



Low driving voltage characteristics of polyaniline–silica nanocomposites as hole-injection material of organic electroluminescent devices

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ABSTRACT

Polyaniline–fumed silica (PANI–SiO₂) nanocomposites were synthesized through in-situ chemical oxidative polymerization of the aniline in the presence of SiO₂ nanoparticles by using ammonium peroxydisulphate (APS) as an oxidant. The resulting PANI–SiO₂ nanocomposites with different SiO₂ contents were characterized using FT-IR, XRD, FE-SEM and UV–vis techniques. The influence of SiO₂ contents on the sheet resistance of the nanocomposite was investigated by four-point probe system. The optical properties of poly [2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylenevinylene] (MEH-PPV) in toluene solution studied by UV–vis and photoluminescence (PL) measurements at room temperature. For the first time, the application of the prepared PANI–SiO₂ nanocomposites as a hole-injection layer (HIL) of electroluminescent (EL) devices was examined and the device performance (e.g., turn-on voltage and current density–voltage (*J*–*V*) characteristics) was investigated. The Fowler–Nordheim (FN) field-emission tunneling model was used to explain the *J*–*V* characteristics of the fabricated devices. The hole-injection barrier height (φ) values for the devices were obtained and reported.

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1. Introduction

In recent years there has been increasing interest in developing of organic–inorganic hybrid materials. These materials often have a desired combination of the different properties of its components and play a major role in the development of advanced functional nano- and bulk materials. The improved and unusual features of the hybrid materials open promising applications in many areas such as photovoltaic, photonics, optoelectronics, photocatalysts, sensors, fuel cells, smart membranes and separation devices [1,2]. Among the base materials used to make the hybrid organic–inorganic materials, polyaniline (PANI) is one of the most extensively studied conducting polymers due to its flexibility, good electrical conductivity, low cost, oxidative stability and controlled facile synthesis [3–5]. Considering their great potential applications such as batteries [6], sensors [7,8], electronic devices [9], supercapacitors [10] and anticorrosion coatings [11–13], the modified PANI films have been prepared by dopant ions used in

the polymer synthesis process. The electrical conductivity which is a key property in many applications of PANI composites can be enhanced by incorporating nano-metal oxides into the polymer matrix [14–16]. These hybrid systems are expected to display synergistic properties between PANI and the nano-metal oxides, making them potential candidates for applications in electronic devices [17–19].

In recent decades, several reports have been published related with the enhancement of stability, durability, electrical properties and optical properties of conjugated polymer based nanocomposites for improving the performance of polymer light emitting diodes (PLEDs). The effect of nanoparticles anatase titanium oxide (TiO₂) dispersed within MEH-PPV as photoactive material was studied by Ruhstaller et al. [20]. Hussain et al. improved the electrical properties and luminous efficiency of MEH-PPV based PLEDs by incorporation of Au-capped TiO₂ to the light-emitting MEH-PPV polymer layer [21]. Recently, the role of poly(3,4-ethylenedioxythiophene):poly(*p*-styrenesulfonate) (PEDOT:PSS)–TiO₂ nanocomposites as a HIL on the current–voltage characteristics of PLEDs was reported by Dinh et al. [22]. Among the nanofillers used in the polymer-based nanocomposite preparation, nanosized SiO₂ is one of the most common fillers due to its some unique optoelectronic activities and transport properties [23–25].

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In this work, we have prepared PANI–SiO₂ nanocomposites using in-situ chemical oxidative polymerization. The prepared nanocomposites with different SiO₂ contents have been used to fabricate EL devices with structure of PET (poly(ethylene terephthalate))/ITO (indium tin oxide)/PANI–SiO₂/MEH-PPV/aluminum (Al) at room conditions. The effect of PANI–SiO₂ films as a HIL with different SiO₂ weight ratios on output parameters of devices has been investigated. For the fabricated devices, the φ values were estimated using the FN field-emission tunneling theory.

2. Experimental

2.1. Synthesis of PANI and PANI–SiO₂ nanocomposites

PANI–SiO₂ nanocomposites were synthesized through in-situ chemical oxidative polymerization process by the oxidation of aniline in the presence of hydrochloric acid (HCl) and APS. At the first, an appropriate amount of SiO₂ nanoparticles (AEROSIL[®] 200 with average primary particle size of 12 nm, Degussa AG, Germany) were suspended and sonicated in 50 ml of 1 M HCl for 30 min to get well-dispersed nanoparticles. Then distilled aniline monomer (2.63 mmol) was added to a glass reactor containing the well-dispersed SiO₂ nanoparticles. The mixture was stirred with magnetic stirrer in ice water bath for 2 h to get a uniform suspension. Then 50 ml of 1 M APS solution was added to the mixture drop by drop for 30 min. The clear and colorless solution mixture at the beginning changed to a light blue color and finally to a dark green color after 4 h, indicating that PANI–SiO₂ nanocomposite was produced. Finally, the products were washed with deionized water until the filtrate became colorless, and then dried at 70 °C for further characterization. The schematic representation of the synthesis of PANI–SiO₂ nanocomposites is shown in Fig. 1. In this work, PANI–SiO₂ nanocomposites with the different filler contents were synthesized using 0, 10, 15 and 20 wt% of SiO₂ with respect to aniline monomer and the weight ratios of SiO₂ were checked by burning the weighted PANI–SiO₂ nanocomposite samples at 500 °C for 5 h in a furnace and weighing the amount of white residual powders.

2.2. Fabrication of EL devices

The devices with the configuration ITO/HIL/MEH-PPV/Al were fabricated. The ITO-coated on flexible PET, with a sheet resistance of about 60 Ω/square purchased from Sigma–Aldrich Company used as the substrate for fabricating the devices. The PET/ITO substrates were cleaned using ultrasonic baths of deionized water, acetone, methanol and isopropanol and then spin-coated by a dispersed solution of the PANI–SiO₂ nanocomposites in *N,N*-dimethylformamide, followed by drying in oven at 80 °C for 1 h. The dried PANI–SiO₂ films were spin-coated by MEH-PPV from toluene solution and subsequently followed by deposition of an Al metal layer using thermal evaporation with a shadow mask at a

Table 1
Diodes fabricated with different HIL.

Devices	HIL	φ (eV)
A	PANI	0.132
B	PANI-10% SiO ₂	0.122
C	PANI-15% SiO ₂	0.140
D	PANI-20% SiO ₂	0.203

base pressure of 5×10^{-5} mbar. The fabricated devices are summarized in Table 1.

Fourier transform infrared (FT-IR) of the prepared PANI and PANI–SiO₂ nanocomposites were recorded on spectrometer (Magna 550, Nicolet) in the range of 400–4000 cm⁻¹. The X-ray diffraction (XRD) pattern of the samples were obtained on a Philips X'Pert Pro diffractometer using 35 mA and 40 kV current with a Ni-filtered Cu K α radiation ($\lambda = 0.154056$ nm) over the 2θ range of 0–80°. Morphological study of nanocomposites was carried out using field-emission scanning electron microscopy (FE-SEM) on a Hitachi S-4160 (Japan). The optical absorption spectra of sample solutions were obtained using PerkinElmer Lambda 2S spectrophotometer. The sheet resistance of the prepared PANI–SiO₂ nanocomposites was measured by a four-point probe system (Keithley 196 & 224) at room temperature. Current–voltage characteristic of the fabricated devices was measured by a computer-controlled Keithley 238 Source Measure Unit. All the measurements were performed at room temperature in air without any device encapsulation.

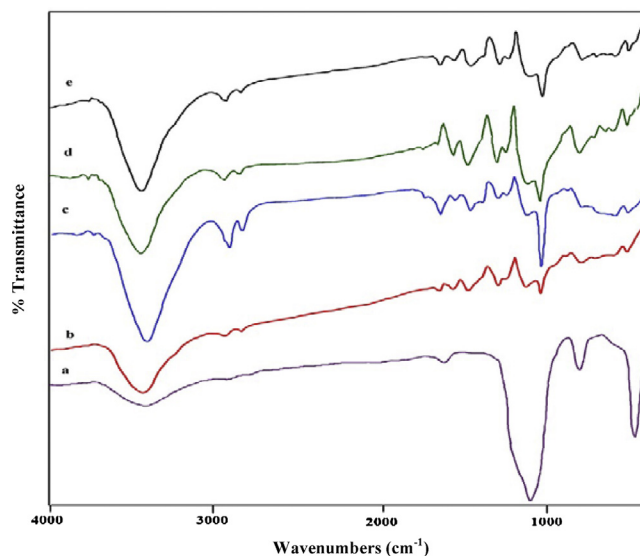


Fig. 2. FT-IR pattern of (a) SiO₂ nanoparticle, (b) PANI and (c)–(e) PANI-10% SiO₂, PANI-15% SiO₂ and PANI-20% SiO₂ nanocomposites.

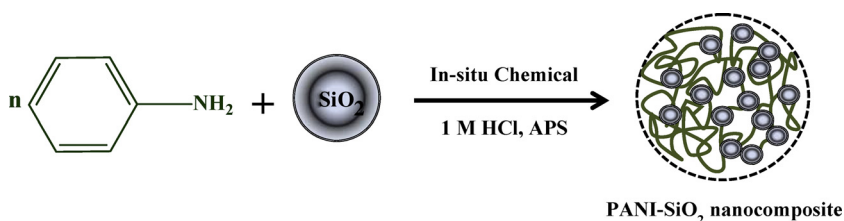


Fig. 1. Schematic representation of the synthesis of PANI–SiO₂ nanocomposites.

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