Inverted Top-Emitting Organic Light-Emitting Devices with An Effective Cathode Structure

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Summary

In this paper, we report an effective bottom cathode structure for enhancing the electron injection in inverted top-emitting OLEDs. Such a cathode structure does not involve handling reactive metals during fabrication and permits use of highly reflective materials such as Al and Ag as the bottom cathode.

One effective cathode structure often used in conventional OLEDs is to deposit an ultrathin layer (~nm) of LiF between the Al cathode and the organic electron-transport layer such as tris-(8-hydroxyquinoline) aluminum (Alq_3). Previous studies have revealed that the formation of an effective cathode with the relatively high-work-function Al is attributed to the release of Li from LiF in the coexistence of Alq_3, LiF, and Al and its subsequent reaction with Alq_3 to form Alq_3^- anion through the thermodynamically allowed reaction of 3 LiF + Al + 3 Alq_3 → AlF_3 + 3 $Li^+Alq_3^-$ [1,2]. Such reactions result in enhanced electron injection due to formation of a thin n-doped Alq_3 layer as in the cases of using Li-doped Alq_3 as the electron-injection layer or using Li as the low-work-function cathode [3,4]. It is also shown that an ultrathin Al overcoat (~nm) on LiF on Alq_3 is sufficient to induce such a reaction, and that the reacted layer has a shallow nature of only 1 nm or less [1,2]. It is therefore possible to employ the ultrathin Alq_3/LiF/Al trilayer as a composite electron-injection layer between various organic electron-transport layers and conductive cathode layers in conventional OLED structures (with the cathode on top), as demonstrated in few cases [1,5]. Whether such an electron-injection scheme may be applied to bottom cathodes of inverted top-emitting OLEDs, however, has not been explored and thus is the subject of this study.

To evaluate the effect of the ultrathin Alq_3/LiF/Al trilayer on electron injection from the bottom cathode, symmetrical "double-cathode" devices with the structure of substrate/bottom cathode (Ag or Al, 80 nm)/ultrathin Alq_3-LiF-Al trilayer/Alq3 (80 nm)/LiF (0.5 nm)/Al (100 nm) were fabricated and tested. The composite trilayer on top of the bottom cathode in sequence consists of 0.2 nm Alq_3, 0.2 nm LiF, and 0.3 nm Al. For comparison, control devices with nearly the same configuration but without the ultrathin trilayer were also fabricated. Fig. 1(a) and 1(b) show the current-voltage characteristics of the double-cathode devices with Ag and Al bottom cathodes, respectively. Without the ultrathin Alq_3-LiF-Al trilayer, the I-V characteristics are somewhat rectified with the reverse-bias current (injection from the bottom) being lower than the forward-bias current (injection from the top). The differences are over three orders of magnitudes for Ag and over one order of magnitude for Al. With the ultrathin trilayer inserted, the injection current under reverse bias is significantly enhanced for both cases to become nearly symmetrical with that under forward bias, indicating comparably effective electron injection from top and bottom cathodes. These results indicate that the reaction in the ultrathin Alq_3-LiF-Al trilayer produces a structure or composition that largely facilitates electron injection regardless of the cathode being deposited before or after the formation of this injection layer and regardless of the cathode being Ag or Al. Such features certainly give much greater flexibility in the design of cathode structures and in applications to various device configurations.

To demonstrate the usefulness of the present cathode structures, in inverted top-emitting OLEDs with the structure of Fig. 2 were fabricated and characterized. The devices were built on glass substrates precoated with Ag or Al (80 nm), which was used as the cathode directly (Fig. 2(b)) or was covered with the ultrathin Alq_3-LiF-Al trilayer before following deposition (Fig. 2(a)). The organic multilayer structure followed sequentially consists of Alq_3 (50 nm) as the electron-transport and emitting layer, α-naphthylphenylbiphenyl diamine (α-NPD, 40 nm and 45 nm for devices using Ag and Al cathodes, respectively) as the hole-transport layer, 4,4',4"-tris(3-methylphenylphenylamino)triphenylamine (m-MTDATA, 20 nm) doped with 2 wt.% of tetrafluorotetracyano-quinodimethane (F4-TCNQ) as the hole-injection layer. A thin layer (20 nm) of Ag, which has relatively low optical absorption and the highest conductivity among all metals, is deposited as the (semi-)transparent anode with a low sheet resistance of ~1 Ω/square. The Ag anode is further capped with a high-refractive-index dielectric layer of thermally evaporated TeO_2 (n~2.2-2.3) as an index-matching (or antireflection) layer to enhance the optical transmission of the thin metal anode [6]. The thicknesses of organic layers and the TeO_2 layer (30 nm) had been optimized for colors and efficiencies of devices according to a methodology described in refrence 6.

Fig. 3(a) shows the measured electroluminescence (EL) spectra with relative intensities (symbols) at viewing angles of 0°, 30°, and 60° off the surface normal for the top-emitting device using the Ag cathode with the ultrathin

trilayer. EL spectra of devices using the Al cathode (not shown) are similar to those in Fig. 3(a). EL of the top-emitting devices shows more saturated colors in comparison with photoluminescence (PL) of Alq_3 due to the microcavity effect [6]. Usual variation of colors with viewing angles associated with microcavity effects have been nearly eliminated in the present case through optimizing optical structures of devices (i.e. thicknesses of organic and dielectric layers) according to a methodology developed previously [6]. Fig. 3(a) also shows that the simulated EL spectra (lines) calculated using the electric-dipole-based classical electromagnetic model [6] are in excellent agreement with measured results (symbols). In the simulation, it is also found that the effects of the ultrathin injection trilayer on optical characteristics are negligible and that the optimal layer thicknesses are slightly different for Al and Ag cathodes due to differences in their optical constants.

Fig. 3(b) compares the I-V characteristics of four inverted top-emitting devices examined. As expected, devices with the electron-injection trilayer exhibit lower voltages than devices without the trilayer no matter whether Ag or Al is used as the cathode. Devices using the Al cathode show somewhat higher voltages than devices using the Ag cathode due to thicker organic films used for optimizing optical characteristics. Fig. 3(c) and the inset of Fig. 3(c) compare the brightness-current (L-I) characteristics and the efficiency characteristics of the four devices, respectively. Without the electron-injection trilayer, the device uisng the Al cathode shows a low EL efficiency of 1.7 cd/A and the device using the Ag cathode gives no emission at all. Such results indicate that electron injection from Ag to Alq_3 is extremely unbalanced with hole injection, and that the situation for Al is less worse yet still not competitive with hole injection. With the electron-injection trilayer, the Ag and Al devices achieve efficiencies of 5.3 cd/A and 4.5 cd/A, respectively. Both are comparable to or somewhat higher than EL efficiencies of a state-of-the-art non-doped (bottom-emitting) Alq_3 device [1,3,4], indicating that electron injection has been enhanced to become rather balanced with hole injection.

In summary, we report the use of the ultrathin Alq_3-LiF-Al trilayer as an effective composite electron-injection layer for bottom cathodes of inverted top-emitting OLEDs. Such an approach does not involve handling reactive metals during fabrication and permits use of highly reflective materials such as Al and Ag as the bottom cathodes. Efficient inverted top-emitting devices employing such a cathode scheme has been demonstrated.

References

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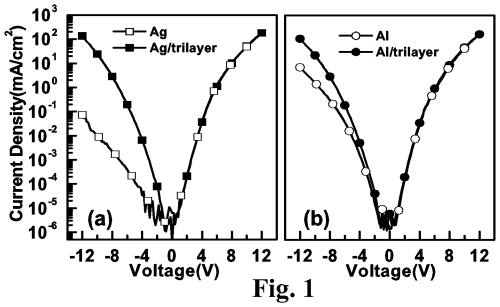

Fig. 1

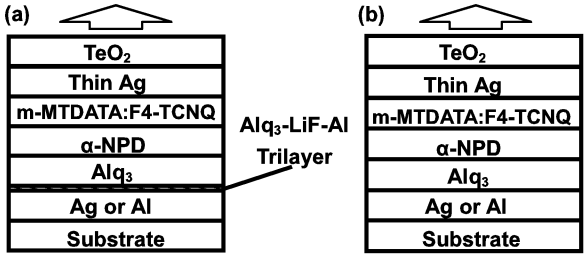

Fig. 2

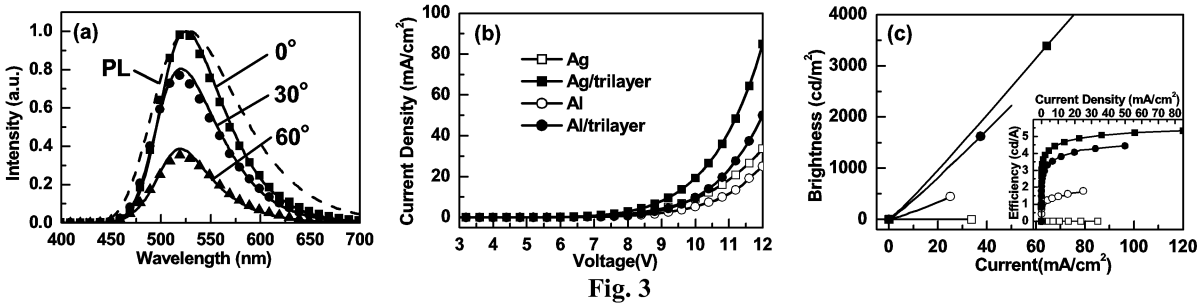

Fig. 3