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Letter

Flexible light-emitting electrochemical cells with single-walled carbon nanotube anodes

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ABSTRACT

In this work, we demonstrate flexible solution processed light emitting electrochemical cells (LECs) which use single-walled carbon nanotubes (SWCNTs) films as the substrate. The SWCNTs were synthesized by an integrated aerosol method and dry-transferred on the plastic substrates at room temperature. The addition of a screen printed poly (3,4-ethylene dioxythiophene) doped with poly (styrene sulfonate) (PEDOT:PSS) film onto the nanostructured electrode further homogenizes the surface and enlarges the work function, enhancing the hole injection into the active layer. By using an efficient phosphorescent ionic transition metal complex (iTMC) as the active material, efficacies up to 9 cd/A have been obtained. These values are among the highest reported so far for light-emitting diodes employing CNTs as transparent electrode.

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1. Introduction

In the last two decades, organic light-emitting diodes (OLEDs) have been the focus of extensive research which led to their entry in the display market. Despite the successful development of this technology, OLEDs have not yet been implemented at their full potential. The use of OLEDs in general lighting applications, for example, is not yet feasible. Efficient OLEDs rely on sequentially evaporated multilayer stack, which increase considerably the overall device cost [1]. For this reason, much simpler devices like light-emitting electrochemical cells (LECs) are actively being investigated. LECs consist of a single layer of an ionic organic semiconductor, either a polymer blended with salts or a pure ionic transition metal complex (iTMC), sandwiched between two electrodes. Thanks to the presence of freely moving ions, the active layer can withstand alone all processes taking place in an electroluminescent device, i.e. charge injection, transport, exciton formation and recombination, which are normally absolved by the different functional layers in OLEDs. Hence, LECs represent a competing technology for inexpensive future light sources. An additional feature of OLEDs and LECs is the possibility of being processed through simple solution deposition techniques at low temperature, making them compatible with a variety of substrates, including flexible ones. Unfortunately, the most widespread transparent conductor nowadays is indium tin oxide (ITO), and its application to large area flexible devices is limited by its insufficient conductivity. Furthermore, ITO is polycrystalline, and suffers from cracking after repeated bending or strain which further increases the surface resistivity. Within these premises, several alternative thin film transparent conductors have been explored, in particular metal nanowires [2,3], graphene [4–6] and, most notably, carbon nanotubes (CNTs) [7–13]. In terms of performances and cost reduction, single walled carbon nanotubes (SWCNTs) networks have demonstrated to be competitive with ITO as well as with most of organic materials that have been extensively studied as low-cost alternatives [14]. Furthermore, high flexibility of the SWCNTs opens avenues beyond ITO, i.e. creation of completely new components, needed in the flexible and transparent electronics [15]. In this work,

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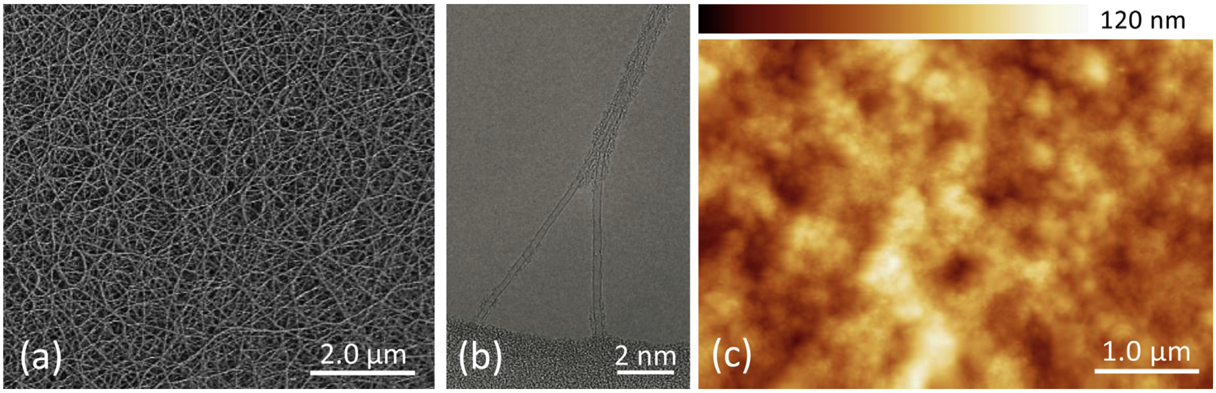

Fig. 1. (a) SEM and (b) TEM images of bare SWCNTs film at low and high magnification. (c) AFM topography of the same after screen printing of a 100 nm thick PEDOT:PSS layer.

we demonstrate flexible solution processed LECs employing SWCNTs foils as the substrate. An integrated aerosol chemical vapor deposition (CVD) synthesis of SWCNTs and their consequent dry deposition at room temperature was used [16,17]. The SWCNTs produced by ferrocene decomposition in a CO atmosphere are usually first collected from the outlet flow on filters and then can be transferred from the filter to practically any substrate, including heat sensitive materials. Hence, the SWCNTs synthesized by the aerosol method meet all the requirements for low-cost, flexible and transparent electronics, which could be used in many high technology applications like touch sensors and displays [18–20]. The addition of a screen printed poly (3,4-ethylene dioxythiophene) doped with poly (styrene sulfonate) (PEDOT:PSS) film onto the nanostructured electrode further homogenizes the surface, which is needed for the preparation of high quality diodes with low leakage current. Moreover, PEDOT:PSS enlarges the work function, enhancing the hole injection into the active layer. By using an efficient phosphorescent iTMC as the active material, efficacies up to 9 cd/A have been obtained. These values are among the highest reported for light-emitting diodes employing CNTs as transparent electrode [21].

2. Experimental details

2.1. Preparation and characterization of the flexible conducting substrates

SWCNTs were synthesized by an aerosol (floating catalyst) CVD method based on ferrocene vapour decomposition in a CO atmosphere described elsewhere [22,23]. Briefly, the catalyst precursor was vaporized by passing ambient temperature CO through a cartridge filled with ferrocene powder. The flow containing ferrocene vapour was then introduced into the high-temperature zone. In order to obtain stable growth of SWCNTs, a controlled amount of carbon dioxide was added together with the carbon source [24]. SWCNTs were collected downstream of the reactor by filtering the flow through nitrocellulose or silver membrane filters. 20–80 nm thick SWCNTs films on PET or glass substrates were formed by a dry press transfer technique [16]. Subsequently, a 100 nm thick layer of PEDOT:PSS was deposited by screen printing onto the SWCNTs film. SWCNTs film morphology was investigated using a scanning electron microscope Helios Nanolab 660 (SEM, FEI) with a 30 kV accelerating voltage. Transmission electron microscopy (TEM) images were obtained with a Tecnai G2 F20 microscope with point resolution up to 0.24 nm and line resolution up to 0.144 nm at 80 kV. The surface morphology of the thin films was analysed using atomic force microscopy (AFM, Multimode SPM IVa, Veeco, USA). The work function of the SWCNTs and PEDOT:PSS-coated SWCNTs substrates were measured with an Air Photoemission System (APS02, KP Technology Ltd., UK).

2.2. Device fabrication and characterization

The iTMC used as the light-emitting material is the [Ir(ppy)$_2$(dtb-bpy)](PF$_6$), where ppy is 2-phenylpyridinato and dtb-bpy is 4,4′-di-(*tert*-butyl)-2,2′-bipyridine), synthesized according to previously described methods [25]. The emitting layers (100 nm thick) were prepared by spin-coating an acetonitrile solution of the iTMC (4 wt%) onto the PET/SWCNTs-PEDOT:PSS foils. After deposition of the emitting layer, the samples were transferred into a nitrogen glovebox, where the aluminum electrodes (100 nm) were thermally evaporated in a vacuum chamber through a shadow mask. The active area of the devices was 24 mm^2. The devices time dependent parameters were obtained by applying a pulsed current while simultaneously monitoring the luminance with a True Colour Sensor MAZeT (MTCSICT Sensor), using a Lifetime Test System designed by BoTEST (Botest OLT OLED Lifetime-Test System). Electroluminescence spectra were recorded with an Avantes fiber-optics photo-spectrometer. The devices were not encapsulated and were characterized inside the glovebox.

3. Results and discussion

The SWCNTs morphology on the PET substrate has been investigated by SEM and TEM, as depicted in Fig. 1(a–b). The large area SEM image (Fig. 1a) demonstrates high quality SWCNTs connected between each other randomly. The high magnification TEM (Fig. 1b) demonstrates that the deposited nanotubes are single-walled. The SWCNTs layer was subsequently coated with PEDOT:PSS, in order to smooth the surface and minimize the possibility of isolated CNT protruding perpendicularly from the substrate. This is necessary to avoid direct contact with the counter electrode, considering that the active layers of organic devices such as LECs do not exceed 100 nm in thickness. The resulting surface was analyzed by AFM, as reported in Fig. 1(c).

The morphology of the PET/SWCNTs/PEDOT:PSS surface appears smooth, at least compared to the bare SWCNT layer as seen by SEM (Fig. 1a), indicating full coverage of the underlying carbon nanostructure by the conducting polymer layer. The corresponding root mean square roughness R_{rms} is 15.8 nm, with a height distribution centered at about 50 nm. Besides the relatively high R_{rms} (for comparison, ITO has an R_{rms} typically < 3 nm [26]), a more significant quantification of the surface quality in the case of OLEDs is the peak-to-valley roughness R_{pv}, which directly correlates with the

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