μm . Water dispersible nanostructured conducting PANi-PSS composite for patterning was synthesized by bulk polymerization of aniline templated with polystyrene sulphonic acid. For the formation of stable dispersion of PANi-PSS with desired conductivity, concentration of the precursor acid used in the synthesis was optimized. The conductivity of films thus obtained from a stable dispersion was increased by more than two orders of magnitude by secondary doping with dichloroacetic acid. Elucidation of the mechanism by which this conductivity improvement occurs was done using Ultraviolet Visible spectroscopy and X-ray diffraction analysis. Morphology characterization of the synthesized nanoparticles and thin films in OTFT was carried out using Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM). Electrical characteristics of bottom contact pentacene TFTs with PANi-PSS electrodes and pentacene active material is superior to those with palladium electrodes due to a lower charge injection barrier. Contact resistance was extracted in both the cases by Transfer Line Method (TLM).

1. Introduction

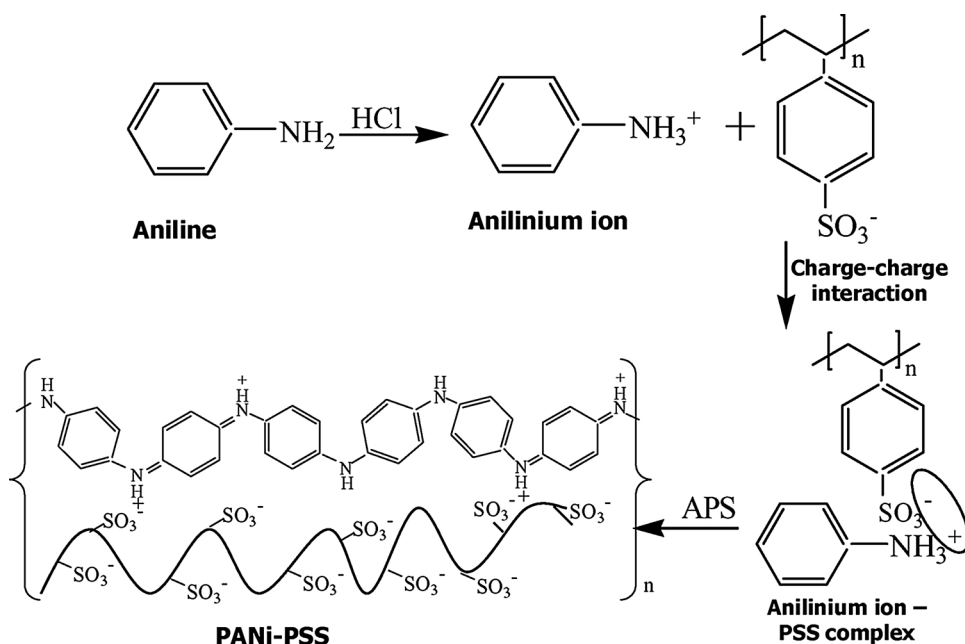
In recent years, a growing interest has been directed to the development and fabrication of large area flexible sensors [1–5]. As flexibility and large-area processing are mandatory requirements for this application, organic electronics can definitely play a role in this field, allowing the fabrication of devices on large, not necessarily planar surfaces at relatively low costs [6,7]. In order to exploit the potential of organic field-effect transistors, the process technology has to be optimized for low-cost techniques [8,9].

Organic electronic device performance is significantly limited by poor charge injection and therefore, a small enhancement in injection would lead to a considerable improvement of OTFT performance. Materials for source/drain electrodes in OTFT have a suitable work function to reduce the charge-carrier injection barrier and form an ohmic contact with organic layers. When semi-conducting layer consists of a p-type material, the work function of source/drain electrode material match or are close to highest occupied molecular orbital (HOMO)

level of the semiconductor [10]. Hence, metals with a high work function are preferred for the source/drain electrodes, such as gold, platinum and palladium. Gold and platinum, which form good ohmic contacts and usually used in OTFTs, are not economical to be used for commercial applications. Palladium is a cheaper electrode material, but has been reported to have relatively high contact resistance due to non-ohmic nature of contacts [11,12]. Substitution of metal electrodes with a conductive polymer, of suitable work function [13,14], is a significant step towards providing lower-energy barriers for hole carrier injection with the added advantage of reducing material cost and for fabrication on flexible substrates. Promising materials for flexible electrodes to match with organic active layers are poly (3, 4-ethylenedioxythiophene), polypyrrole or polyaniline [15,16]. These materials have relatively high electrical conductivity, flexibility, ease of synthesis, and environmental stability. An effective and scalable patterning technique is critical for the low-cost fabrication of organic electronic devices with conductive polymer electrode. Previously investigated techniques, such as screenprinting [17,18], microcontact printing [19,20] and ink-jet

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Scheme 1. Reaction for the synthesis of PANi-PSS composite.

printing [21,22] either have low resolution or involve further process steps, which may degrade the functionality of polymer materials.

One of the difficulties in patterning conducting polymers is that the conducting polymer itself is not easy to process with conventional microfabrication techniques, such as photolithography and etching. Conducting polymer films are easily diluted in the water-based developer used in lithography and can be broken down in solvents that remove photoresist [23]. Parylene lift-off process has been used in recent years for patterning electrodes, dielectrics, proteins etc. for several applications [24–26]. A novel, modified Parylene lift-off process is reported here to pattern conducting polyaniline (PANi) as electrode in organic TFT. Most of the PANi synthesis methods result in powders, which are difficult to process and solubilize. Obtaining a polymer soluble in water and/or in polar solvents is strongly related to the dopant employed in the synthesis process. In the present study, nanostructured PANi, templated with polystyrene sulphonic acid (PANi-PSS) has been synthesized and its conductivity enhanced by more than two orders of magnitude by secondary doping with DCA. This material is readily processed from solution and hence makes excellent alternative to costly metals and metal oxide conductors in organic electronics. Parylene lift-off was employed to pattern this processable conducting polymer as electrodes in pentacene TFT. Performance of the fabricated device was compared with pentacene-based OTFT with non-ohmic palladium electrode, which is a relatively low cost metal with matching work function for p-type pentacene. The contact resistance of the fabricated devices were extracted by Transfer Line Method.

2. Experimental details

2.1. Materials

Aniline (99.9%), Ammonium peroxydisulfate (APS) (> 98%), Poly (sodium 4-styrenesulfonate) (PSS-Na), Mw ~70,000, Poly-(4-vinyl phenol) (PVP), Mw 20,000, Poly (melamine-co-formaldehyde) methylated (84 wt% in 1-butanol), Propylene glycol methyl ether acetate (99.5%), Pentacene (99%), Dichloroacetic acid (DCA) (99%) and Hydrochloric acid (HCl) were purchased from Sigma-Aldrich Co. Ltd. and used without further purification. Aniline was distilled under reduced pressure and stored at 0 °C. APS was dried over molecular sieves and under vacuum overnight. Biaxially oriented, poly (ethylene naphthalate) (PEN) film, of thickness 50 μm (Goodfellow, UK) was used after

cleaning with acetone, isopropyl alcohol and deionized water. Parylene C dimer was purchased from SCS Inc., USA.

2.2. Conducting polymer synthesis

PANi-PSS composite was obtained by oxidative polymerization of aniline in aqueous PSS medium using APS as initiator. For the synthesis of PSS doped PANi in the ratio 1:1, 0.03 mol (based on $-\text{C}_6\text{H}_4\text{NH}$ -repeat unit) aniline was added to 60 mL, 1 M HCl. Separately, 0.03 mol PSS-Na (based on acidic repeat unit) was dissolved in 40 mL, 1 M HCl. 0.01 mol APS was added to acidified PSS-Na solution and stirred for 1 h. PSS-Na was turned to PSS by sodium-hydrogen ion exchange due to solution acidity. The solution was brown in colour. APS-PSS solution was added dropwise to aniline solution and stirred for 24 h at 0 °C. The solution turned green upon stirring. The doped PANi was washed with deionized water and precipitate separated by a centrifuge. The synthesized sample was designated PSN4. PSS doped PANi samples with composition PSS: PANi, 2:1, 1.5:1, 1.25:1, 0.75:1, 0.25:1, 0.1:1, 0.05:1 were synthesized and designated as PSN1, PSN2, PSN3, PSN5, PSN6, PSN7 and PSN8, respectively.

In the oxidative reaction to produce PANi or PANi-PSS composites, acids assist in the polymerization. HCl was chosen as the precursor acid in all syntheses. Since PSN3 gave the highest conductivity, the ratio of PSS: PANi, 1.25:1 was selected for studying the effect of concentration of precursor acid, HCl, on the stability of the synthesized dispersion and to obtain optimum conductivity for the thin film in the fabrication of organic transistor. The concentration of the precursor acid, HCl was varied from 0.1 mM to 2.0 M in the synthesis of PANi-PSS. The polymers with the PSS: PANi ratio, 1.25:1 were designated as PSN3-1, PSN3-2 (same as PSN3), PSN3-3, PSN3-4, PSN3-5, PSN3-6 and PSN3-7, synthesized with the precursor acid concentration, 2 M, 1 M, 0.5 M, 0.1 M, 0.01 M, 0.001 M and 0.1 mM, respectively. The scheme of synthesis of PANi-PSS composite in the presence of precursor acid HCl is depicted in Scheme 1.

After the synthesis and purification of PANi-PSS, to obtain thin films, its 3 wt% aqueous dispersions were prepared. PSN3-3 was chosen for film formation and device fabrication as PSN3-3 gave a stable dispersion with optimum conductivity (discussed in Section 3.4.1). PSN3-3 films were spin casted on glass slide and Si/SiO₂ wafer for further characterization. Secondary doping with DCA was done by soaking the film in preheated DCA at 95 °C for five minutes. The samples were

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