Thermal Evaporation Process Based Organic/Ag/ Organic Transparent Conducting Electrode for Flexible Optoelectronic Applications

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ABSTRACT

Herein, we report a new organic/Ag/organic (OAO) multilayer flexible transparent electrode fabricated a thermal evaporation process. This OAO electrode showed superior optical and electrical characteristics of which transmittance of 81.34% at 550 nm wavelength and low sheet resistance of 9.51 Ω/sq. Its flexible reliability is also very excellent as sheet resistance variation at the radius of curvature of 5 mm with bending cycles of 2,000 is negligible. Fabricated green phosphorescent organic light emitting diodes with this OAO electrode showed high current efficiency of 75.1cd/A.

1. INTRODUCTION

Indium-tin-oxide (ITO), currently used as a transparent conducting electrode (TCE), has been widely used in various optoelectronic applications due to its high transparency with low sheet resistance. However, ITO electrode accompanies not only the sputtering process but also a complicated photolith-ography process for patterning, which is disadvantage-ous in the production cost. Also, since ITO has high rigidity, it makes significant difficulty to apply ITO for flexible applications due to a drastic increment of sheet resistance caused by the cracking for bending stress. To surmount the problems, several flexible TCE technologies to replace the ITO electrode have been studied. Among them, dielectric/metal/dielectric (DMD) structure in which ultrathin metal layer such as Ag and Au is sandwiched within the dielectric layers, especially metal oxide, is strong candidate for flexible TCE.[1-2] The DMD multilayer electrode enables direct patterning through a fine metal mask during thermal evaporation, and it also has a merit in that a successive process between electrode and organic light emitting diode (OLED) fabrication is possible. In addition, it anticipates DMD electrode is suitable for flexible optoelectronic devices owing to the high ductility of the metal. However, metal oxides such as MoO_3 and WO_3, which are mainly used as a dielectric layer, have a relatively low transmittance as compared with organic materials and a high deposition temperature which are likely to suffer thermal damage to a plastic substrate. Hence, it is needed to study the electrode structure for improving them. Ultrathin metal based electrodes with thermal evaporation based organic materials have been rarely reported. Thus, we are convinced that our electrode has a high novelty compared with previously reported studies.

In this study, we propose the thermal evaporated TCE with organic/Ag/organic (OAO) multilayer structure. As organic material which is used instead of the metal oxide requires relatively lower deposition temperature in the fabrication of the electrode, there is less concern for damage of plastic-based substrates. In suggested electrode, we obtained the excellent characteristics of high transparency (81.34% at 550 nm) and low sheet resistance (9.51 Ω/sq) with high bending durability.

2. EXPERIMENT

In this study, DMD and OAO structures were fabricated on a 125 μm thick PET substrate for a valid comparison of electrode characteristics. The DMD and OAO electrode is following structure: (PET / MoO_3(40 nm) / Ag(12 nm) / MoO_3(40 nm), (PET / KHU-1 organic(40 nm) / Ag(12 nm) / KHU-2 organic(40 nm)).

These electrodes were used only thermal evaporation technique. PET substrates were cleaned with acetone, and isopropyl alcohol by ultrasonication and rinsed by deionized water followed by ultraviolet-ozone treatment for 600 seconds. All materials were thermally deposited under high vacuum pressure (~1.0 ×10-7 torr). The deposition temperature of MoO_3 was 556 ºC. The thin Ag layer was deposited at a rate of 0.1Å/s.

We used Konica Minolta CS-100A luminance and color meter and Keithley 2635A source meter unit for evaluate current density-voltage-luminance (J-V-L) characteristics of fabricated devices. Efficiency, EL spectra and Commission Internationale de l'Eclairage (CIE) 1931 color coordinator were evaluated using a spectroradiometer (Konica Minolta CS-2000A). The transmittance was evaluated using the Jasco V-700 UV-visible spectrophotometer and the sheet resistance was evaluated by DASOLENG FPP-2000A 4-point probe sheet resistance meter.

To verify bending reliability, mechanical flexibility and durability tests of the two electrodes were carried out 2000 consecutive bending cycles with a bending radius of 5 mm using the bending test system.

3. RESULTS AND DISCUSSION

3.1 Characteristics of Electordes

In spite of the excellent conductivity characteristics of Ag, but it is difficult to obtain high transmittance with low sheet resistance in ultrathin sole Ag layer. It is originated from the non-uniform film by the Ag agglomeration which is caused by the higher cohesion between Ag than the adhesion between Ag and substrate. [3] Therefore, the bottom dielectric layer which increases the adhesion force with silver should be applied under Ag layer. Hence, as a wetting inducer, we introduced KHU-1 organic material which generate the complex well with silver. Since the generated [Ag−KHU-1] complex acts as nuclei of Ag growth, we expect the Ag agglomeration might be prevented.

Meanwhile, we also capped the KHU-2 organic layer on the Ag to enhance the optical transmittance by antireflection effect in accordance with the destructive interference. [2,4] Moreover, KHU-2 layer also has high hole injection characteristics, thus, we believe that the performances of OLED device are also improved.

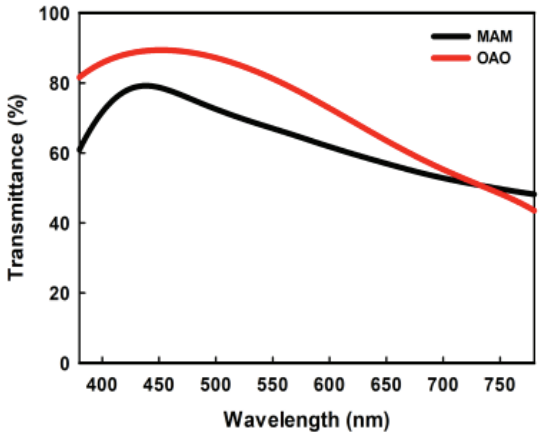

Fig. 1 Transmittance versus wavelength characteristics of OAO and MAM electrode

Based on the above electrode concepts, we demonstrated OAO electrode. To verify the excellence of suggested electrode, the performances of OAO electrode were compared with those of MoO3/Ag /MoO3 (MAM) electrode which is previously reported electrode of DMD structure. As shown in Fig. 1, the transmittance at 550 nm wavelength was improved from 67.18% to 81.34% according to the change of dielectric layers. The sheet resistances of the OAO and MAM structures were 9.51 Ω/sq and 13.11 Ω/sq, respectively. Its low sheet resistance implies that the wettability of Ag is favorable in OAO electrode. As a result, the OAO electrode showed better optical and electrical characteristics than those of MAM electrode.

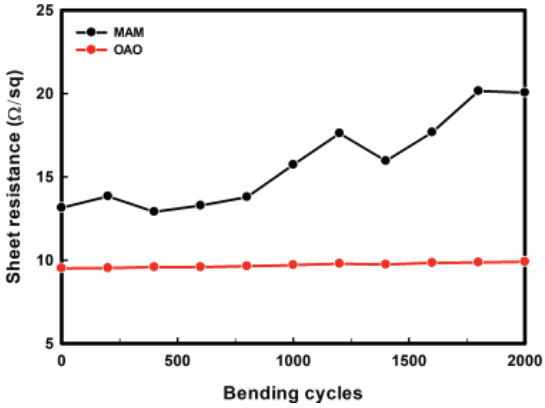

Fig. 2 Sheet resistance change of MAM and OAO electrodes according to the bending cycle and photographs of bending reliability test

To verify the flexibility of suggested electrode, we conducted the bending reliability test for OAO and MAM electrode with a radius of curvature of 5 mm and 2,000 bending cycles. After bending test, the sheet resistance of OAO electrode was only increased up to 9.90 Ω/sq while that of MAM electrode dramatically changed up to 20.58 Ω/sq, as depicted in Fig 2. Consequently, durability for bending stress of OAO electrode was superior to that of MAM electrode, and thus, we anticipate the OAO electrode is appropriate to apply in flexible optoelectronic devices.

3.2 Using of Multilayer Electrode in Green Phosphorescent OLED

We fabricated a green phosphorescent OLED in which OAO and MAM electrode is applied and compared the performances of each device to demonstrate the suggested electrode is also suitable for OLED device.

Considering optical simulation results, we designed OLED with the following configuration. PET / MoO_3(40 nm) / Ag (12 nm) / MoO_3(10 nm) / NPB (15 nm) / TCTA (15 nm) / Bepp2 : 5wt% Ir(ppy)2(acac) (20 nm) / TmPyPB (45 nm) / LiF (1.5 nm) / Al (100 nm). For faithful comparison of device performances, identical OLED device with OAO electrode is also fabricated on PET substrate under the same deposition conditions. Herein, the OLED structure comprises, bis[2-(2-pyridinyl-N)phenyl-C](2,4-pentanedionato-O^2,O^4)iridium(III) ($Ir(ppy)_2(acac)$) and bis(2-(2-hydroxyphenyl)pyridinato) beryllium ($Bepp_2$) as the green phosphorescent dopant and host, respectively, 4,40-bis[N-(1-naphthyl)-N-phenylamino]biphenyl (NPB) and Tris(4-carbazoyl-9-ylphenyl)amine (TCTA) as the hole transport layer (HTL) and the exciton blocking layer (EBL), respectively, 1,3,5-Tri(m-pyridin-3-ylphenyl)benzene (TmPyPB) as the

electron transport layer.

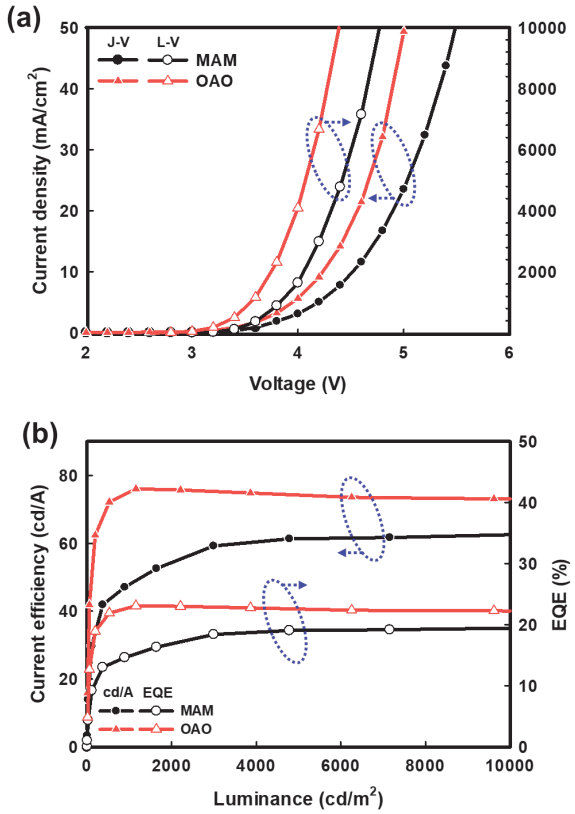

Fig. 3 Device performances of green phosphorescent OLED with different electrodes on flexible substrate (a) J-V-L characteristics, (b) current efficiency and EQE versus luminance properties of OLEDs with OAO and MAM electrodes.

As can be seen in Fig 3(a), the OAO electrode applied device indicated faster driving voltage (3.9 V) than that of MAM electrode (4.2 V) at 3,000 cd/m2 due to the better hole injection property of OAO electrode. Owing to the synergetic effect between improved charge balance via high hole injection characteristics and increased transmittance of electrode, the OAO electrode contained device also presented excellent current efficiency of 75.1 cd/A and external quantum efficiency (EQE) of 23.0% at 3,000 cd/m2 whereas those of MAM electrode device was 62.7 cd/A and 19.5% as portrayed in Fig 3(b). Consequently, it is established that the suggested electrode was also suitable for optoelectronic device.

4. CONCLUSIONS

We obtained thermal evaporation fabrication process based OAO TCE with high transparency (81.34% at 550 nm), low sheet resistance (9.51 Ω/sq) and high bending durability. Because the whole fabrication process of OLED can be carried out at once, we anticipate that the suggested electrode can simplify the fabrication process, thus the manufacturing costs might be lowered. Additionally, its high flexible durability is appropriate for flexible applications. Therefore, we expect that OAO can be one of the good alternatives for ITO electrode.

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