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Solid-State Electronics 48 (2004) 2085–2088

SOLID-STATE ELECTRONICS

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Dependence of film morphology on deposition rate in ITO/TPD/Alq_3/Al organic luminescent diodes

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Received 10 December 2003; accepted 15 March 2004

Abstract

Organic multilayers of *N,N′*-diphenyl-*N,N′*-bis(3-methylphenyl)-1,1′-biphenyl-4,4′-diamine(TPD)/aluminum tris(8-hydroxyquinoline)(Alq_3) were evaporated on the indium tin oxide (ITO) coated glass substrates under different conditions. The effect of deposition rate on the surface morphology of top Alq_3 films and *I–V* curves of organic luminescent diodes (OLED)s is studied, in order to optimize the Alq_3 film growth conditions. SEM and AFM images show dense, uniform surface for high deposition rate (>0.4 nm/s) devices, and local pinholes on the surface for device of low (<0.15 nm) deposition rate. *I–V* characteristic measurement shows resistive characteristics with no luminescence for low deposition rate devices, and diode characteristics with characteristic green luminescence for the high deposition rate. The deposition rate is identified as one of the key factors in the performance of the TPD/Alq_3 device due to its great influence on the surface morphology of top Alq_3 films.

PACS: 68.65.–z

Keywords: TPD/Alq_3 organic diode; Growth rate; Surface morphology

1. Introduction

In the last decade, organic light emitting diodes (OLED) have attracted great attention in both fundamental research and device fabrication because of their potential application to display components [1–8]. Tremendous progress has been made, and OLED technology is expected to show great impact on the future of general lighting application and flat-panel displays [9,10]. Achieving this will require significant advances in efficiency and life at high brightness. However, problems remain in reliability and fabrication in these OLED structures. A key obstacle to the development of large area OLED is the presence of local defects which cause electrical shorts [11,12]. Some shorts are relatively benign in that they "burn out" during operation resulting in only a small nonemisssive area [13]. However, some do not burn out and develop over time [14].

In particular, pinhole shorts can appear between the organic layers, short-circuiting current which should be directed into the recombination layer of the OLED. In localized areas, these shorts can cause dark spots; with sufficient pinhole density, the device no longer luminesces at reasonable voltages since current flows through the short rather than the working areas of the device. Cause of shorting defects include particle contamination during fabrication, asperities from electrode roughness and nonuniformities in organic layer thickness [13,14]. In this paper, the effect of growth kinetics, governed by deposition conditions, on the formation of pinholes is studied, in order to optimize the film growth conditions.

2. Experiment

In order to study the mechanism for this dense pinhole formation, test layers of Alq_3 films on TPD on ITO

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0038-1101/$ - see front matter
doi:10.1016/j.sse.2004.05.067

glass were deposited under different conditions. Before film fabrication, all ITO substrates were immersed in ethanolamine heated to 80 °C for 20 min and subsequently ultrasonic cleaned in acetone and alcohol for 20 and 10 min, respectively. Cleaned samples were rinsed in deionized water, dried and mounted in the vacuum chamber immediately. The basic structure of our organic luminescent diode is a double layer structure with a hole-transporting layer of *N,N′*-diphenyl-*N,N′*-bis(3-methylphenyl)-1,1′-biphenyl-4,4′- diamine (TPD) and an emitting/electron transport layer of aluminum tris(8-hydroxyquinoline) (Alq_3) sandwiched between a metal anode contact (Al) and a transparent cathode (ITO) on glass fabricated in a metal cross-linked configuration (see Fig. 1). TPD and Alq_3 organic films were deposited sequentially by vacuum evaporation/sublimation in a thermal evaporator with thicknesses of 50 and 40 nm, respectively. The growth rate of TPD films was kept at 0.3 nm/s and that for Alq_3 was varied. The ITO-coated glass substrate was not heated during the depositions. Al films, applied as metal anode, were sputtered on the top of the TPD/Alq_3 organic multilayer with the thickness of 150 nm on the same equipment. These devices had an area of about 1cm^2. The base vacuum for the evaporation and sputtering process was approximately 5×10^{-6} Torr. Typically, the Alq_3 and TPD were deposited without breaking vacuum, and the Al was deposited after a brief vacuum break.

Current–voltage (*I–V*) characteristics were measured between Al and ITO electrodes using Keithley source meter (2400 series), and the morphology of the resultant Alq_3 film were observed under scanning electron microscope (SEM) (Hitachi S-4000 Field Emission) and Atomic Force Microscope (AFM) (Digital Instruments, Dimension ™3100 series). All tests were performed in air at room temperature.

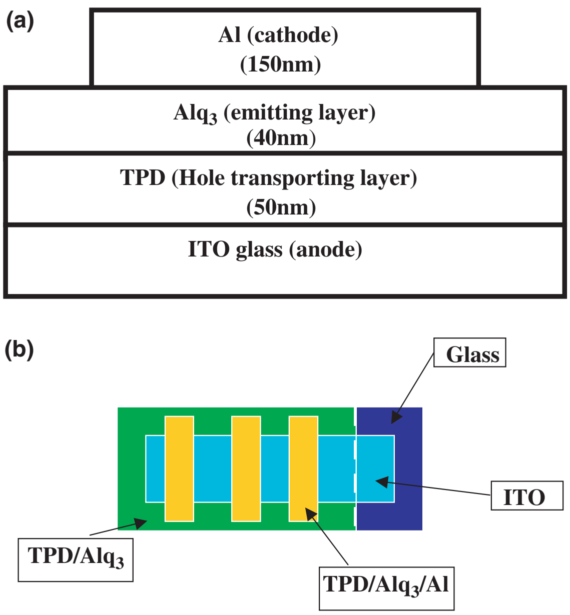

Fig. 1. (a) Schematic drawing of the TPD/Alq_3 organic luminescent diode structure. (b) Cross-linked pattern on ITO glass.

3. Results and discussion

Fig. 2 shows the SEM pictures of the surface morphology of TPD/Alq_3 (50/40 nm) layer fabricated on ITO glass with the evaporation rate of 0.15 nm/s at room temperature. The SEMS were taken with an environmental SEM operating at a pressure of a few Torr. Pinholes with the size of 10–30 nm and randomly oriented crystals (~20 nm) with fairly wide gaps can be observed on the surface, and the pinhole density, estimated from SEMS of small area, was of the order of 10^9/cm^2. AFM images (inset) also showed pinholes as well as

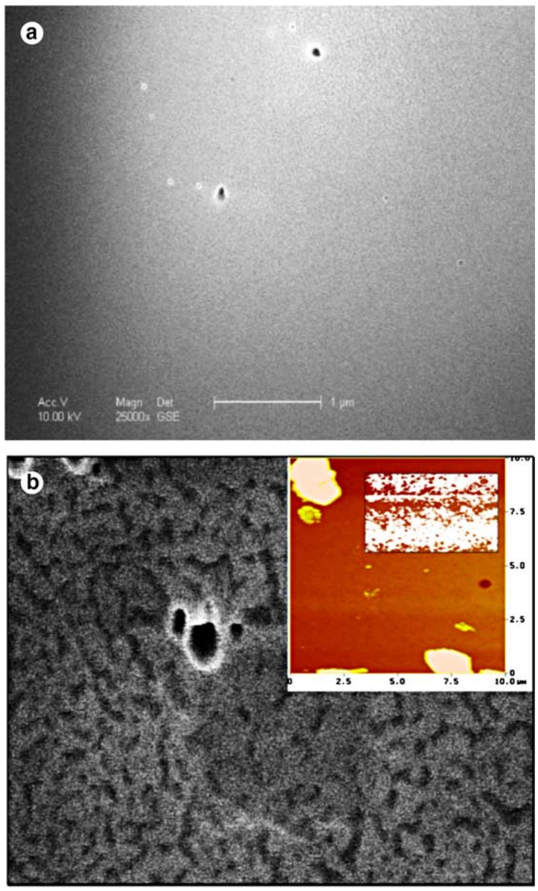

Fig. 2. SEM images of TPD/Alq_3 double layer fabricated with the evaporation rate of 0.15 nm/s. (a) 25 μm^2 region, (b) 0.25 μm^2 region.

pillars where the height was much higher. These pinholes will short current from the anode directly to the underlying TPD and cathode, preventing the current from contributing to luminescence, and precluding luminescence at reasonable current levels.

Fig. 3 shows the surface morphology of a TPD/Alq_3 (50/40 nm) double layer fabricated with a higher evaporation rate of 0.5 nm/s. A dense and smooth surface image was found, with no pinholes observed and a smaller, more ordered surface texture was seen under higher magnification. The roughnesses for both films, obtained from AFM measurements in regions where there were no pinholes, is about 1.7 nm. Higher evaporation rate is more favorable for the formation of pinhole-free TPD/Alq_3 double layer, which is a key factor for the success of organic luminescent diodes. The Alq_3 layers which are kinetically driven at rapid deposition are uniform, while those that are slower allow for molecular redistribution and form high densities of pinholes. The mechanism for this pinhole formation is not clear.

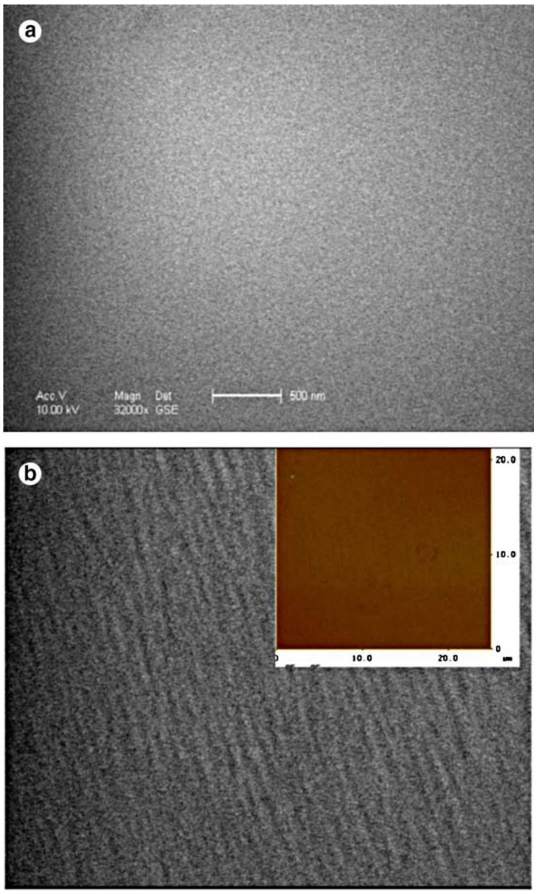

Fig. 3. SEM images of TPD/Alq_3 double layer fabricated with the evaporation rate of 0.5 nm/s. (a) 12.25 μm^2 region, (b) 1 μm^2 region.

The best obtained diode characteristic *I–V* curve is shown in Fig. 4(a) with a turn-on voltage about 5 V for device fabricated with high Alq_3 deposition rate. The TPD/Alq_3/Al diode (30/40/90 nm) was fabricated with the growth rate of Alq_3 at 0.4 nm/s. (This device had Al thermally deposited rather than sputtered). Fig. 4(b) shows the *I–V* characteristics of two diodes identically fabricated except for different deposition rates for Alq_3, high (0.4 nm/s) and low (0.15 nm/s). The *I–V* curve for a low deposition rate device was resistive with an impedance of about 25 kΩ. It was observed that all of the devices fabricated with a slow deposition rate (<0.2 nm/s) acted as electrical resistive shorts with impedances of several kΩ, while devices fabricated with fast Alq_3 growth rates (>0.5 nm/s) luminesced with a turn-on voltages of 5–18 V. Based on the images of surface

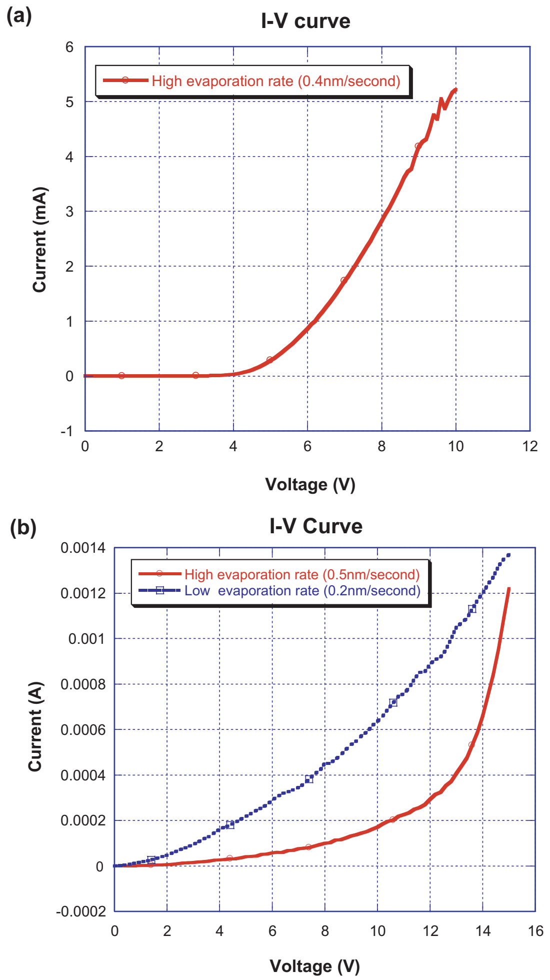

Fig. 4. (a) *I–V* curve for the ITO/TPD/Alq_3/Al diode fabricated with high growth rate for Alq_3 layer with turn-on voltage around 5 V. (b) *I–V* curve for the ITO/TPD/Alq_3/Al diode fabricated with high and low growth rate for Alq_3 layer.

morphology, the resistive characteristics are due to the many small electrical shorts through the pinholes in the Alq_3 layers. Uniform, dense Alq_3 layer surface deposited at higher rates did not have observable pinholes and formed luminescing devices with low turn-on voltages. The deposition rate of thermally deposited Alq_3 is a critical factor in the performance of Alq_3-based OLEDs due to its effect on film morphology.

4. Conclusion

Organic luminescent diodes formed from ITO/TPD/Alq_3 layers were fabricated with thermal evaporation/sublimation of the organic layers, and sputtering/evaporation of the metal cathodes. The surface morphology of thermal evaporated TPD/Alq_3 double layer showed strong dependence on the growth rate. Higher growth rates (>0.4 nm/s) produced uniform, pinhole-free films than low (<0.2 nm/s) growth rates, and is more favorable for the illumination of organic luminescent diode. The exact mechanism for pinhole formation and how it is influenced by deposition is unclear. The best devices fabricated had turn-on voltages as low as 5 V.

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