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Enhancement of electron injection in inverted top-emitting organic light-emitting diodes using an insulating magnesium oxide buffer layer

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We report the enhancement of the electron injection by inserting a 1-nm-thick magnesium oxide (MgO) buffer layer between Al cathode and tris (8-hydroxyquinoline) aluminum in an inverted top-emitting organic light-emitting diode (OLED). The turn-on voltage of OLEDs decreased from 10 to 6 V and the luminance increased about 61% as the MgO interfacial layer was employed. The MgO interfacial layer played a role in reducing the energy barrier of electron injection, leading to the reduction of the turn-on voltage and the enhancement of luminance. © 2005 American Institute of Physics. DOI: 10.1063/1.2033129

Inverted top-emitting organic light-emitting diodes (ITOLEDs), utilizing cathodes as a bottom contact, are more preferable in active-matrix organic light-emitting devices due to the use of superior n-channel transistors.1 Achieving improved device performance of ITOLEDs requires a high reflective bottom cathode, optimization of charge injection, and transport at the interfaces.

In ITOLEDs, aluminum (Al) has been mainly used as a reflective bottom cathode.2,3 Unfortunately, the comparatively high-work function (~4.3 eV) of Al brings the poor OLEDs performance such as an increase of the turn-on voltage and a decrease in efficiency.4 Therefore, it needs to insert a proper electron injection layer (EIL) between the cathode and the emitting materials for the effective electron injection.

Several kinds of EIL such as poly(methyl methacrylate),5 CsF,5 NsSt,6 and Al2O37 were reported to reduce the turn-on voltage. However, no works were conducted on magnesium oxide (MgO) as an EIL. The magnesium oxide (MgO) has the wide band gap (6.0–7.8 eV)8 and the good insulating property with a high dielectric constant of 9.96.9 Thus, it is expected that the electron injection could be improved by inserting a thin MgO buffer layer.

In this letter, we report the enhancement of the electron injection in ITOLEDs using the thin MgO buffer-layer on Al cathode. The change in the work function with the insertion of MgO was examined using synchrotron radiation photoelectron spectroscopy. From this, the effect of MgO layer on the improvement of device performance is discussed.

The glass was used as the starting substrate. The surface of glass was cleaned in sequence with the acetone, isopropyl alcohol, and de-ionized water, and then dried with high purity nitrogen gas. In order to enhance the adhesion of Al film on glass, Ti (20 nm) was used as a glue layer for Al cathodes (150 nm).10 In order to enhance the adhesion of organic layer with Al, the substrate was exposed to N2-plasma (“sample A”) in a plasma treatment chamber.11 Two kinds of EIL, LiF (“sample B”) and MgO (“sample C”), were separately deposited on the plasma-treated Al cathode by thermal evaporator and rf magnetron sputtering, respectively. The electron injection efficiency between the cathode and the emitting layer depends on the EIL thickness.2,4–7,12 Thicknesses of LiF and MgO changed and corresponding current density-voltage (J-V) characteristics were investigated, summarized in Table I. The minimum turn-on voltages for both samples were obtained when thicknesses of LiF and MgO were 0.5 and 1 nm, respectively. The three types of samples were simultaneously loaded to the thermal evaporator, on which tris (8-hydroxyquinoline) aluminum (Alq3, 22 nm), 4, 4′-[N-(1-naphthyl)-N-phenyl-amino]biphenyl (α-NPD, 35 nm), copper phthalocyanine (CuPc, 18 nm) were deposited in sequence. Finally, Au (30 nm) layer as an anode was deposited. The active area of the device was 3 × 3 mm2. The J-V characteristics and luminance of the samples were measured.

In order to investigate the onset of photoemission corresponding to the vacuum level at the surface of Al, the three kinds of samples were loaded into a vacuum chamber, equipped with the electron analyzer, at 2B1 beam line in Pohang Accelerator Laboratory. The negative bias of −20 V on the sample was applied to avoid the work function of detector.

Figure 1 shows the J-V characteristics of the three types of samples. The insertion of the thin insulating buffer is ef-

![Figure 1](image-url)
effective in reducing turn-on voltage of $J-V$ curve. The turn-on voltage was most significantly reduced by inserting a MgO layer. The operating voltages at the current density of 50 mA/cm$^2$ were 12.5, 10.5, and 8.9 V, respectively, for samples A, B, and C. The tolerance in MgO thickness to improve the ITOLEDs was in the range of 0.5–2 nm, as given in Table I.

The dependence of the luminance on the injected current is shown in Fig. 2. The maximum luminance value in sample $A$ was 620 cd/m$^2$, but it increased to 1000 cd/m$^2$ in sample $C$. Such improvement in the luminance might be attributed to the decrease of the energy barrier by the MgO layer, enhancing the injection of electrons into the organic emitting layer.

To clarify the enhancement of electron injection by the MgO layer, the electron only devices were prepared with the following structures: Ti (20 nm)/Al (150 nm)/Alq$_3$ (50 nm)/Au, Ti (20 nm)/Al (150 nm)/LiF (0.5 nm)/Alq$_3$ (50 nm)/Au, and Ti (20 nm)/Al (150 nm)/MgO (1.0 nm)/Alq$_3$ (50 nm)/Au. As clearly shown in Fig. 3, the electron injection is the most effective as a thin layer of MgO was employed. Therefore, the enhanced electron injection promoted the balanced recombination with holes, resulting in the increase of the luminance, as shown in Fig. 2.

The relative change of the work function was measured using secondary electron emission spectra, as shown in Fig. 4. The onset of the secondary electron was determined by extrapolating two solid lines from the background and straight onset in the spectra. As shown in Fig. 4, the onset of the secondary electron for the samples with insulating layers shifted to the higher kinetic energy with respect to the onset for sample $A$. This result means that the work function of sample $C$ is higher than that of sample $A$. This could not explain the results in Fig. 1 because the increase in work function of the cathode means the increase of the turn-on voltage in the $J-V$ curve of OLED. As a result, the enhancement of the electron injection by inserting a 1-nm-thick MgO buffer layer between the Al cathode and Alq$_3$ cannot be explained by the decrease of the work function of the interfacial layer, but related to the formation of dipoles at both sides of MgO.

Such enhancement of the electron injection by the insertion of the insulating buffer in Figs. 1 and 2 can be understood on the basis of the energy band diagram, shown in Fig. 5. The electron injection barrier ($\Phi_{b}$) from the cathode to organic materials corresponds to the energy difference between the Al work function (4.3 eV) and the lowest-unoccupied molecular orbital (LUMO) energy level (2.55 eV) (Ref. 7) of Alq$_3$. Thus, the electron injection barrier, $\Phi_{b}$, is determined to be 1.75 eV, as shown in Fig. 1. As the forward bias is applied, the surface band bending changed to Fig. 5(b). When a forward bias is applied to the OLED, a considerable voltage can be dropped across the MgO layer through the formation of dipoles, resulting in the reduction of energy barrier for electron injection from Al to Alq$_3$. Thus, the LUMO energy level of Alq$_3$ aligned with the Fermi level of Al, enable direct tunneling of electrons through the thin insulating layer, as shown in Fig. 5(c). As a result, the electron injection could be enhanced by inserting the optimal thickness of insulating layer.

The MgO buffer layer is more effective in improving device performance than the LiF buffer layer. The relative potential drops in insulating layers of LiF and MgO can be determined from the equations proposed by Kim et al. considering the optimal thickness ($d$) and the dielectric constant ($\varepsilon$) of insulating layers. The ratios of $\varepsilon/d$ for LiF and MgO are 18.06 and 9.98, respectively. From the equation, the potential drop across the MgO layer induced by the electric dipoles is larger than LiF layer at a forward bias. Therefore, the energy barrier for electrons from Al to Alq$_3$ could be much lowered by the larger potential drop, resulting in the increase of balanced recombination with holes. Consequently, the emission intensity increased and the turn-on voltage of ITOLEDs reduced.

<table>
<thead>
<tr>
<th>LiF thickness (nm)</th>
<th>MgO thickness (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.5</td>
</tr>
<tr>
<td>Voltage at 100 mA (V)</td>
<td></td>
</tr>
<tr>
<td>15.7</td>
<td>10.9</td>
</tr>
</tbody>
</table>
In conclusion, the luminance and the turn-on voltage in ITOLEDs could be improved by inserting the optimal thickness of 1 nm MgO. The enhancement was due to the decrease of the electron potential barrier between Al and Alq3 through the potential drop across MgO layer induced by the electric dipoles. The enhancement of emission intensity originated from the balanced injection of electrons and holes into the ITOLEDs.

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