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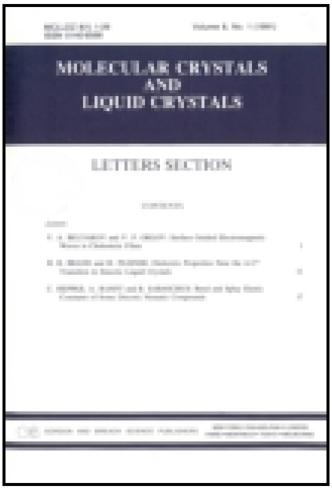

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Close-Spaced Sublimation of Organic Materials for Organic Light-Emitting Devices

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Close-Spaced Sublimation of Organic Materials for Organic Light-Emitting Devices

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We have demonstrated organic light-emitting devices (OLEDs) fabricated by using a close-spaced sublimation (CSS) method. We have deposited tris(8-hydroxyquinolinato)aluminum (Alq_3) layer using a CSS process for the formation of electron transport layer of the PVK/Alq_3 bilayer OLEDs. The distance between the sublimation source and substrate was fixed to be less than 5 mm. The substrate temperature was maintained less than 70°C while the sublimation source temperature was varied from 185 to 200°C during the deposition of Alq_3 film. We investigated the photoluminescence characteristic of the CSS processed Alq_3 film. We also investigated the emission and electro-optical properties of the PVK/Alq_3 bilayer OLEDs with CSS deposited Alq_3 film.

Keywords Organic light-emitting diode; close-spaced sublimation

Introduction

Since the first report on double-layered organic light-emitting devices (OLEDs) more than two decades ago by C. W. Tang and S. A. Vanslyke, these devices have attracted much attention because of their many advantages, such as fast response time, wide viewing angle, and thin thickness for use in full color displays and solid-state lighting applications [1]. Especially, the external quantum efficiency has been substantially enhanced by using phosphorescent organic emitters which can harvest both the singlet and triplets excitons generated by the recombination of holes and electrons [2]. The OLEDs are typically fabricated by using a thermal vacuum evaporation or solution coating methods [1, 3]. The high efficiency and long lifetime devices have been generally obtained by the thermal vacuum evaporation method which can provide multiple functional organic layers that improve the carrier injection, recombination efficiency, confinement of excitons within the emission layer, and driving characteristics. However, this method requires high vacuum system, long deposition time, complicated fabrication procedures, expensive equipment, and low efficiency of utilization of organic materials. The solution coating methods have also been widely used for the fabrication of OLEDs because of their simplicity and large area compatibility. However, the solution coated method have a difficulty in preparing

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multiple layers of organic materials because the typical organic materials are soluble in common solvents such as toluene and chlorobenzene.

In this paper, we demonstrate a close-spaced sublimation (CSS) method for depositing organic layers of OLEDs. The CSS process has been used to fabricate semiconductor solar cells [4]. The substrate and sublimation source are closely spaced (typically less than 2 mm) and the low vacuum (typically higher than 10^{-3} Torr) is used in the CSS process. Since the distance between the sublimation source and the target substrate is very small in the CSS process, the consumption of organic materials can be reduced in fabricating OLEDs. In addition, the CSS process doesn't need the complicated fabrication system because the low vacuum is required to deposit the functional layers. In this paper, we used the CSS process for depositing functional organic layers of bilayer OLEDs. We investigated the effect of sublimation source temperature on the characteristics of CSS deposited tris(8-hydroxyquinolinato)aluminum (Alq_3) layers. We also investigated the emission and current efficiency characteristics of the OLEDs fabricated by the CSS process.

Experimental

Figure 1 shows the schematic diagram of the CSS system and process. The CSS system is composed of the low vacuum chamber, low vacuum pump, substrate holder, substrate, sublimation source loader, IR lamp, and quartz window. At first, the organic source substrate is prepared by spin coating of the mixed solution on the bare glass substrate. The mixed solution is fabricated by mixing organic material in a common solvent such as chlorobenzene. The organic source substrate and the target substrate are loaded to the sublimation source holder and substrate holder, respectively. After then, the organic source substrate is heated by IR lamp through the quartz window. The IR lamp is located outside the low vacuum chamber. The nitrogen gas is used to purge the chamber. The thermocouple on the target substrate is used to measure the temperature of the target substrate during the sublimation of organic material.

For the fabrication of OLEDs by the CSS of organic material, ITO coated glass substrates were prepared. The sheet resistance and thickness of ITO film were about 13 ohm/square and 150 nm, respectively. After defining ITO anode patterns using a standard photolithography process, the patterned ITO substrates were cleaned with acetone, isopropyl alcohol, and methanol, and rinsed with deionized water. After then, a 60 nm thick poly(9-vinylcarbazole) (PVK) layer was formed on the ITO substrate by spin coating of the PVK solution solved in chlorobenzene solvent. The PVK layer acts as a hole injection and transport layer. After loading the PVK coated substrate into the CSS chamber, a 70 nm thick Alq_3 layer was coated by close-spaced sublimation. The Alq_3 layer acts as an electron transporting and emitting layer. We also coated the Alq_3 layer by vacuum thermal evaporation at a base pressure of about 1×10^{-6} Torr for comparing with the CSS coated layer. After the formation of organic layers, a 0.5 nm thick LiF layer and a 100 nm thick Al layer were sequentially evaporated through a shadow mask. The active area of the device was 4 mm × 4 mm. Current density-voltage-luminance characteristics the OLEDs were measured using computer controlled Keithley 2400 source-measure unit and a calibrated photodiode. The photoluminescence (PL) and electroluminescence (EL) spectra of organic materials and devices were measured with a spectroradiometer (Minolta CS1000).

Results and Discussion

The organic source material is closely spaced to the substrate in the CSS process so that the organic material consumption can be very small compared to the vacuum thermal

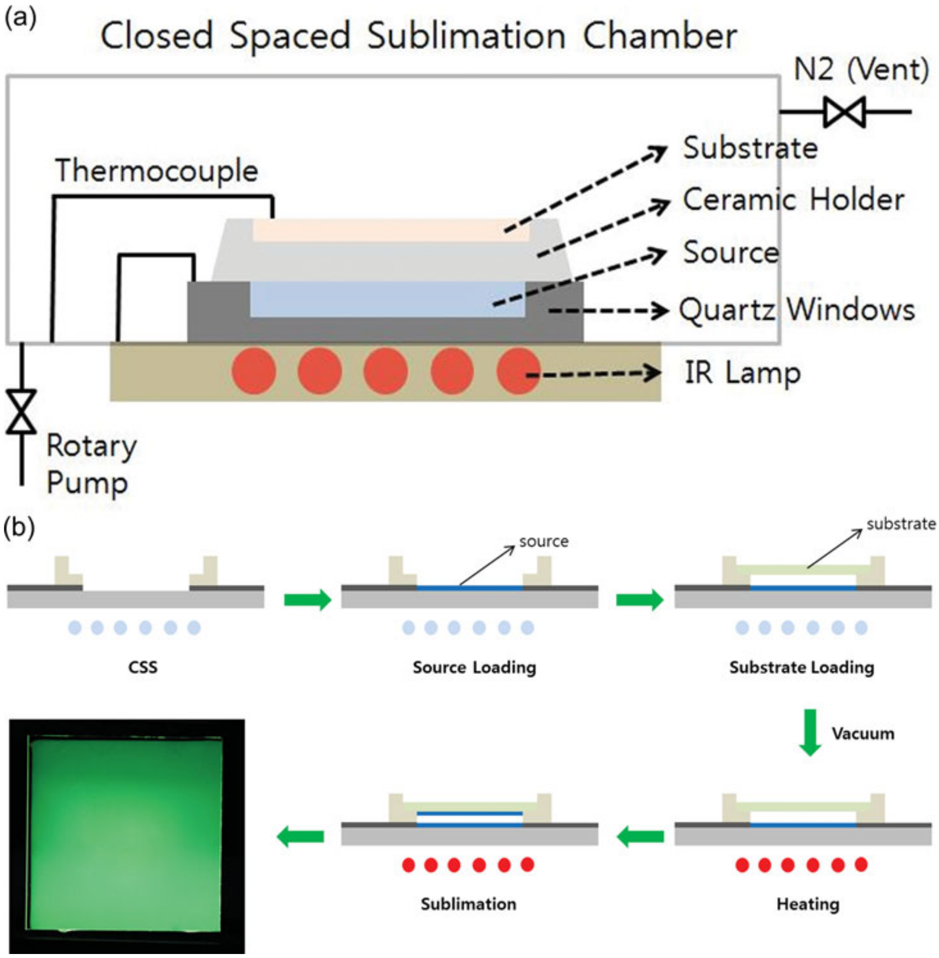

Figure 1. Schematic views of the CSS system and process.

evaporation process where the distance between source and substrate is larger than 10 cm. The material efficiency of CSS process was investigated by comparing the weight of deposited organic layer with that of the source organic material. Alq_3 was used as an organic material for measuring the material efficiency of CSS process. For depositing the Alq_3 layer with a weight of 0.9 mg at a sublimation temperature of 210°C, the source material of 1.0 mg was consumed so that the material efficiency of CSS process was measured to be about 90%. In the typical vacuum thermal evaporation process equipped with point organic sources, the material efficiency is typically less than 5%. The material efficiency is also less than 50% in the evaporation process with linear organic sources. The material efficiency is extremely high in the CSS process.

Figure 2 shows the substrate temperatures as a function of sublimation time for various source temperatures. Alq_3 molecules were used as an organic source material. The IR lamp was used for heating organic source material. The lamp was turned on for 5 min. for sublimation of Alq_3 molecules. In our CSS process the graphite source holder absorbs the IR light from the lamp and reaches to the source temperature within several seconds, whereas the substrate temperature increases with increasing sublimation time up to about 300–400 sec.

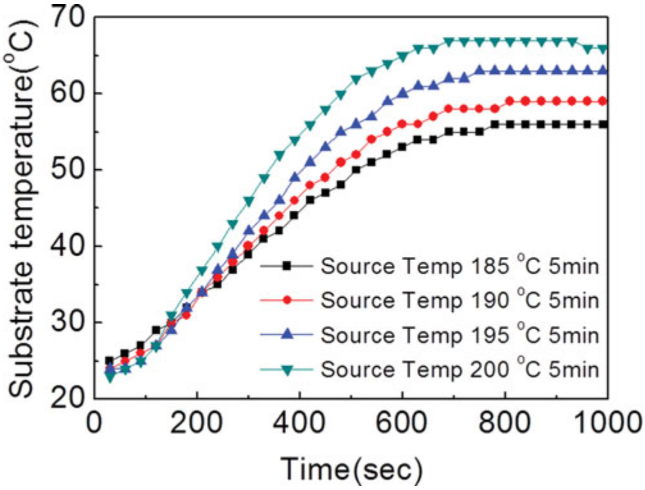

Figure 2. Substrate temperatures as a function of sublimation time for various source temperatures.

and then saturates. The rise of substrate temperature is attributed to the heat transfer from the graphite source holder. Therefore, the maximum substrate temperature increases as the source temperature increases. For example, the maximum substrate temperature increases from 57 to 66°C as the source temperature increases from 185 to 200°C. The rise of the substrate temperature at high source temperature may affect to the material characteristics of deposited Alq_3 layer. In order to investigate the material property of the Alq_3 layer deposited by CSS process, PL spectrum of the CSS deposited Alq_3 layer was measured. Figure 3 shows the PL spectrum of the Alq_3 layer deposited by the CSS process. Although the emission intensity is weak because the thickness of the layer is thin (less than 100 nm), the emission peak is observed at 520 nm, which is consistent with the reported value. Hence, it indicates that the emission property is not modified by the rise of substrate temperature during the close-spaced sublimation of Alq_3 molecules. Therefore, the result suggests that the material properties of Alq_3 may not be changed since the substrate temperature is much less than the glass transition temperature of Alq_3 [5].

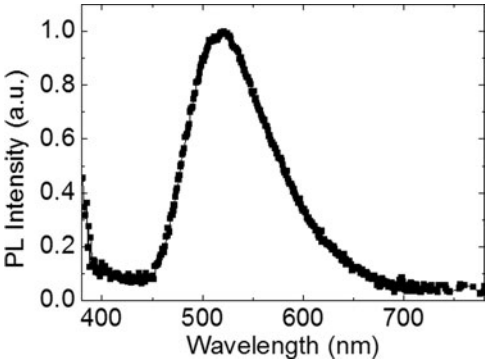

Figure 3. PL spectrum of the Alq_3 layer deposited by the CSS process.

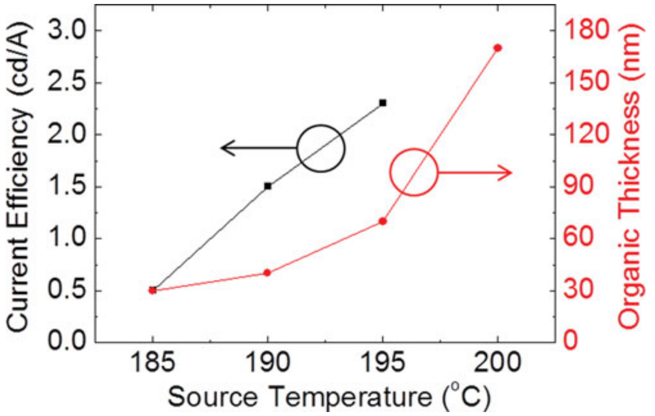

Figure 4. Thickness of the Alq_3 layer and the current efficiency of the bilayer OLEDs as a function of CSS source temperature.

Figure 4 shows the thickness of the Alq_3 layer and the current efficiency of the bilayer OLEDs as a function of CSS source temperature. The turn on time of IR lamp was fixed to be 5 min. The device structure was ITO (150 nm)/PVK (60 nm)/Alq_3 (30–180 nm)/LiF (0.5 nm)/Al (100 nm). The PVK layer was formed by spin coating of the PVK solution solved in the chlorobenzene. The thicknesses of the Alq_3 layer were about 30 nm at a source temperature of 185°C. As the source temperature increases to 195°C, the thickness increases to 70 nm. The thickness of Alq_3 layer abruptly increases to 170 nm as the source temperature increases to 200°C. The current efficiency of the device is 0.5 cd/A at a source temperature of 185°C. This low current efficiency is attributed to the low recombination efficiency since the electrons and holes are not balanced when the Alq_3 layer is thin. As the Alq_3 thickness increases to 70 nm at a source temperature of 195°C, the current efficiency increases to 2.3 cd/A. These results also indicates that the PVK and Alq_3 layers are not damaged because the substrate temperature is maintained less than 70°C during the close-spaced sublimation of organic materials, as shown in figure 2. The substrate temperature is much less the glass transition temperatures of PVK and Alq_3 [6].

Figure 5 shows the current density-voltage curves of the OLEDs fabricated by the CSS and vacuum thermal evaporation methods. The device structure is ITO/PVK (60 nm)/Alq_3 (60–70 nm)/LiF/Al. The driving voltage is slightly high in the PVK/Alq_3 architecture since the energy barrier for hole injection is high. The highest occupied molecular orbital (HOMO) and the lowest molecular orbital (LUMO) energy levels of PVK are 5.5 and 2.0 eV, respectively [7], as shown in Figure 6. The barrier for hole injection from ITO to PVK is about 0.8 eV, which is higher than the typical hole transporting materials. The driving voltage of the CSS processed device is higher than the vacuum evaporated one. It might be attributed to the microstructures of Alq3 film although the reason of this difference is not clear. The deposition pressure in the vacuum evaporation process is lower compared to the CSS process, so that the molecules may be easily reoriented to make a dense film in the vacuum evaporation process, resulting in shorter distances between adjacent molecules.

Figure 7 shows the EL spectra for the OLEDs fabricated by the CSS process and vacuum thermal evaporation. The CSS processed and thermally evaporated devices exhibit almost same EL spectra. The EL peak at 530 nm is due to the radiative emission from the Alq_3 molecules. The HOMO and LUMO energy levels of Alq_3 are 5.7 and 3.0 eV, respectively [8]. The energy barrier for hole injection from PVK to Alq_3 is 0.2 eV, as

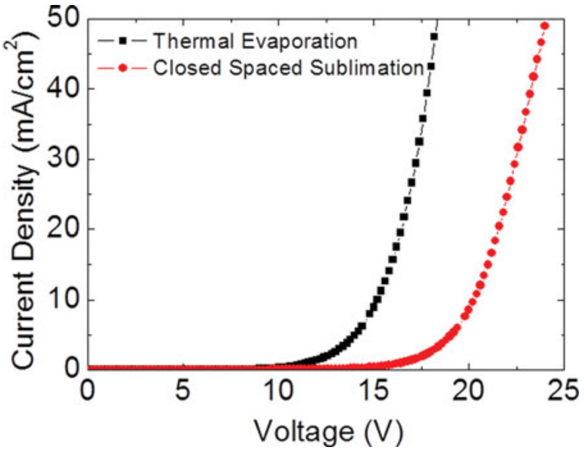

Figure 5. Current density-voltage curves of the OLEDs fabricated by the CSS and vacuum thermal evaporation methods.

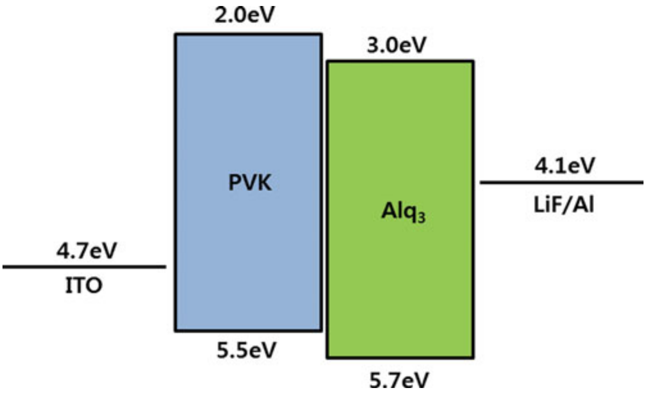

Figure 6. Energy diagram of the PVK/Alq$_3$ bilayer OLED.

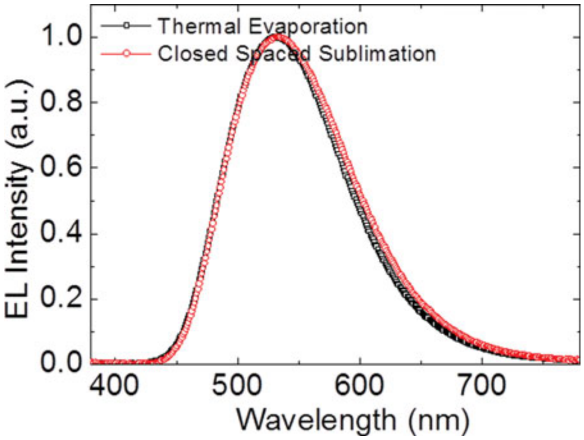

Figure 7. EL spectra for the OLEDs fabricated by the CSS process and vacuum thermal evaporation.

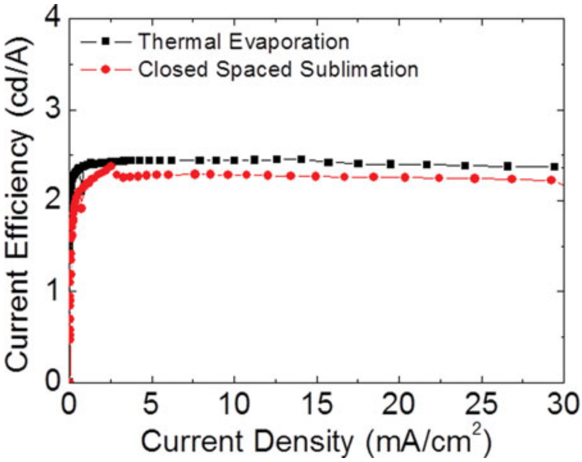

Figure 8. Current efficiency characteristics of the devices fabricated by the CSS process and vacuum thermal evaporation.

shown in the energy diagram of Figure 6. On the other hand, the energy barrier for electron injection from Alq_3 to PVK is 1.0 eV. The electrons should overcome high energy barrier for injecting from Alq_3 to PVK. Therefore, electron-hole recombination takes place in the Alq_3 layer adjacent to the PVK so that the EL emission from PVK is not observed in the Figure 7.

Figure 8 shows the current efficiency characteristics of the devices fabricated by the CSS process and vacuum thermal evaporation. The maximum current efficiency of the CSS processed device is 2.4 cd/A. On the other hand, the maximum current efficiency of the thermally evaporated device is 2.5 cd/A. Hence, both devices exhibit similar current efficiency, indicating that the CSS process can be a good candidate in fabricating OLEDs.

Conclusions

We have investigated the Alq_3 layer and PVK/Alq_3 bilayer devices deposited by the CSS process. The organic material consumption in the CSS process was very small. The material efficiency of the CSS process was about 90%. The substrate temperature was maintained less than 70°C when the organic materials were sublimed at source temperature of 180–200°C. There was no difference in EL spectrum between the CSS processed and thermally evaporated devices. The maximum current efficiency of the CSS processed device was also similar with the vacuum evaporated device. Therefore, the CSS process can be a strong candidate in fabricating OLEDs.

Funding

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