Enhanced performance in organic light-emitting diode by surface modification of ITO with Graphene oxide

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Abstract

We demonstrated that surface modification of ITO by graphene oxide (GO) brings about hole injection property and enhanced device stability in OLED. Photoelectron yield spectra of GO modified ITO suggested that the shit of vacuum level shift, which is responsible for the enhanced performances.

1. Introduction

OLEDs have attracted attention to flexible displays and lighting applications, where low voltage driving and long lifetime are indispensable. We have reported that insertion of MoO_3 reduces charge injection barrier and increase in device stability. [1] Recently, it was reported that insertion of GO as hole injection layer in OLED reduces the hole injection barrier and enhances device performance of OLED. [2] However, evaluation of lifetime characteristics of the OLED with GO has not been reported yet. In this paper, we demonstrated that surface modification of ITO by GO brings about not only enhanced hole injection efficiency but also enhanced device stability in OLED.

2. Experimental method

GO was synthesized by means of oxidation of graphite powder as described in the literature. [3] Chemical composition of GO was characterized using Fourier transform infrared (FT-IR) spectroscopy and Raman scattering spectroscopy. Morphological characterization of GO was done by atomic force microscope (AFM) measurement, scanning electron microscope (SEM) measurement. To evaluate work function of GO, GO on ITO and ITO, photoelectron yield spectroscopy (PYS) was employed. To apply GO layer as the hole injection, GO was deposited on cleaned ITO electrode by air-liquid interface technique. [4] The GO modified ITO substrates were set in a vacuum evaporator to fabricate devices. To investigate of hole injection characteristic, we fabricated the hole-only devices whose structure was glass substrate/ITO (150 nm) /GO/a-NPD (100 nm) /MoO₃ (10 nm) /Al (100 nm). We also fabricated the OLED, where the device structure was glass substrate/ITO (150 nm) /GO/α-NPD (90 nm) /Alg₃ (60 nm) /LiF (0.5 nm) /Al (100 nm). All organic layers and electrodes were deposited by vapor deposition in a vacuum chamber at the pressure of 10⁻⁷ Torr. The devices were encapsulated using a glass cap and an UV curing epoxy resin together with a desiccant sheet, which absorbs both oxygen and water. All fabrication processes were conducted without exposing the samples to air. The device area was 4 mm², which was defined as the overlapped area of the ITO layer and the Al layer.

The current density-voltage (J-V) characteristics of the hole-only devices (HODs) and OLED were measured using a computer-controlled sourcemeter (2400, Keithley). The luminance of the OLEDs was measured using a luminance meter (BM-9, TOPCON) during the measurement of J-V characteristics. The operational stability of the OLEDs were measured at a constant current density at 50 mA/cm².

3. Results and discussion

The morphology of GO layer on ITO electrode was measured by SEM (Figure 1). The ITO electrode was covered with the planer GO sheets where the grey area and light area represent GO sheets and uncovered ITO, respectively. The size of the GO sheets ranged from 1 to 4 μ m. In the separate experiments, AFM image of the GO layer on Si wafer revealed that the thickness of GO sheets was around 1 nm which corresponds to the GO monolayer. [2]



Fig. 1 SEM image of GO layer on ITO electrode. Scale bar is $10 \ \mu m$.

Figure 2 shows the current density-voltage (*J-V*) characteristics of hole-only devices. The current density markedly increased when GO layer was inserted. For instance, the current density of HOD with GO layer measured at 1 V (8.30 mA/cm^2) is 430 times larger than that of the HOD without GO layer (0.02 mA/cm^2).

To investigate origin of the increase in current density, we measured work function of ITO and GO on ITO electrodes with PYS. Figure 3 shows photoelectron yield spectra of ITO and GO on ITO. The work function of UV/O_3 treated ITO electrode was -5.01 eV. On the other hand, the

photoelectron yield spectra of ITO with GO layer exhibits two inflection points at -5.20 and -5.79 eV. To assign each point, we measured thick GO layer on a silver substrate and the work function of GO layer was found to be -5.80 eV. Based on these results, we assign the values of work function of -5.20 eV and -5.79 eV for ITO and GO. We noticed that the work function of ITO shift from -5.01 eV to -5.20 eV by inserting GO layer. This result suggests that the shift of vacuum level of ITO took place by depositing GO. Thus, we concluded that the increase in current density in hole-only device is realized by not only the reduction of



Fig. 2 J-V characteristic of hole-only devices. \circ : With GO layer, \blacktriangle : Without GO layer



Fig. 3 Photoelectron yield spectra of ITO, GO modified ITO and thick GO on Ag.

hole injection barrier but also the shift of vacuum level of ITO.

Figure 4 shows *J*-*V* characteristics of OLEDs. Similar to the *J*-*V* characteristics of hole-only device, we observed reduction of voltage by inserting of GO layer such that the driving voltage at 10 mA/cm² with ITO with GO (6.50 V) is 2.75 V lower than that of the OLED without GO (9.25 V).

Figure 5 shows the voltage-time (Fig. 5a) and relative luminance (L/L_0) -time (Fig. 5b) characteristics of OLEDs driven at the constant current density (50 mA/cm²). In general, increase in driving voltage is frequently observed during the degradation of OLEDs. Indeed, the OLED without GO layer showed drastic increase in the driving voltage. On the other hand, the increase in the driving voltage of OLED with GO layer was suppressed significantly. It has been reported that since the suppression of increase of the driving voltage can be realized by coating of ITO with fluorinated polymer layer [5], where the reason for the increase in driving voltage would be caused by the degradation of ITO surface. Based on this, we suggested that the electronic state of ITO surface was improved stability by insertion of GO layer. Although the initial degradation was observed regardless the GO layer (Fig. 5b), the long-term stability after 100 h of the OLED with GO layer showed better enhanced stability.





Fig. 5 Lifetime measurement of OLEDs.

Conclusions

In summary, we demonstrated that the reduction of voltage due to the shift of vacuum level shift of ITO by insertion of GO layer. In addition, the device stability was increased by inserting GO layer, which improved stability of the electronic state of ITO surface. Although, the initial degradation did not change by the GO layer, the long-term stability can be further improved with optimization of GO modification technique

References

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