



# High-performance polymer light emitting diodes with interface-engineered graphene anodes

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## ABSTRACT

Recently, graphene-based organic light emitting diodes (OLEDs) were successfully demonstrated using graphene as anodes. However, the graphene electrodes have not been utilized for polymer light emitting diodes (PLEDs) yet, although the simpler device structure and the solution-based fabrication process of PLEDs are expected to be more advantageous in terms of time and cost. Here we demonstrate high-performance polymer light emitting diodes (PLEDs) with simple two-layer structures using interface-engineered single-layer graphene films as anodes. The single-layer graphene synthesized by chemical vapor deposition methods was transferred onto a glass substrate utilizing an elastic stamp, and its work function was engineered by varying the duration and the power of ultraviolet ozone (UVO) treatment. Thus, we were able to optimize the contact between silver electrodes and the graphene anodes, leading to the considerable enhancement of light-emitting performance.

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## 1. Introduction

Since the first isolation of graphene in 2004 [1], graphene-based transparent electrodes have received remarkable attentions in the photonic and optoelectronic applications [2] including organic light emitting diodes (OLEDs) [3–6], photovoltaic cells [7], light emitting electrochemical cells (LECs) [8,9], and touch sensors [10]. Although indium tin oxide (ITO) is being mainly used as transparent electrode materials in various display and photovoltaic applications, its brittleness, high fabrication cost, and poor surface property have limited the actual performance of optoelectronic

devices [11]. Moreover, the diffusion of the indium into the organic layer causes OLED luminance degradation due to luminescence quenching [12]. On the contrary, the graphene electrode is expected to show not only high conductivity and outstanding transmittance but also ultrahigh flexibility, chemical inertness, and smooth interface with controllable work functions, which made it one of the most promising candidates that can replace ITO in the future [10]. In practice, several papers have already reported the use of graphene electrodes for OLED applications [3–6]. However, most of them show rather low current and power efficiencies unless additional hole injection materials or complicated multi-layered structures are adopted. On the other hand, the use of graphene electrodes for polymer light emitting diodes (PLEDs) with simple bi-layer structures has not been reported yet, although PLEDs are expected to be more advantageous than OLEDs in terms of processibility, flexibility, fabrication and materials costs [13].

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In most cases, multi-layer graphene electrodes are being used rather than single-layer graphene to achieve higher conductivity [3–6]. However, the sheet resistance of graphene does not critically limit the optoelectronic performance in the case of bottom emission type active-matrix organic light emitting diodes (AMOLED) because the transparent electrodes are used only in a patterned manner just for individual pixels if the pixel can be emitted uniformly. Therefore, in this case, appropriate methods of patterning and work function engineering to achieve lower contact resistance between graphene electrodes and metal lines are more crucial than conductivity itself.

Thus, we demonstrate, for the first time, the fabrication of high-performance PLEDs using chemically modified single-layer graphene electrode as an anode to maximize the optoelectronic performance. Ultraviolet ozone (UVO) treatment was utilized to pattern and engineer the work function of the single-layer graphene films. The contact resistance between single-layer graphene films and several metal layers were also investigated.

## 2. Experiment and measurements

### 2.1. Preparation of graphene substrates

Monolayer graphene was synthesized by utilizing a chemical vapor deposition (CVD) process described in the literature [10]. The graphene film grown on the copper foil was covered by poly(methyl methacrylate) (PMMA) and floated on the surface an aqueous solution of 0.1 M ammonium persulphate ( $(\text{NH}_2)_4\text{S}_2\text{O}_8$ ). After all the copper layers were etched away, submerging a clean PET film into the etchant and picking up the floating PMMA/graphene film to transfer it into DI water. The cleaning process was repeated 5 times. The graphene film with PMMA support was transferred to the glass. To measure Raman spectrum, the monolayer graphene film was transferred to the  $\text{SiO}_2/\text{Si}$  substrate. The sample was dried with blowing nitrogen gas immediately, and was baked for 8 h on the hot plate at 60 °C, followed by cleaning in acetone at room temperature for 30 min to remove the PMMA support layer. Finally, the sample was dried and baked as previously done.

### 2.2. Patterning of graphene films and preparation of the TLM measurements

Graphene film on a glass substrate was etched with the shadow mask by illuminating the UVO in the air. The etching time was 15 min. Then, the several metal such as Ag (99.99%, Materion Advanced Chemical, USA), Al (99.999%, CERAC, USA), Ti (99.995%, CERAC, USA) was deposited in the vacuum chamber under  $5.0 \times 10^{-6}$  torr with the deposition rate at 5.0 Å/s, 2.0 Å/s, and 1.0 Å/s, respectively.

### 2.3. Fabrication of PLEDs

For a reference device, the glass substrate with ITO patterns purchased from Free M Tech (Korea) was cleaned in a ultrasonic bath with acetone, isopropyl alcohol (IPA), and deionized water for 10 min each at room temperature,

consecutively. Then, the substrate was dried in an oven for 1 h at 120 °C. For the PLEDs with single-layer graphene anodes, additional Ag electrodes were deposited under  $5.0 \times 10^{-6}$  torr in a vacuum chamber. The rate of deposition and the thickness of the Ag electrode is 5.0 Å/s and 1500 Å, respectively. To control the work function of anodes and to remove any organic contaminants, the substrate was treated by UVO for 5 min for the graphene PLEDs, and for 10 min for the ITO PLEDs. The power of UV lamp (Low Pressure Mercury Vopar Grid Lamp, Jelight) was 28 mW/cm<sup>2</sup>. After the UVO treatment, the substrate was transferred to a globe box to minimize the concentration of H<sub>2</sub>O and O<sub>2</sub> during device fabrication. PEDOT:PSS (Poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate), CLEVIOS™ P VP Al 4083, Heraeus, Germany) was spun-coated at 2000 rpm for 60 s. Then, annealing process was followed at 120 °C for 10 min. SPG-01T (Merck, Germany) that is a green emitting polymer was spun-coated by using 0.8 wt% concentrations in toluene solvent at 2000 rpm for 60 s. The substrate was annealed at 90 °C for 60 min, and then, Ca (99.99%, Sigma-Aldrich) was deposited at 1.0 Å/s rate in a vacuum chamber under  $5.0 \times 10^{-6}$  torr. Without breaking vacuum, Al was deposited at 2.0 Å/s rate under  $5.0 \times 10^{-6}$  torr.

### 2.4. Measurements

The SEM images of device structures and the atomic force microscopy (AFM) images of graphene films were taken by a field emission scanning electron microscope (FE-SEM, Hitachi S-48000) and by a non-contact mode AFM system (XE-100, Park system), respectively. The work function was measured by photoemission yield spectroscopy (AC-2, Riken Keiki). The Raman spectrums were measured by a Raman spectrometer (Raman microsystem 2000, Renishaw). The thickness and the sheet resistance were measured by a surface profiler (Surfcorder ET3000i, Kosaka) and a four point probe measuring system (FPP-5000, Changmin, Korea), respectively. The resistance between adjacent electrodes for a transfer length method (TLM) and the patterning process optimization were measured by a semiconductor parameter analyzer (HP4145B, HP). Current (I) – Voltage (V) – Luminance (L) characteristics were measured by a digital multimeter (Keithley 2000, Keithley) and a source-measure unit (Keithley 236, Keithley) while sweeping voltages. The measured data were calibrated by a spectroradiometer (CS-1000A, Konica Minolta).

## 3. Results and discussion

Fig. 1a shows the fabrication process of our PLEDs. The thickness of PEDOT:PSS and SPG-01T layers are 24 and 70 nm, respectively. The thickness of Ca and Al electrodes are 30 and 150 nm, respectively. Schematic structure of the device with ITO (reference), and single-layer graphene anodes are shown in Fig. 1b and c, respectively. Corresponding cross-sectional SEM images are shown in Fig. 1d and e. Except for anode layer, device structures of both PLEDs are identical. The emission area was  $1.6 \times 1.4 \text{ mm}^2$ .

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