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Non-oxidized graphene nanoplatelets as an efficient hole transport layer in organic light-emitting diodes

Hoang-Tuan Vu [a], Hsin-Chieh Yu [a], Ying-Chih Chen [a], I-Wen Peter Chen [b], Chun-Yuan Huang [b], Fuh-Shyang Juang [c], Yan-Kuin Su [a,d,*]

[a] Institute of Microelectronics and Advanced Optoelectronic Technology Center, National Cheng Kung University, Tainan 701, Taiwan
[b] Department of Applied Science, National Taitung University, Taitung 950, Taiwan
[c] Institute of Electro-Optical and Materials Science, National Formosa University, Huwei, Yunlin 632, Taiwan
[d] Department of Electrical Engineering, Kun Shan University, Tainan 710, Taiwan

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ABSTRACT

In this work, we have synthesized and applied non-oxidized graphene nanoplatelets (GNPs) with high work function of 5.61 eV into the aluminum tris(8-hydroxyquinoline) (Alq_3)-based organic light-emitting diodes (OLEDs) as an efficient hole transport layer (HTL). By the assistance of Py^+ ions from the pyridinium tribromide salt ($Py^+Br_3^-$), the exfoliation of GNPs can be easily obtained via sonication. After proper filtering, the concentration of GNPs dispersed in water–ethanol mixture is about 0.2 mg/mL. To be the HTL, we presumed that a nearly full coverage of GNPs on ITO substrates can be achieved by multiple spin-coat process. Thereby, low turn-on voltage of 4.1 V (@100 mA/cm^2) and high luminance of 36,000 cd/m^2 (at 8.4 V) were obtained, which corresponded a luminous efficiency (2.33 cd/A) about 200 times and 1.4 times higher than that in OLEDs without any HTL and with a N,N-bis-(1-naphyl)-N,N-diphenyl-1,1′-biphenyl-4,4″-diamine (NPB) HTL, respectively.

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1. Introduction

Graphene and its derivatives have extensively motivated and attracted research worldwide in last few years. The fascinating electrical and mechanical properties such as high transparency, mobility, conductivity and flexibility of graphene and its derivatives make then suitable for potential optoelectronic applications as electrodes or semiconducting layers [1–5]. For example, the strong demand of alternatives to replace vulnerable indium tin oxide (ITO) have driven the extensive investigation of synthesis and process techniques of graphene toward applications including inorganic photovoltaic cells (OPVs), organic thin film transistors (OTFTs), organic memory devices, and organic light-emitting diodes (OLEDs) [6–11].

Indeed, most of the present studies have been focused on utilizing graphene as the anode in OLEDs [6,12]. In those accomplishments, in order to obtain highly efficient graphene electrode, graphene surfaces were treated with week O_2 plasma [13] or modified by nitric acid doping-HNO_3 [6] which facilitated the conductivity and the injection of hole by increasing the graphene's work function. However, these methods still have drawbacks due to the increasing of surface roughness by O_2 plasma treatment or the instability of nitric acid doping. Some studies even tried to obtain high performance by using hybrid anodes of graphene and conducting polymer [14] or graphene/Ag/Al-doped zinc oxide [15] which more or less decreased the graphene's unique advantage, high transparency.

Besides, graphene oxide (GO) and reduced graphene oxide (rGO) have been studied in OLEDs [16,17]. Though

* Corresponding author at: Institute of Microelectronics and Advanced Optoelectronic Technology Center, National Cheng Kung University, Tainan 701, Taiwan. Tel.: +886 6 275 7575x62382.
E-mail address: yksu@mail.ncku.edu.tw (Y.-K. Su).

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the GO is an inherent insulator due to the disruption of sp^2 conjugation in the graphene lattice, the residual sp^2 clusters in GO is considered as reason that can allow hole or electron transport to occur at the Fermi level by hopping in the same sub-lattice [18]. As a result, GO and rGO have widely used as a efficient HIL/HTL in OLEDs. In Lee's report, a polymer light-emitting diodes (PLEDs) with a 4.3 nm solution-processable GO interlayer have efficiency 2.2 times higher than that in devices with a conventional hole transport polymer, (poly (3,4-ethylenedioxythiophene)–polystyrenesulfonic acid) (PEDOT–PSS). On the other hand, Shi et al. reported that an undoped solution-processable GO layer used in fluorescent OLEDs is capable to achieve high luminance of 40,785 cd/m^2 at only 8.8 V [17].

In this paper, we have successfully synthesized the dispersion of high work function non-oxidized graphene nanoplatelets (GNPs) in water–ethanol mixture by the assistance of pyridinium salt ($Py^+Br_3^-$). Raman spectra shows the GNPs are with low structure defects. By precisely controlling the coverage of GNPs on ITO/glass substrates for hole transport, high luminance of 36,000 cd/m^2 and luminous efficiency (LE) of 2.33 cd/A were demonstrated in aluminum tris(8-hydroxyquinoline) (Alq_3)-based OLEDs.

2. Experimental

2.1. Preparation of graphene suspension

The preparation of GNPs solution was conducted by a facile synthesis route depicted as following: Firstly, 0.5 mg-weight highly oriented pyrolytic graphite (HOPG) purchased from SPI supplies was mixed with pyridinium tribromide salt ($Py^+Br_3^-$) in the water:ethanol mixture (1:1). The Py^+ cations were adsorbed on the exposed surface of graphite by π–π interactions. With proper sonication, the repulsion force between Py^+ cations on adjacent graphene layers competes to original van der Waals forces, leading to the gap opening and further exfoliation of GNPs. Finally, the charge transfer complexes of graphene and Py^+ cation were dispersed in the solution. Since no chemical oxidation or reduction is involved during the exfoliation, defect-associated deterioration in electronic properties of graphene is supposed to be ruled out.

2.2. OLEDs fabrication

The OLEDs were fabricated on pre-patterned ITO/galss substrates which were purchased with a sheet resistance of ~13 Ω/□. The substrates were cleaned via acetone, isopropyl alcohol, and de-ionized water, followed by an UV-ozone surface treatment for 25 min. Graphene suspension was spin-coated on substrates through a syringe filter (0.45 μm) at 500 rpm for 10 s and 3000 rpm for 30 s. In this study, various coverages of the GNPs films were investigated and they were realized by repeating the same spin-coating process for multiple times. After that, non-oxidized GNPs on ITO were baked at 120 °C for 1 h in the air ambient to dry out the solvent. All organic and metal layers were subsequently deposited by thermal deposition at pressure of 1×10^{-6} Torr. The active area of the device was 1×1.5 mm^2, defined by the shadow masks exchanged in a glove box directly connected with the evaporator. The structure of all fabricated devices in this study is shown in Fig. 1(a), which can be shortly described as ITO/(GNPs or NPB)/Alq_3 (50 nm)/LiF (0.5 nm)/Al (200 nm). The NPB, which was used as HTL in conventional devices, has thickness of 40 nm after optimization.

2.3. Characterization and analysis

The electrical characteristics of devices were measured using a Keithley 2400 source-meter, a Keithley 2000 multimeter, a silicon photo-detector and a PR650 colorimeter in a nitrogen-filled glove box with oxygen and water vapor less than 1 ppm. The transmittance of GNP films and the NPB film were measured by UV–Vis spectrometer (Perkin–Elmer, Lambda 35). Raman spectra of GNP films and HOPG were measured by a micro-Raman spectrometer (Bruker Senterra).The surface roughness of the GNPs was measured by atomic force microscopy (AFM). The thickness of GNPs films and the grain sizes of GNPs were estimated by transmission electron microscopy (TEM). The work function of GNPs film and HOMO level of NPB film were obtained by a photo-electron spectrometer measured in air (Model AC-2) and an ultraviolet photoemission spectroscopy (UPS).

3. Results and discussion

In this study, the energy level diagram of each layer is exhibited in Fig. 1(b), accordingly, the fluorescent material Alq_3 played a role as electron transport layer and emitting layer with electroluminescence at 520 nm (inset of Fig. 1(c)). The curves of square root of the photoelectron emission intensity for GNPs and NPB films as a function of irradiated photon energy are shown in Fig. 1(c). From the curves, the work function of GNPs was derived as 5.61 eV, which is much higher than the previous reported values (4.4 or 4.6 eV for pristine graphene) [6,13]. The NPB layer experienced a HOMO level of 5.45 eV which is near its previous value reported in [19]. Moreover, we also checked the work function an of GNPs film on ITO/glass substrate by the ultraviolet photoemission spectroscopy (UPS) measurement. From UPS spectra, the estimated work function of GNPs film was 5.5 eV and similar with the result obtained by AC-2. The energy level diagram intuitively shows that the higher work function of the GNPs HTL induces almost no hole barrier between GNPs and Alq_3 interface ($\Delta E = -0.01$ eV). In our work, graphene was synthesized by solution process to obtain GNPs in water: ethanol (1:1) mixture. Therefore, the conductivity of graphene film strongly depends on the overlaps among GNPs after spin-coating process. In fact, GNPs obtained a lot of edge disorders which directly affects the physics of electrons in graphene and its transport properties due to the disruption in sp^2 conjugation at the edge of flakes lattices. However, the residual sp^2 clusters remained in GNPs lattices still aids the transportation of charge carriers. Hence, GNPs film could be utilized as HTL of OLEDs in this study. As a

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