



Effect of the thermal evaporation rate of Al cathodes on organic light emitting diodes



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ABSTRACT

The relationship between the thermal evaporation rate of Al cathodes and the device performance of organic light-emitting diodes (OLEDs) was investigated to clarify the source of leakage current. Time-of-flight secondary ion mass spectrometry was applied to identify the diffusion of Li and Al fragments into the underlying organic layer during the thermal evaporation process. We prepared various OLEDs by varying the evaporation rates of the Al cathode to investigate different device performance. Interestingly, the leakage current level decreased when the evaporation rate reached $\sim 25 \text{ \AA/s}$. In contrast, the best efficiency and operational lifetime was obtained when the evaporation rate was 5 \AA/s .

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1. Introduction

Since Tang et al. [1] reported organic light-emitting diodes (OLEDs) as the first thin film devices based on small molecular organic materials, great progress has been made, such as high efficiency, fairly low driving voltage, very low power consumption, significantly long lifetime, and so on, enabling OLEDs to be applied to commercial products [2–10].

The lowering of the electron injection barrier from the cathode side has been one of the important developments contributing to such great device performance [11]. The most common cathode system being utilized for mass production in the industry is Mg:Ag (magnesium doped with silver) [12]. However, due to its high reactivity toward air (or moisture) and high sheet resistance, both of which are not appropriate for large area display fabrication, this cathode system requires perfect encapsulation [13,14]. Thus, the cathode system based on a thick aluminum (Al) layer combined with lithium fluoride (LiF) as an electron injection layer is still very important because of its basic ability to realize dramatically improved quantum efficiency, stability, color purity, and so on [15,16]. In particular, the electron injection process, which has a strong influence on device performance, could be enhanced by controlling the thickness of the LiF layer [17].

Meanwhile, Wang et al. [18] reported that LiF could act as a diffusion barrier toward Ag metal fragments during successive evaporation, although it is utilized as a very thin layer (5 Å). However, if the LiF layer cannot be sufficiently protected from Al cathode diffusion during thermal deposition, some Al atoms could penetrate into the underlying organic layers in the LiF/Al cathode system. Nevertheless, the use of the Al cathode is inevitable because it dissociates LiF to Li and AlF₃. Consequently, the diffusion of Li into organic materials in this process could improve the electron injection into the electron transport layer (ETL), as this process may result in n-doping of the underlying layer ($3\text{LiF} + \text{Al} \rightarrow 3\text{Li} + \text{AlF}_3$) [19,20].

Besides, to realize appropriate and reliable device performance, control of the leakage current level is very important because device performance could promptly be degraded when the leakage current level is remarkably high. There have been many reports proposing methods to reduce such an unfavorable factor. The most common method to minimize the leakage current level is to get rid of all the possible particles, which are known to be one of the most probable sources of such a failure mode, in the substrate [21]. A second method could be controlling the roughness of the underlying layers (e.g., ITO, certain organic layers, etc.) utilized during the building of an OLED device structure [22,23]. The third important method associated with cathode formation is reducing the penetration of the electrode materials during the evaporation or sputter process, and so on [24]. This specific issue is particularly critical for achieving reasonable device performance, because such a kind of degradation could also cause imperfect carrier injection or transportation.

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Thus, we investigated the relationship between such metal diffusion and device characteristics of the fabricated OLEDs.

2. Experimental

To fabricate OLED devices, 150 nm-thick patterned indium tin oxide (ITO) substrates with active areas of 4 mm² polyimide insulator were cleaned by sonication in isopropyl alcohol and acetone, rinsed in deionized water, and finally, treated in a UV-ozone chamber. All organic materials were deposited by the vacuum evaporation technique under the pressure of $\sim 1 \times 10^{-7}$ Torr. The deposition rate of organic layers was about 0.5 Å/s. The deposition rate of LiF was about 0.15 Å/s. Specifically, we varied the deposition rates of Al (1, 5, 10, 15, 20 and 25 Å/s) to investigate the different behaviors of the fabricated devices.

The current density–voltage (J – V) and luminance–voltage (L – V) data of OLEDs were measured by Keithley 2635 and Minolta CS-100A, respectively. The electroluminescence (EL) spectra and CIE coordinates were obtained using a Minolta CS-2000A spectroradiometer. The lifetime was measured using OLA300-007 (S-FAC).

The depth profile was obtained using time-of-flight secondary ion mass spectrometry (TOF-SIMS) with a TOF.SIMS 5 instrument prepared by ION-TOF. Bismuth ions (Bi⁺) were used as primary ions with a kinetic energy of 25 keV and a current of 1.0 pA for the detection. The analyzed area was 50 μm × 50 μm. For the evaluation of the depth profile, a sputtering was performed using cesium ions (Cs⁺) of 3 keV and 33.0 nA.

Except TOF-SIMS, all measurements were made at room temperature under ambient conditions with encapsulation.

N,N'-di-[(1-naphthyl)-*N,N'*-diphenyl]-1,1'-biphenyl)-4,4'-diamine (NPB) as a hole-transporting material, Tris-(8-hydroxyquinoline) aluminum (Alq₃) as an electron-transporting and light-emitting material, LiF as an electron injection layer and Al as a cathode were purchased from commercial suppliers and used without purification.

3. Results and discussion

Before investigating the device characteristics, we fabricated a specimen having the structure of a glass substrate/NPB (50 nm)/Alq₃ (50 nm)/LiF (1 nm)/Al (100 nm) to investigate the composition of organic multi-layers after the deposition of Al metals (evaporation rate: 1, 10, and 25 Å/s) by TOF-SIMS analysis. Fig. 1 shows the SIMS depth profile of the sample with a thick Al layer deposited by thermal evaporation. Since both NPB and Alq₃ layers were composed of carbon elements, we determined the mid-points of the rising and falling edges of carbon ion (C⁺) intensity as the interface of the metal/organic layers and that of the organic layers/glass substrate. Also, we assumed that the interface between organic layers (Alq₃/NPB) was located at the mid-point of the steady C⁺ intensity region, because the thickness of each organic layer was exactly the same. The long tails of aluminum ion (Al⁺) and lithium ion (Li⁺) signals were detected in the whole range of the organic layers from the Alq₃ layer through to the NPB layer. Unexpectedly, the samples prepared at different Al deposition rates showed a very similar SIMS depth profile. In other words, Al as well as Li was substantially diffused throughout all of the organic layers in the OLED devices regardless of the evaporation rate of the Al metal.

To investigate the role of metal diffusion, we prepared several devices fabricated with various evaporation rates of the Al cathode, because the evaporation rates could influence the device performance because they could affect the size of the metal fragments penetrated into the underlying organic layers, although the depth profiles are very similar regardless of the evaporation condition.

The configuration of the OLEDs fabricated by varying evaporation rates of the Al cathode (1, 5, 10, 15, 20, and 25 Å/s) were as follows:

Device A: ITO/NPB (50 nm)/Alq₃ (50 nm)/LiF (1 nm)/Al (100 nm, 1 Å/s)

Device B: ITO/NPB (50 nm)/Alq₃ (50 nm)/LiF (1 nm)/Al (100 nm, 5 Å/s)

Device C: ITO/NPB (50 nm)/Alq₃ (50 nm)/LiF (1 nm)/Al (100 nm, 10 Å/s)

Device D: ITO/NPB (50 nm)/Alq₃ (50 nm)/LiF (1 nm)/Al (100 nm, 15 Å/s)

Device E: ITO/NPB (50 nm)/Alq₃ (50 nm)/LiF (1 nm)/Al (100 nm, 20 Å/s)

Device F: ITO/NPB (50 nm)/Alq₃ (50 nm)/LiF (1 nm)/Al (100 nm, 25 Å/s).

Fig. 2 shows the J – V – L characteristics of the fabricated OLEDs, and the representative results are summarized in Table 1. At a given constant voltage of 5.0 V, current density values of 22.7, 13.5, 14.9, 16.6, 16.2, and 10.7 mA/cm² were observed from the fabricated Devices A, B, C, D, E, and F, respectively. Interestingly, the absolute values of the leakage current density level at -2 V were observed to be diverse as follows: 4.1×10^{-2} , 1.0×10^{-3} , 7.4×10^{-4} , 6.8×10^{-4} , 4.5×10^{-4} , and 1.7×10^{-4} mA/cm² for Devices A, B, C, D, E, and F, respectively. Similar behavior was also observed at the lower operating voltage region below the turn-on voltage, where we assumed that those kinds of leakage current levels could originate from the trapping sites by the diffusion of the Li or Al fragments into the underlying organic layers detected by TOF-SIMS. As a result, the leakage current level decreased when the evaporation rate of the Al cathode increased to ~ 25 Å/s. This means that the higher evaporation rate could result in an extremely low leakage current level originating from the different diffusion behavior of Li or Al metal fragments. Conversely, the devices obtained from the very slow evaporation rate (1 Å/s) showed a very high leakage current level. Meanwhile, Devices B through E (5–20 Å/s), which were fabricated at the medium evaporation rates, caused middle levels of leakage current, which were very similar and in between 1.0×10^{-3} and 4.5×10^{-4} mA/cm². This tendency is very similar to the one obtained in a conventional electron beam evaporation system. In other words, the average grain size increased monotonically with the increase in deposition rate, while the films consisting of small grains with almost uniform size were obtained at low deposition rates, as reported by Bordo [25]. Therefore, when deposited at a very low evaporation rate, the size of metal fragments or particles diffused into the underlying organic layers could be very small and uniformly distributed. For this reason, we expected that the device fabricated in the lower evaporation rate would result in larger levels of leakage current. Meanwhile, the driving voltages to reach 1000 cd/m² were 5.7, 5.5, 5.5, 5.6, 5.7, and 6.0 V for Devices A, B, C, D, E, and F, respectively (Table 1). To understand this behavior, we fabricated electron only devices (EODs) as follows:

EOD 1: ITO/Alq₃ (150 nm)/LiF (1 nm)/Al (100 nm, 1 Å/s)

EOD 2: ITO/Alq₃ (150 nm)/LiF (1 nm)/Al (100 nm, 10 Å/s)

EOD 3: ITO/Alq₃ (150 nm)/LiF (1 nm)/Al (100 nm, 25 Å/s)

We chose only three evaporation conditions as above because Devices B–E showed very similar leakage current levels. Fig. 3 shows the electron current characteristics of EODs prepared with the evaporation rate of 1 Å/s (EOD 1), 10 Å/s (EOD 2), and 25 Å/s (EOD 3), respectively. In principle, the increase of applied voltage may increase the density of free carriers resulting from injection to such a value that the quasi-Fermi level moves up over the shallow electron-trapping level, and fills up most traps. The traps-filled limit (TFL) is the condition for the transition from the trapped J – V

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