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Study of the Yellow Organic Light Emitting Diodes using ZnS/Metal/ZnS Multilayer Anodes and Investigation of the Effect of BCP on Electroluminescent Spectra of the Devices

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Abstract: The effects of ZnS/Metal/ZnS multilayer anodes, including ZnS/Au/ZnS (ZAuZ), ZnS/Ag/ZnS (ZAgZ) and ZnS/Cu/ZnS (ZCuZ) on device performance in organic light emitting diodes based on rubrene were investigated systematically. The device with ZAuZ as anode gave an improved performance comparable with ITO based devices. A low driving voltage of 5.61 V and a high luminance of 1836 cd/m^2 were obtained in the ZAuZ based device. Then the effect of BCP as hole blacking layer on emissive spectra of ZAuZ based OLEDs was investigated and white spectra with (0.32,0.31) colour coordinates was obtained for ZAuZ/PEDOT:PSS/TPD/BCP/Rubrene/BCP/BPhen/LiF/Al multilayer structure.

Keywords: Transparent conductive anode; Yellow OLED; Rubrene; BCP

Introduction

In general, transparent conductive anodes are an integral part of all organic light emitting devices [1]. Nowadays demand will grow for low-cost anodes while retaining good optoelectronic properties. The studies show that dielectric/metal/dielectric (D/M/D) multilayer systems with optimum structures, possessing very low sheet resistance and high transparency, are good candidates for use as transparent conductive anodes [2]. The multilayer ZnS/Metal/ZnS (ZMZ) anodes are Suitable alternatives to ITO for use in OLED devices [3].

In this paper we study the effects of ZMZ multilayer anodes, including Au, Ag and Cu as a metal layer on OLED devices performance. Also, effect of hole blocking layer (BCP) on EL spectra of ZAuZ based device has been investigated.

Experimental

Prior to fabricating the OLED devices, Nanostructured thin films of ZMZ with the optimized structures obtained by characteristic matrix theory, where detailed simulation procedures are presented elsewhere [4,5], were deposited on a glass substrate by thermal evaporation.

OLED devices under study were based on PEDOT:PSS as a Hole Injection Layer (HIL), TPD as a Hole Transport Layer (HTL), BPhen as an Electron Transport Layer (ETL), BCP as a Hole Blacking Layer (HBL), Rubrene as an emitting layer, and LiF/Al as a cathode with different transparent ZMZ anodes. All the inorganic and organic materials were purchased from Aldrich Chemical Co. and used as received. Also, control samples on conventional ITO-coated glasses (Aldrich Chemical Co. R_s=15 Ω/sq) were fabricated in the same batch for comparison.

The electroluminescent (EL) spectra, current density–voltage (J-V), and luminance characteristics of the samples were measured using Keithley 2400 source-meter and a Jaz spectrometer (Ocean optics). All the measurement was carried out in room temperature and under ambient conditions without any encapsulation.

Results and Discussion

Fig. 1 (a) and (b) depicts current density-voltage (J-V) and luminance-voltage (L-V) characteristics for ZMZ/PEDOT:PSS/NPB/Rubrene/BPhen/LiF/Al devices with Au, Ag and Cu as a metal layer for anodes.

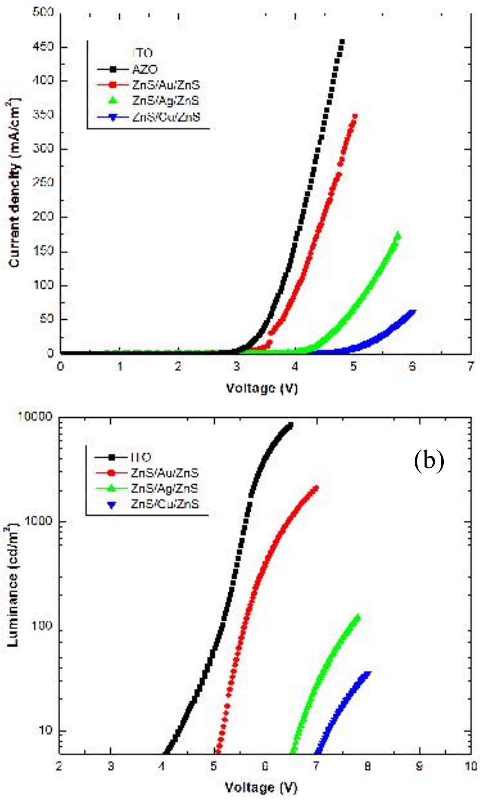

Fig. 1: (a) Current density-Voltage and (b) Luminance-Voltage characteristics of Anode/ PEDOT:PSS/ NPB/ Rubrene/ BPhen/ LiF/ Al devices with different anodes.

The J-V and L-V characteristics of the optimized reference control device (ITO based) is also given for comparison. When Au was used as a metal layer for the multilayer anode, we observed that a high current density and luminance which is comparable to that of the ITO based control device at the same operating voltage. Also, as it can be seen in Table 1, the ZAuZ based devices exhibit desirable OLED characteristics such as a turn-on voltage (V_{TO}) (defined as the voltage to start the driving current) as small as 3.214 V and a driving voltage (V_D) (defined as the voltage of OLEDs to emit a light with luminance of 100 cd/m^2) of 5.61 V. This level of performance is well compared with those of ITO based control samples which exhibited V_{TO} of 3.016 V and V_D of 5.16 V.

Table 1 The effect of the ZMZ multilayer anodes on the electro-luminescent characteristics of the yellow OLEDs.

Anode structure	V_{TO} (V)	Current density at 5 V (mA/cm^2)	V_D (V)
ITO	3.016	458	5.16
ZnS/Au/ZnS	3.214	348	5.61
ZnS/Ag/ZnS	4.16	69	7.67
ZnS/Cu/ZnS	4.769	9	-

The surface properties of the ZMZ anodes as well as ITO anode can also affect the characteristics of the device. Therefore, it is very important to examine the surface morphology of these anodes. Fig. 2 shows SEM images of the surface of ZMZ multilayer films annealed at temperature 200 °C and surface of ITO film.

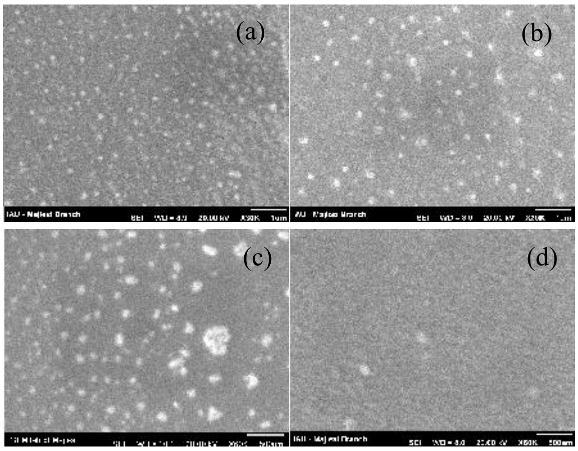

Fig. 2: SEM images of different anodes (a): ITO, (b) ZnS/Au/ZnS, (c) ZnS/Ag/ZnS, (d) ZnS/Cu/ZnS.

The surface morphology of all ZMZ transparent conducting films and ITO film showed a dense and uniform surface. However, grain size of ITO film is larger than that of other films and it has improved structural homogeneity. Thus, it is expected that it's carrier dispersion and mobility is lower and higher, respectively, compared to the other films, respectively. These results show that the OLED device performance made with the ITO film as an anode is superior to the other devices due to increased hole injection and transport efficiency from the ITO anode layer to the organic layer. Although, among the ZMZ multilayer systems, the ZAuZ film exhibited the improved structural properties compared to other films, (ZAgZ and ZCuZ). As a result, ZAuZ based OLEDs show improved performance compared to the ITO based OLEDs and so ZAuZ multilayer films can be used as the anode in OLED devices.

In following, we investigated the effect of BCP layer on EL spectra of yellow organic light emitting diodes. The structures used for the yellow OLEDs fabricated in this work are as follows:

Device 1: ZAuZ/PEDOT:PSS/TPD (30nm)/Rubrene (1nm)/BPhen (35nm)/LiF/Al.

Device 2: ZAuZ/PEDOT:PSS/TPD (30nm)/Rubrene (1nm)/BCP (10 nm) /BPhen (35 nm)/LiF/Al.

Device 3: ZAuZ/PEDOT:PSS/TPD (30nm)/BCP (8nm)/ Rubrene (1nm)/BCP (10nm)/BPhen (35nm)/LiF/Al.

Fig. 3 shows the proposed energy level diagram of the devices studied, with the relative alignment of the HOMO and LUMO levels of each layer.

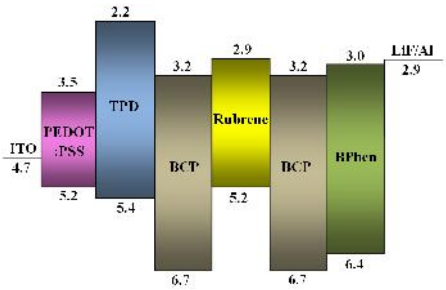

Fig. 3: Schematic illustration of energy level diagram of the devices.

It can be seen from Fig. 3 that in these devices, BCP with high HOMO was used as hole blocking layer. Fig. 4 shows the EL spectra of device D1, D2 and D3 under driving voltage of 12 V.

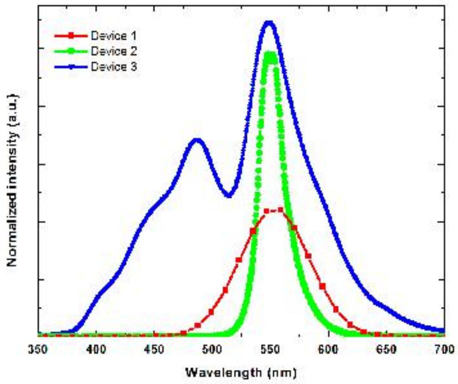

Fig. 4: The normalized EL spectra of the different devices under the driving voltage of **12 V.**

As it can be seen in Fig. 4, for D1 and D2 devices there is only one emission peak at 550 nm which can be attributed to Rubrene. However, the higher EL intensity of D2 could be attributed to hole blocking characteristic of BCP layer. In fact, BCP as an HBL has a strong hole blocking ability, as well as more chance to aggregate of holes into EML and recombination probability in the emitting layer with electron carriers. As a result, the balance in the number of holes and electrons improves and EL intensity of D2 increases.

In addition to Rubrene exciton emission, we also observed one emission peak around 486 and four shoulders around 401, 442, 590 and 630 nm from EL spectrum of D3 device. According to the previous reports [6] the 401 emission shoulder originate from the exciton emission of TPD molecule, and the 442 and 486nm emissions are singlet exiplex from the interface between TPD and BCP. In addition, the 590 and 630 nm emissions that are red shift than the singlet exiplex emission maybe belong to electroplex emission. However, according to results reported in Ref [7], it can be seen that the emission shoulder at 590 nm should be ascribed to the (TPD:BCP) exciplex triplet and only the emission shoulder at 630 nm could belong to an electroplex emission.

The EL spectra of D3 device changes with the applied voltages, as shown in Fig. 5.

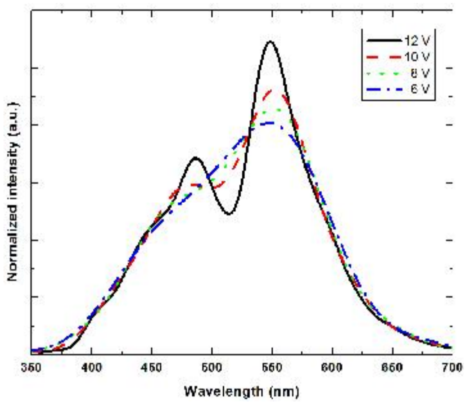

Fig. 5: The normalized EL spectra of devices under driving voltages varied from 6 to 12 V**.**

The first, it is observed that there is a yellow emission at 550 nm in the EL spectra. It is known that the rubrene dye as yellow emitter has PL quantum efficiency about %100 and recombination probability is efficient in it. At bias voltages lower than 12 V, in addition to the yellow emission, the shoulder of exiplex emission can be seen due to the strong exiplex mechanism at interface of TPD/BCP and its intensity increases with increase in applied voltage. However, the singlet exciton emission at 401 nm was not observed because a number of the exciton states are low in TPD layer. Furthermore, two emissions at 590 and 630 nm may not be observed due to the decrease in electric field at the interface of TPD/BCP. The pure white light emission can be observed at the driving voltage of 12 V, with CIE coordinates of (0.31, 0.33), which is very close to the equi-energy white point (0.33, 0.33).

Conclusions

This study examined the effect of the ZnS/Metal/ZnS multilayer systems with Au, Az and Cu as a metal layer, on the performance of yellow OLEDs. The optoelectrical properties of OLEDs based on the Z/Au/Z multilayer anode show improved characteristics compared to ITO-based devices. The OLEDs fabricated with ZAuZ as the anode show excellent brightness of more than 1800 cd/m^2. Also, the effect of BCP layer on EL spectra of yellow OLEDs was investigated and pure white light emission via mixing organic molecular, exciplex and electroplex has been obtained in ITO/PEDOT:PSS/TPD/BCP/Rubrene/BCP /BPhen/LiF/Al nano-multilayer structure.

Acknowledgment

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