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Micromesh-structured flexible polymer white organic light-emitting diodes using single emissive layer of blended polymer and quantum dots

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ABSTRACT

In this study, poly(N-vinylcarbazole) was blended with CdSe/ZnS core-shell quantum dots of various dimensions to be used as a single emissive layer of flexible polymer white organic light-emitting diodes (WOLEDs). A structure of WOLEDs was deposited on polyethylene terephthalate (PET) substrates. A luminance and maximum luminous efficiency of 4070.2 cd/m² and 2.84 cd/A were respectively obtained for the WOLEDs prepared using n-type gallium zinc oxide (GZO) and n-type indium gallium zinc oxide (IGZO) films as the electron transport layer and cathode, respectively. Because the current density can be increased by increasing the carrier transport surface area, the 2-μm-periodic SiO₂ micromesh was fabricated on PET substrates to enhance the surface area of a MoO₃ hole transport layer and a GZO electron transport layer. Furthermore, the GZO electron transport layer could provide a moisture barrier function. Compared with WOLEDs without the SiO₂ micromesh, the prepared WOLEDs had an increased current density of 404.5 (compared with 222.8) mA/cm² when operating at 11 V. Moreover, their associated luminance and maximum luminous efficiency increased to 7016.1 cd/m² and 3.04 cd/A, respectively.

1. Introduction

During the past half century, various inorganic materials and structures have successfully been used to fabricate inorganic light-emitting diodes (LEDs) and widely applied to various modules and systems. Recently, organic LEDs (OLEDs) have received considerable attention because of their inherent advantages, including wide viewing angle, self-emitting property, low cost, simple fabrication process, large-scale production, and flexibility [1,2]. Single-colored OLEDs are widely used in various applications; however, white OLEDs (WOLEDs) are urgently required for applications in solid-state lighting fields, full-color flat-panel display system, and optical biosensors [3–5]. Resonant cavities, single emissive layers, and tandem-stacked layers are commonly developed to fabricate WOLEDs [6–8]. For simplifying fabrication, the single

inherent advantages of quantum confinement for improving performance, various QDs have been widely used in LEDs [9–11]. In this work, the single emissive layer in flexible polymer WOLEDs was developed by blending poly(N-vinylcarbazole) (PVK) with various dimensional CdSe/ZnS core-shell QDs. Moreover, regarding the promising transparent ZnO-based materials used in transparent thin-film transistors and solar cells [12,13], n-type gallium zinc oxide (GZO) and n-type indium gallium zinc oxide (IGZO) films were used as an electron transport layer and a cathode electrode of flexible polymer WOLEDs, respectively. Furthermore, to fabricate a moisture barrier layer and to roughen the surface of a GZO electron transport layer, a 2-μm-periodic SiO₂ micromesh was formed on flexible polyethylene terephthalate (PET) substrates by using a two-beam He–Cd laser interference photolithography system.

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emissive layer is a promising structure. However, to obtain white-light emission, various mixed organic materials are required in the single emissive layer. In addition to high color rendering, the emission of various colors of light can be achieved by quantum dots (QDs) with different dimensions. By considering the

2. Experiment

Fig. 1(a) and (b) illustrates the schematic configuration of flexible polymer WOLEDs without and with the 2-μm-periodic SiO₂ micromesh, respectively. In

a structure, 35-nm-thick n-type GZO layer, 65-nm-thick blended PVK with ODs (QDs:PVK) layer, and 10-nm-thick p-type

Fig. 1. Flexible polymer WOLEDs (a) without and (b) with the 2- μm -periodic SiO_2 micromesh.

molybdenum oxide (MoO_3) layer were used as electron transport, single emissive, and hole transport layers, respectively. In addition, 300-nm-thick IGZO and 100-nm-thick gold (Au) layers were employed as a cathode and an anode, respectively. Furthermore, the SiO_2 micromesh [Fig. 1 (b)] was used as the moisture barrier layer, and the surface roughness of the GZO electron transport layer was enhanced. A 500-nm-thick SiO_2 film was first deposited on PET substrates by using an electron beam evaporator. Subsequently, a pattern of 2- μm -periodic SiO_2 micromesh was obtained using a two-beam He–Cd laser interference photolithography system in accordance with the process reported in Ref. [14]. The period of the SiO_2 micromesh was obtained by changing the intersection angle of the two-beam He–Cd lasers. An etched 30-nm-thick SiO_2 micromesh was obtained using a formed mask of a photoresist (AZ 6112) micromesh pattern by employing a chemical solution of buffered-oxide etch. Because the resulting SiO_2 micromesh caused roughening of the SiO_2 surface, the surface of subsequently deposited IGZO and GZO films was roughened to increase the carrier-injection surface area of the GZO electron transport layer. Fig. 2 (a) displays the top view of the 2- μm -periodic SiO_2 micromesh obtained using a scanning electron microscope. Afterward, a 300-nm-thick IGZO and a 35-nm-thick GZO films were respectively deposited on the PET substrates and the SiO_2 micromesh using a radio frequency (RF) magnetron sputter, sequentially. For fabricating the QDs:PVK single emissive layer, a dispersion solution of toluene, blended PVK, and various dimensional CdSe/ZnS core-shell QDs was first prepared. The dispersion solution was then stirred for 1 day and sonicated for 1 h. The amounts of toluene, PVK, and total CdSe/ZnS core-shell QDs in the dispersion solution were 1 mL, 5 mg, and 25 mg, respectively (the weight ratio for QDs with a dimension of 5.2 nm, 4.2 nm, 1.6 nm was 1.5:1.3:2.2). The 65-nm-thick QDs:PVK single emissive layers were spread on GZO layers by employing the prepared dispersion solution and annealed in a glove box at 90 °C for 30 min. Subsequently, a hole transport layer of a 10-nm-

thick MoO_3 film and an anode of a 100-nm-thick Au film were sequentially deposited on the QDs:PVK

Fig. 2. Scanning electron microscope image of top view of 2- μm -periodic SiO_2 micromesh.

single emissive layer by using a thermal evaporator. To study the electrical properties of 35-nm-thick GZO and 300-nm-thick IGZO films, they were deposited on PET substrates through RF magnetron sputter deposition.

3. Results and discussion The electron concentration of $7.86 \times 10^{20} \text{ cm}^{-3}$ and $1.07 \times 10^{21} \text{ cm}^{-3}$ and the electron mobility of $4.20 \text{ cm}^2/\text{V}\cdot\text{s}$ and $9.96 \text{ cm}^2/\text{V}\cdot\text{s}$ were respectively obtained for 35-nm-thick GZO and 300-nm-thick IGZO films by using Hall measurement, and their associated resistivities were $1.67 \times 10^{-3} \Omega\cdot\text{cm}$ and $5.08 \times 10^{-4} \Omega\cdot\text{cm}$, respectively.

Fig. 3 presents the photoluminescence spectra of PVK and CdSe/ZnS core-shell QDs that were obtained using the He–Cd laser as an excited source and normalized with respect to a green emissive intensity by employing 5 mg of each used material. The emission peaks of QDs with a dimension of 1.6 nm, 4.2 nm, 5.2 nm appeared at 451 (blue color), 523 (green color), and 611 nm (red color), respectively. The white emission could be obtained by varying the content of various dimensional CdSe/ZnS QDs. By using the weight ratio of 1.5:1.3:2.2 for the RGB emissive quantum dots, the Commission Internationale de l’Eclairage (CIE) of (0.33, 0.33) was obtained for both the WOLEDs without and with the 2- μm -periodic SiO_2 micromesh (Fig. 4). Its associated color rendering index (CRI) was 73.2. The SiO_2 micromesh and roughened surface of the GZO electron transport layers did not influence white emission.

Fig. 3. Normalized photoluminescence spectra of PVK and CdSe/ZnS core-shell QDs with dimension of 5.2, 4.2, and 1.6 nm.

Fig. 4. CIE coordinate of WOLEDs without and with the 2- μ m-periodic SiO₂ micromesh.

Fig. 5 presents current density as a function of voltage applied to WOLEDs without and with the 2.0- μ m-periodic SiO₂ micromesh. Current density was increased in WOLEDs with SiO₂ micromesh. The current density of WOLEDs without and with the 2- μ m-periodic SiO₂ micromesh at an applied voltage of 11 V were 222.8 and 404.5 mA/cm², respectively. The surface area of MoO₃ hole transport layer and GZO electron transport layer roughened by the SiO₂ micromesh was larger than that of the flat MoO₃ hole transport layer and flat GZO electron transport layer. Consequently, the increase in current density was attributed to the larger carrier injection surface area. The inset of Fig. 5 illustrates the dependence of luminance on the applied voltage. When the applied voltage was 11 V, the luminance of 4070.2 and 7016.1 cd/m² was obtained for WOLEDs without and with the 2- μ m-periodic SiO₂ micromesh, respectively. The luminance of WOLEDs with the SiO₂ micromesh was greater than that of WOLEDs without the SiO₂ micromesh for the same applied voltage. Moreover, regarding to the low carrier transportation velocity in the QDs:PVK single emissive layer owing to the low carrier mobility, the carrier recombination opportunity would be increased by shortening the propagation distance caused by the surface roughness of both the surfaces. It was deduced that the increased carrier recombination opportunity would increase its emissive

luminance. Consequently, the phenomenon of improve luminance was attributed to a larger injection current density and a larger carrier

Fig. 5. Current density as a function of voltage applied to WOLEDs without and with the 2- μ m-periodic SiO₂ micromesh. The inset shows associated dependence of luminance on applied voltage.

recombination opportunity obtained in the WOLEDs with the SiO₂ micromesh for the same applied voltage. Luminance gradually saturated at a higher applied voltage.

Fig. 6 presents luminous efficiency as a function of voltage applied to WOLEDs without and with the SiO₂ micromesh. At the same applied voltage, the WOLEDs with the SiO₂ micromesh had higher luminous efficiency than the WOLEDs without the SiO₂ micromesh. This phenomenon was attributed to the larger luminance of WOLEDs with the SiO₂ micromesh. In both WOLEDs, the luminous efficiency increased with an increase in the applied voltage until the voltage corresponded to the peak luminous efficiency, and then its luminous efficiency decreased with a further increase of apply voltage. Because the injection current density increased with an increase in the applied voltage, a larger current density led to a higher hole-electron recombination rate via QDs, which enhanced luminance and the associated luminance efficiency. However, because the total number of QDs within the QDs:PVK single emissive layer was kept constant, it was deduced that the injection current density was restricted for obtaining effective hole-electron recombination via QDs. Consequently, the luminance of WOLEDs was gradually saturated by applying a higher injection current density caused by a higher apply voltage (as shown in the inset of Fig. 5). The luminous efficiency gradually decreased with an increase in the applied voltage beyond the voltage corresponding to the maximum luminous efficiency. For WOLEDs without and with the 2- μ m-periodic SiO₂ micromesh, the maximum luminous efficiency of 2.84 and 3.04 cd/A were respectively obtained at the applied voltage of 8.1 and 7.8 V. The applied voltage corresponding to the maximum luminous efficiency of WOLEDs with the 2- μ m-periodic SiO₂ micromesh was lower than that of WOLEDs without the SiO₂ micromesh. This phenomenon was attributed to the fact that the injection current density required for saturating effective hole-electron recombination via QDs was obtained by applying a lower voltage to the WOLEDs with the 2- μ m-periodic SiO₂ micromesh.

4. Conclusions

In this work, a single emissive layer constructed by blending PVK with various dimensional CdSe/ZnS core-shell QDs was employed to fabricate flexible polymer WOLEDs on PET substrates. RGB light could be emitted through CdSe/ZnS QDs with the dimensions of 5.2, 4.2, and 1.6 nm, respectively. By using the weight ratio of 1.5:1.3:2.2 for RGB emissive QDs, the CIE coordinate and CRI of (0.33, 0.33) and 73.2 were respectively obtained for WOLEDs. The luminance and the maximum

Fig. 6. Luminous efficiency as a function of voltage applied to WOLEDs without and with the 2- μ m-periodic SiO₂ micromesh.

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The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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