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Color-tunable organic light-emitting devices using a thin N,N'-bis-(1-naphthyl)-N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine layer at heterojunction interface

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Abstract

Novel types of multilayer color-tunable organic light-emitting devices (OLEDs) with the structure of indium tin oxide (ITO)/N,N'-bis-(1-naphthyl)-N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB)/aluminum (III)bis(2-methyl-8-quinolinato)4-phenylphenolato (BAlq)/tris-(8-hydroxyquinolate)-aluminum (Alq₃)/Mg:Ag were fabricated. By inserting a thin layer with different thickness of a second NPB layer at the heterojunction interface of BAlq/Alq₃, the emission zone of devices shifted greatly and optoelectronic characteristics underwent large variation. Although BAlq was reported as a very good hole-blocking and blue-light-emission material, results of measurements in this paper suggested that a certain thickness of NPB layer between BAlq and Alq₃ plays an important role to modify device characteristics, which can act as recombination-controlling layer in the multilayer devices. It also provides a simple way to fabricate color-tunable OLEDs by just changing the thickness of this "recombination-controlling" layer rather than doping by co-evaporation. © 2006 Elsevier B.V. All rights reserved.

Keywords: Organic light-emitting devices; Hole-blocking layer; Color tunable

1. Introduction

Organic light-emitting devices (OLEDs) have been attracting a lot of attention because of their potential application in flat panel displays (FPD). In the past two decades since efficient double-layer OLEDs were first reported by C.W. Tang et al., tremendous progress has been made in device fabrication and performance optimization, highly efficient and colorful devices have already been fabricated for commercial use [1–5]. Almost all giants in display around the world, e.g., Samsung, Sony and Philips, were involved in developing OLEDs for the next generation of FPD. Some people even estimate that OLEDs will replace liquid crystal displays in the next decade. However, there still remain some fundamental

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mechanisms of devices for scientists to deal with, such as long-term stability of organic materials or device, transportation and recombination of carriers and energy transferring process. In addition, finding a simple fabricating way to realize full-color OLEDs is also an important and imperative approach to reduce manufacturing cost in industrial production.

In this paper, instead of being partially or totally doped in the emissive layer or carriers-transport layer in conventional devices, color-tunable devices with simple multilayer structures were achieved. The device structure in this work is indium tin oxide (ITO)/N,N'-bis-(1-naphthyl)-N,N'diphenyl-1,1'-biphenyl-4,4'-diamine (NPB)/aluminum (III) bis(2-methyl-8-quinolinato)4-phenylphenolato (BAlq)/NPB/ tris-(8-hydroxyquinolate)-aluminum (Alq₃)/Mg:Ag without doping in any layer of the device. The recombination of charge carriers and energy transferring processes during device operation was investigated. Results suggested that a thin NPB layer at the interface of BAlq/Alq₃ can act as

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recombination-controlling layer except for its common employment as hole transport layer (HTL) [6,7] or light-emitting layer [8,9] in OLEDs.

2. Experimental

ITO glasses, as the anode substrates, were sequentially subjected to a routine cleaning procedure, namely sonicated in diluted detergent solution, ethanol and acetone for 15 min, respectively. After each step, the substrates were rinsed with deionized water thoroughly. The cleaned substrates were sequentially dried in nitrogen gas, exposed to oxygen plasma treatment for 15 min and then loaded into vacuum chamber for organic materials deposition at a pressure of 1×10^{-4} Pa and metal cathode deposition in $\sim 10^{-3}$ Pa without breaking the vacuum. Device configuration was as following: ITO/NPB (30 nm)/BAlg (10 nm)/ NPB $(x \text{ nm})/\text{Alq}_3$ (10 nm)/Mg:Ag(100 nm). Here, in these device samples, the first thicker layer of 30 nm NPB was used as HTL, followed by a 10 nm layer of BAlq as holeblocking and light-emitting layer [8-10]. The second thinner NPB layer was as recombination-controlling layer (referred to as the second NPB layer below), of which the thickness ranges from 0 to 18 nm, followed by 10 nm Alq₃ as light-emitting and electron-transporting laver and 100 nm Mg:Ag(10:1) layer as cathode. All these devices are denoted as A, B, C, D, E, F, which correspond to x = 0, 1.8, 3.6, 4.2, 6, 18 nm, respectively. The effective emitting areas of all samples are $5 \times 5 \text{ mm}^2$. Fig. 1 shows the chemical structure of organic materials and device configuration used in this study. Electroluminescence (EL) spectra of devices were recorded with OPT-2000 photo-



NPB

BAlq



Fig. 1. Chemical structure of organic materials and device configuration used in this study.

meter. The characteristics of current–voltage (I-V) and luminance–voltage (L-V) were measured with a Keithley 4200 source meter. All of the measurements were performed at room temperature in air.

3. Results and discussion

Fig. 2 shows the normalized EL spectra of all devices. It can be obviously seen that EL spectrum of device without the second NPB layer between BAlq and Alq₃ layers (device A) peaks at 483 nm which comes from BAlq layer, and the color of light emission from device A appears to be white blue, which is in good agreement with the results reported by Itoh et al. in Refs. [11–14]. While a thin NPB layer, 1.8 and 3.6 nm for devices B and C, respectively, was inserted between BAlq and Alq₃ layers, no changes of EL spectra of these devices took place and the emissive peak still locates at 483 nm. Spectra of these three samples overlapped very well.

As the thickness of the second NPB layer was further increased to 4.2 nm in device D, the peak of EL spectrum of the device was dramatically removed from 483 to 530 nm and EL color turns to be yellow green accordingly. It clearly indicates that the recombination of charge carriers occurred in Alq₃ layer rather than in BAlq layer in this device.

These phenomena can be well explained by the energy band diagram of devices under forward-biased voltage as shown in Fig. 3. It can be seen that if there is no the second NPB layer at heterojunction interface, electrons injected from the cathode can be easily transported through Alq₃ and BAlq layer [10,11] and accumulated in the bulk of BAlq layer near the interface between NPB HTL and BAlq layer. On the other hand, holes injected from the anode can be easily transported through the first NPB HTL and accumulated at the NPB/BAlq interface due to the high hole-blocking capability of BAlq layer, as reported in Ref. [11]. As a result, these concentrated holes and



Fig. 2. EL spectra of devices as a function of film thickness of NPB layer between BAlq and Alq₃.

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