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Dual Ag electrodes for semitransparent organic light-emitting diodes

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ABSTRACT

Thick ReO_3 film (> 30 nm) is found to serve as an active hole-injecting layer as well as a passive buffer layer to suppress point discharge of Ag anode. This buffered Ag electrode enables fabrication of transparent organic light emitting diodes (TOLEDs). The excellent conductivity of ReO_3, second only to metal amongst oxide semiconductors, is made possible to use a thick hole-injecting buffer layer. The use of thick ReO_3 layer alleviates strong local electric field on a rough electrode surface. For device optical design, the transfer matrix method is used to provide theoretical guidance on electrode fabrication. Based on microcavity theory in TOLEDs, the optical path parameters are also optimized to the thicknesses of different functional organic films. To test the device performance, we fabricated normal fluorescent green Alq_3 devices. The data show that TOLEDs with ReO_3-buffered Ag electrodes have good photochromic uniformity on both sides of the devices.

1. Introduction

Organic light-emitting diode (OLED) is a very promising luminescent device for next generation displays and solid-state lighting applications due to its broad viewing-angle, fast response, low driving voltage, and flexibility [1–4]. Presently, it has been applied in mobile phones and digital electronic products. There are further developments towards active full color display, silicon–based micro-display (OLED on Silicon, OLEDoS) [5], transparent flexible display (FOLED) [6], and large area OLED [7,8] lighting panel.

Based on which side the light emerges, OLEDs can be divided into three types: Bottom-Emitting OLEDs (BEOLED), Top-Emitting OLEDs (TEOLED) and Transparent OLEDs [9]. A BEOLED is fabricated on ITO glass with multiple layers of organic films along with a metal film acting as a reflecting cathode. In a BEOLED, the light emerges from the ITO glass side while a TEOLED uses the reflecting metal deposited on the glass as the electrode and has light emerging from the top transparent/semitransparent electrode. If the lights emerge from both sides of the OLED, we call it a TOLED. The basic device structures of a BELOD, TEOLED, and TOLED are shown in Fig. 1.

Due to its double emitting properties, transparent OLEDs open up a variety of new applications including, but not limited to windshield display of a car and illuminated windows of a building.

Fabricating a transparent OLED can be challenging due to the difficulty of finding an electrode material with advantageous characteristics. In order to ensure device efficiency, the material must exhibit high qualities in transparency, injecting efficiency of carriers, and conductivity. ITO (Indium Tin Oxide) [10] is a very good transparent electrode material. Currently, ITO has been widely used in BEOLEDs because of its matured fabrication sputtering process. Generally a thick blocking or buffer layer of SiO_2 is pre-deposited on the glass for smoothness and infiltration requirements. Additionally, ITO can be used as a cathode [11]. However, the work function of ITO is fairly high which suppresses electron injection and thus, decreases the performance of the device. Furthermore, indium is known to be an expensive element that is fairly scarce on the earth. The high energy sputtering technology will damage the organic functional layer. Also, due to its fragility, ITO is unfavorable to be used in flexible devices. Hence, it is significant to explore better alternatives without the use of ITO and its unfavorable sputtering process. A cheaper and simple transparent or semi-transparent electrode material can become an important solution for commercial applications.

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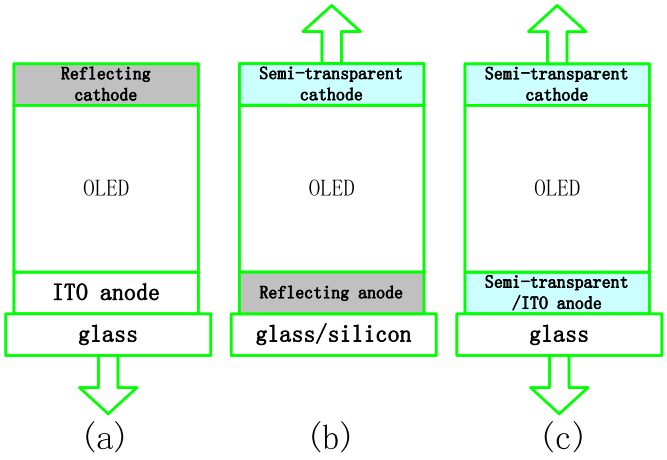

Fig. 1. (a) BEOLD; (b) TEOLED; (c) TOLED.

2. Material and method

2.1. Materials selection

Ideal electrode materials such as Al and Ag are commonly used as reflecting cathodes in typical ITO-based BEOLED because of their appropriate work functions and high conductivity. The light can transmit through a thin metal film, acting as a light emergence electrode, ultimately replacing ITO in the process. Due to its ductility, metal electrode is favorable to be used in flexible devices. In visible wavelengths and for thin films, Ag is better than Al as a semi-transparent electrode due to its high transmittance, conductivity, and stability. More specifically, these metal characteristics arise from smaller refractive index and extinction coefficient.

However, Ag has bad infiltration deposited on normal glass substrates which leads to high surface roughness. In comparison, when it is deposited on the surface of Al, it achieves excellent morphology [12].

During the working process of an OLED, the surface roughness of the Ag metal electrode will lead to point discharge and thus, a short circuit. The point discharge on the anode is more severe than the cathode because of the corona discharge polarity effect [13]. In fact, because this is evident in many fabricated OLED devices, it causes disturbances in research. Given this, we have made great efforts to solve this difficult issue. We finally propose two methods to solve the Ag anode short circuit issues. Essentially, we co-evaporate two metals, like Ag/Al, Ag/Mg to form an alloy electrode. It changes the ambience of Ag vapor crystallization to form a film with good morphology. Secondly, we evaporate some materials with good transparency and conductivity to cover the rough Ag film to alleviate the electric field of point discharge, thereby avoiding a short circuit.

However, the first method has a relatively complex co-evaporation process, can cause high doping ratio of another meta land has decreased transparency. On the other hand, in the second method, we find that the transition metal oxide semiconductor (TMOS) has the potential properties to solve the short circuit problem. To this extent, we focus on ReO_3 and its single layer evaporation.

In previous literature, ReO_3 was commonly used as the hole-injecting layer (< 10 nm) in BEOLED to modify the ITO [14]. Recently, in regard to injecting layers, researchers applied MoO_3 as a better alternative to ReO_3. After experiment conduction, we recognized ReO_3 as an unusual oxide due to its electrical properties: the lowest resistivity $\sim 1 \times 10^{-7}\,\Omega\,m$ of all reported inorganic TMOS at room temperature. This resistivity value is approximately 6 times larger than that of Ag and 1/60 of MoO_3 ($5.98 \times 10^{-6}\Omega\,m$). ReO_3 behaves like a metal with good optical transparency. We use ReO_3 as the buffer layer and hole injecting layer on a 20 nm Ag anode on normal glass with varied thicknesses. Through experimentation, we discovered that OLED short circuit issue is eliminated when the buffer layer thickness is more than 30 nm. This method not only enables the use of Ag but also makes it possible to complete fabrication process in the same chamber. This provides a convenient fabrication process and reduces the overall costs. In comparison, we also tried the same device fabrication process with MoO_3, MoO_2, WoO_3, RuO_2 (common known TMOS with good conductivity) but did not resolve the short circuit issue.

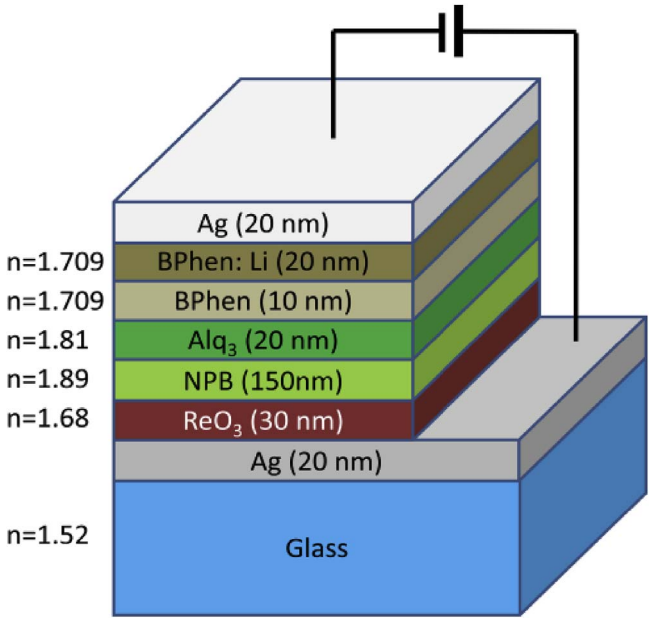

Fig. 2. The structure of TOLED.

2.2. Device fabrications

We select the thickness of every layer by combining our experiences with the optical simulations under the assumption that the light emitting position is on the interface between luminous organic functional layers of Alq_3/NPB. The structure is listed in Fig. 2.

ReO_3 works as the hole-injecting layer and anti-short circuit layer, N,N′-di-[(1-naphthalenyl)-N,N′-diphenyl]-1,1′-biphenyl-4,4′-diamine (NPB) as the hole-transporting layer, 8-hydroxyquinoline, aluminum salt(III) (Alq_3) as the luminescence functional layer, 4,7-diphenyl-1,10-phenanthroline (BPhen) doped with Li as the electron-injecting and transporting layer for reducing the injecting barrier and enhancing the conducting ability, and 10 nm BPhen as a spacing layer to deduce the quenching effect from Li diffusion into the light-emitting layer.

All the materials used in this work are as-received with the purity higher than 99%. The samples were fabricated on normal optical glass substrates without ITO, which was cleaned with acetone, dehydrated alcohol and deionized water for 10 min, and then dried at 120 °C step by step.

The active area of each device is 0.09 cm^2 (3 mm × 3 mm). The devices were fabricated in a normal vacuum thermal evaporation system with multiple evaporation sources including organics and the metal through changing the masks. The base vacuum pressure is about 4×10^{-6} τ. The deposition rates and doping concentrations of materials were controlled and measured in situ using thickness monitors SI-TM606A calibrated by Spectroscopic Ellipsometer alpha -SE. The deposition rate of the host organic and metal materials was 0.2–0.3 nm/s, and that of the doped material were adjusted according to the weight ratio in the host materials. To avoid the decomposition, the deposition rate of ReO_3 is only 0.02 nm/s.

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