Highly efficient tandem p-i-n organic light-emitting diodes adopting a low temperature evaporated rhenium oxide interconnecting layer

Dong-Seok Leem,1 Jae-Hyun Lee,1 Jang-Joo Kim,1,a) and Jae-Wook Kang2
1Department of Materials Science and Engineering and Center for Organic Light Emitting Diode, Seoul National University, Seoul 151-744, Republic of Korea
2Department of Surface Technology, Korea Institute of Materials and Science (KIMS), Changwon 641-831 Republic of Korea

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High quality interconnection units (ICUs) with a high transparency and superior charge generating capability for tandem organic light-emitting diodes (OLEDs) are developed. The ICUs of rubidium carbonate-doped 4,7-diphenyl-1,10-phenanthroline/rhenium oxide (ReO3)-doped N,N'-diphenyl-N,N'-bis(1,1'-biphenyl)-4,4'-diamine layers with or without an additional ReO3 interlayer produce high transmittance (88%–92% at 420–700 nm) and spontaneous internal charge generation properties. A very high efficiency of ~129 cd/A has been demonstrated from only two stacked green p-i-n OLEDs by employing the developed ICUs. The relationship between the device efficiency and internal charge generation within the ICUs is further described by means of the capacitance measurements. © 2008 American Institute of Physics. [DOI: 10.1063/1.2979706]

Stacked (tandem) organic light-emitting diodes (OLEDs) are attractive for next-generation displays and solid-state lightings owing to their significantly high current efficiency and brightness.1–15 High performance tandem OLEDs require formation of high quality interconnection unit (ICU) possessing high optical transparency as well as superior charge generation and injection properties.1,5 Researchers have reported on several types of ICUs including metal-based multilayers,2,6 undoped organic bilayers,3 organic-doped organic p-n junctions,7–9 and doped organic/metal oxide bilayers.10–14 Among them, the use of metal-based or undoped organic bilayer ICUs (Ref. 3) are feasible for simple fabrication of tandem OLEDs. However, the relatively low transmittance (below 70% in the visible range) of metal-based ICUs (Refs. 2 and 6) and high operation voltages of tandem devices employing undoped organic bilayer ICUs (Ref. 3) remain as problems. On the other hand, metal and organic doped n-p junction ICUs, such as Bphen:Cs/N,N'-Di(naphthalen-1-yl)-N,N'-diphenyl-benzidine: tetrafluorotetracyanoquinodimethane (F4-TCNQ),8 and Alq3:Mg/m-MTDATA:F2-TCNQ,9 have been widely used to fabricate efficient tandem OLEDs due to their good electrical and optical properties. However, recent results have shown that the highly diffusive p-dopant (F2-TCNQ) (Refs. 3 and 15) decreases interfacial stability causing an increase in drive voltage during operation.15 Unlike the organic doping method, the application of relatively stable metal oxides as connection systems such as Alq3: Mg/WO3,10 Alq3: Mg/VO5,11 Alq3: Cs2CO3/MoO3,12 and Bphen:Cs2CO3/NPB:WO3,13 also have been developed. The metal oxides, themselves, can produce good electrical conductivity and a high transmittance of 85%–90% in the visible range,9 but their high evaporation temperatures (over 600 °C) diminish compatibility with organic molecules and hinder their practical application in tandem devices.2,3,9,15

In this letter, we report on highly transparent and superior charge generating organic p-n junction ICUs. The developed ICUs consist of rubidium carbonate (Rb2CO3)-doped 4,7-diphenyl-1,10-phenanthroline (Bphen)/rhenium oxide (ReO3)-doped N,N'-diphenyl-N,N'-bis(1,1'-biphenyl)-4,4'-diamine (NPB) with and without a very thin ReO3 interlayer. The developed ICUs showed a high transmittance of 88%–92% at 420–700 nm wavelengths. Excess absorption by the doping and the ReO3 interlayer was below 4% in the entire visible range. A high efficiency of 129 cd/A was obtained from two stacked phosphorescent green p-i-n OLEDs by employing a developed ICU, which was mainly attributed to the superior internal charge generation as well as the high transparency of the ICU.

The patterned indium tin oxide (ITO) substrates were prepared and basically cleaned.16 Before construction of the two stacked p-i-n OLEDs, the single p-i-n OLED structure was fabricated and optimized. This structure consisted of the ITO anode, 4 wt % ReO3-doped NPB hole transporting layer (HTL) (80 nm), undoped NPB (20 nm), a double emission layer (30 nm) of 8 wt % Ir(ppy)3-doped 4,4'-N,N'-dicarbazolylbiphenyl and 8 wt % Ir(ppy)3-doped Bphen, undoped Bphen (40 nm), 15 wt % Rb2CO3-doped Bphen electron transporting layer (15 nm), and an Al cathode. After that, we fabricated tandem p-i-n OLEDs by simply stacking the optimized single p-i-n cell with or without a ReO3 interlayer between the n-p junction ICUs. The structures of the devices are schematically shown in Fig. 1. We employed ReO3 as the p-dopant and the interlayer because it can be evaporated at a lower temperature (~340 °C) than MoO3 (~620 °C), alleviating the drawbacks of metal oxides in a practical manufacturing process induced by a high evaporation temperature.15 Moreover, the device stability can be enhanced by doping ReO3 in HTL; and the doping capability of ReO3 is superior to other metal oxide dopants.16 The current density-voltage-luminance (J-V-L) characteristics of the devices were measured by a Keithley 2400 semiconductor parameter analyzer and a Photo Research (PR-650) spectrophotometer. We measured the angular-dependent electroluminescence (EL) spectra by an optical fiber and a $2000
miniature fiber optic spectrometer (Ocean Optics). The capacitance-voltage measurements of the ICUs were carried out by a 1260 impedance/gain-phase analyzer and a 1287 electrochemical interface (Solartron). The transmittance was measured by means of a UV-visible spectrophotometer (Cary 5000).

Figure 2(a) shows the J-V-L characteristics of single and tandem OLEDs. The two stacked OLEDs with the doped organic p-n junction ICU (tandem A) exhibits an operation voltage of 9.5 V at 1000 cd/m^2, which is larger than twice that of the operation voltage (3.6 V) of a single OLED. However, the incorporation of a very thin (1 nm) ReO_3 interlayer at the interface between the n-doped and p-doped layer in the ICU (tandem B) significantly reduced the operation voltage to 8.1 V at 1000 cd/m^2, indicating that the insertion of a thin ReO_3 layer effectively enhances the electrical properties of the doped organic p-n junction ICU. In addition to that, the tandem B device shows a very high luminance of 68 000 cd/m^2 at a current density of 242 mA/cm^2 and a bias voltage of 14.8 V (not shown). The current efficiency of single and tandem OLEDs are displayed in Fig. 2(b) as a function of the current density. A very high current efficiency of 129 cd/A is obtained from the tandem B, which is almost twice that of the single OLED (68 cd/A). The tandem A device also exhibits a high current efficiency of 111 cd/A, but a little lower than that of the tandem B.

Figure 3 displays the EL spectra of single and tandem p-i-n OLEDs. The stacked OLEDs (A and B) exhibit almost the same EL spectra as a single device without any spectrum narrowing. The EL spectra of tandem OLEDs also show no angle dependence and the emission intensity follows Lambertian distribution (the inset of Fig. 3). These results suggest that the two types of ICUs are optically transparent, and minimize the microcavity effect. The microcavity effect results in EL spectrum narrowing, angle dependent spectral change, and deviation from the Lambertian distribution of emission intensity. This microcavity effect was commonly observed in tandem devices employing metal-based ICUs where the microcavity is formed by metal-based semireflective ICUs and a highly reflective metal electrode (cathode). In contrast, our devices show little microcavity effect due to the high transmittance (low reflectivity) of the ICUs.

We proved high optical transparency of the ICUs by a transmittance measurement, as shown in Fig. 4. Both ICUs
A change in the absorption is induced by the doping of Rb$_2$CO$_3$ and the reference of Bphen which is much higher than that of 88%–92% at wavelengths of 420–700 nm, from the increase in absorption by the formation of charge transfer complexes between the NPB and ReO$_3$. Little change in the absorption is induced by the doping of Rb$_2$CO$_3$ in Bphen. This transparency is much higher than that of an undoped organic bilayer exhibits no change in capacitance with the applied voltages, indicating no generation of internal charge carriers. The samples with ICUs A and B, however, show higher capacitance than that of the reference even at low and negative (forward for the n-p junction) bias. We attribute this higher capacitance to the larger dielectric constants of the doped layers coming from the larger polarization of the charge transfer complexes. In addition to that, the capacitances apparently increase with the increment of positive (reverse for the n-p junction) bias above certain voltages for the devices. These results suggest that additional (internal) charges are generated within the ICUs since no charges can be injected into the ICUs due to the thick LiF insulating layers. We note that the capacitance of Bphen:Rb$_2$CO$_3$ layer in the ICU B generates more charges at a lower voltage than the ICU A and consequently contribute to the enhancement in efficiency as well as the lowering in an operation voltage of tandem OLED B (Fig. 2). Our quantitative analysis of the charge generation efficiency in the ICUs will be published elsewhere.

In summary, we demonstrated high efficiency green tandem OLEDs using Bphen:Rb$_2$CO$_3$/NPB-ReO$_3$ and Bphen:Rb$_2$CO$_3$/ReO$_3$/NPB-ReO$_3$ as ICUs. Two stacked OLEDs adopting one of the ICUs showed a very high efficiency of ~129 cd/A with little angular dependence of the EL spectrum, resulting from high optical transmittance (88–92% at 420–700 nm) and efficient charge generation of the ICUs. The insertion of a thin ReO$_3$ interlayer within the ICUs was very effective in generating charges as demonstrated by the C-V measurement and in lowering the operation voltage of the stacked OLEDs.

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