Effect of Charge Generation Barrier Height for Field-Polarity Dependent Color-Tunable Quantum-Dot Light-Emitting Diodes

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ABSTRACT

We demonstrate a hole-only driven color-tunable tandem QD-LED. The device restrains electron injection from the electrodes but generates electrons from the charge generation layer, resulting the field-polarity controlled color emission. We verify the effect of the charge generation barrier which affects the turn-on voltage and the efficiency of the device.

1 INTRODUCTION

Current display devices are composed of red, green, and blue sub-pixels to reproduce a variety of colors. In general, these three sub-pixels are horizontally arranged[1]. However, the horizontal arrangement has the limitation in improving the geometric fill factor, which is essential for ultra-high resolution displays. One of methods to overcome the low geometric fill factor nature is vertical stack of sub-pixels. Recently, our group have reported a unique structure of electron-only tandem QD-LED that can modulate emission color according to the polarity of the applied voltage[2]. The main mechanism of this structure is restraining the hole injection from electrodes and generating holes from charge generation layer (CGL) inside the device.

Here, we demonstrate hole-only color-tunable tandem QD-LEDs that can control the emission color by changing the polarity of the applied voltage. We have used two different hole transport layer (HTL) to verify the relationship between the charge generation barrier height and the device performance.

2 EXPERIMENT

The hole-only color-tunable tandem QD-LED was fabricated in a full solution process except for the electrodes. The device structure is composed of ITO/PEDOT:PSS/HTL/green QDs/ZnO/PEDOT:PSS/ZnO /red QDs/HTL/PEDOT:PSS/Al, as demonstrated in Fig. 1a. Poly-TPD and PVK are used for the HTL because their highest occupied molecular orbital (HOMO) levels are distinguishable. The HOMO level of poly-TPD and PVK are 5.4 and 5.8 eV, respectively, as shown in Fig. 1b and 1c. First, the pre-patterned ITO glasses were sonicated in the acetone and isopropyl alcohol bath sequentially for 10 min each. Then, the substrates were rinsed with deionized water and baked at 200 °C for 5 min to remove residual waters. The O_2-plasma treatment was carried out on the cleaned substrates at 50 W for 60 s. Next, PEDOT:PSS was spin coated on the substrate at 6000 rpm for 30 s and baked at 120 °C for 25 min. Both HTL materials, poly-TPD and PVK, were dissolved in chlorobenzene with the concentration of 1 wt% and the solution was spin coated on the PEDOT:PSS layer at 8000 rpm for 30 s and annealed at 140 °C for 25 min. Green QD (CdSe@ZnS) layer was spin coated on the HTL at 6000 rpm and 30 s and baked at 120 °C for 40 min. ZnO nanoparticles dispersed in 1-buthanol were deposited on the previous layer with the spin rate of 3000 rpm for 30 s and baked at 140 °C for 25 min. The following PEDOT:PSS, ZnO, red QDs (CdSe@ZnS), HTL, PEDOT:PSS layers were processed consecutively in the same procedure as demonstrate above, except that PEDOT:PSS was mixed with ethanol in 3:2 ratio to improve the wettability. On top of multi-stacked layers, aluminum electrode was thermally evaporated in a high vacuum chamber.

3 RESULTS

The current density(J)-voltage(V)-luminance(L) characteristics of the tandem QD-LEDs with different HTLs are demonstrate with regard to the polarity of the applied voltage in Fig. 2a and 2b. Both QD-LEDs emit green light at the positive bias and red light at the negative bias, as demonstrated in the Fig. 2c. The peak wavelength for green emission was ~536 nm and for red emission was ~647 nm. The overall performances of the devices are summarized in Table 1. The poly-TPD device exhibit lower turn on voltage (V_T, voltage at 1 cd/m^2) and higher maximum luminance (L_{max}) but lower external quantum efficiencies (EQE) than the PVK device. The results indicates that difference in the HOMO level of the HTL affects the device performance.

4 DISCUSSION

The color-tunable emission mechanism of the tandem QD-LED is illustrated in Fig. 3. On the positive bias, holes are injected from the ITO electrode to the green QD layer. However, electrons injected from the Al electrode are blocked by the top HTL and recombine with holes generated from the interface between the red QD layer and the top HTL. Electrons generated in this interface are transported to the central PEDOT:PSS layer and recombine with holes generated from the bottom ZnO layer and the central PEDOT:PSS layer. Generated electrons from ZnO and PEDOT:PSS interface are injected to the green QD layer and contribute to the emission. Thus, only green color is emitted on the positive bias. The red emission on the negative bias occurs in the similar mechanism but in the opposite direction moving pathways.

Here, the effect of HTL HOMO level can be considered. In the case of positive bias, the bottom HTL HOMO level determines the hole injection barrier(Δh_1) height to the green QD layer, while the top HTL HOMO level decides the charge generation barrier(Δh_2). The hole injection barrier is the difference between the bottom HTL HOMO level and the valance band maximum of the green QD layer, which is 1.4 and 1.0 eV for poly-TPD and PVK, respectively. The charge generation barrier is the difference between the top HTL HOMO level and the conduction band minimum of the red QD, which is 0.9 and 1.3 eV for poly-TPD and PVK, respectively. Hence, the lower charge generation barrier of poly-TPD resulted the lower V_T[2], but inferior efficiencies because of the higher hole injection barrier[3].

5 CONCLUSIONS

In summary, we have demonstrated a field-polarity dependent hole-only tandem color-tunable QD-LED. Two different HTLs were used in our devices and the features were characterized with positive and negative bias. The carrier injection and transport paths were interpreted to clarify the color-tunability by the polarity of the applied voltage. Furthermore, the effect of charge generation barrier was verified that it affects the threshold voltage of the device. Meanwhile, the charge generation barrier was related to the hole injection barrier which affects the efficiency of the device. Therefore, we can conclude that the charge generation barrier is in trade-off relationship between the turn-on voltage and the efficiency of the device.

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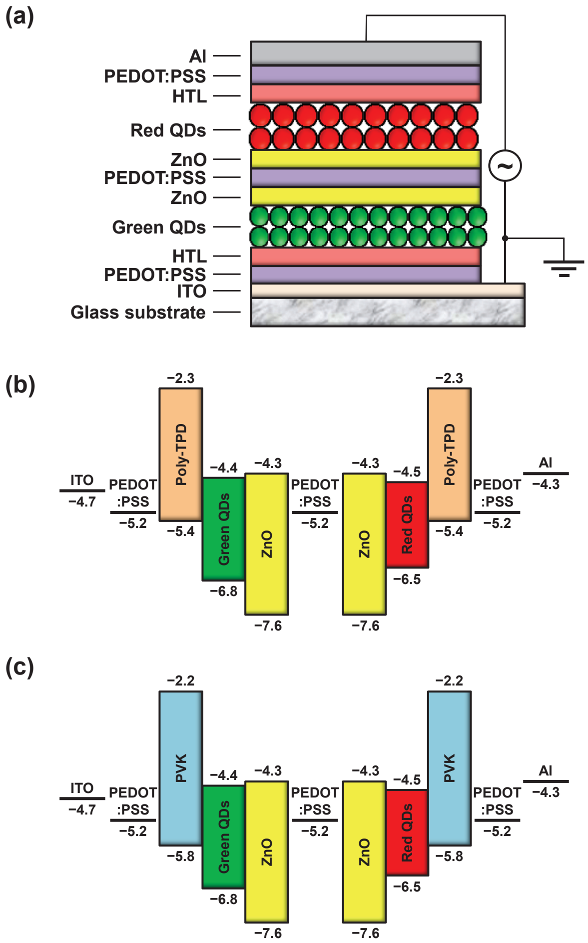

Fig. 1 (a) Schematic of the hole-only tandem QD-LED. (b) Energy level diagram for poly-TPD HTL device. (c) Energy level diagram for PVK HTL device.

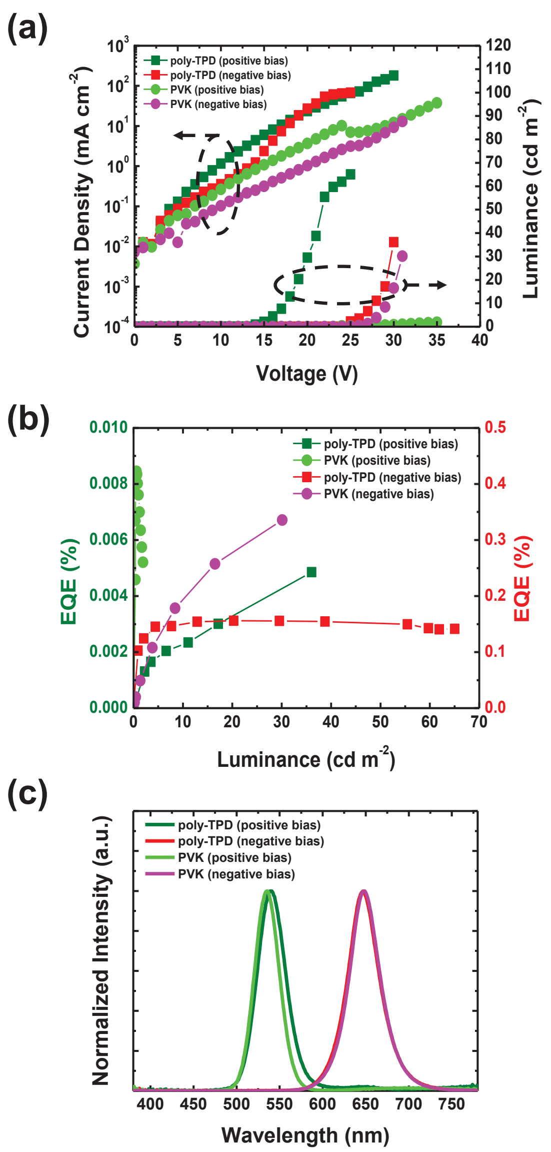

Fig. 2 (a) J-V-L, (b) EQE, and (c) electroluminescence spectra of two different HTL tandem devices under the positive and negative driving.

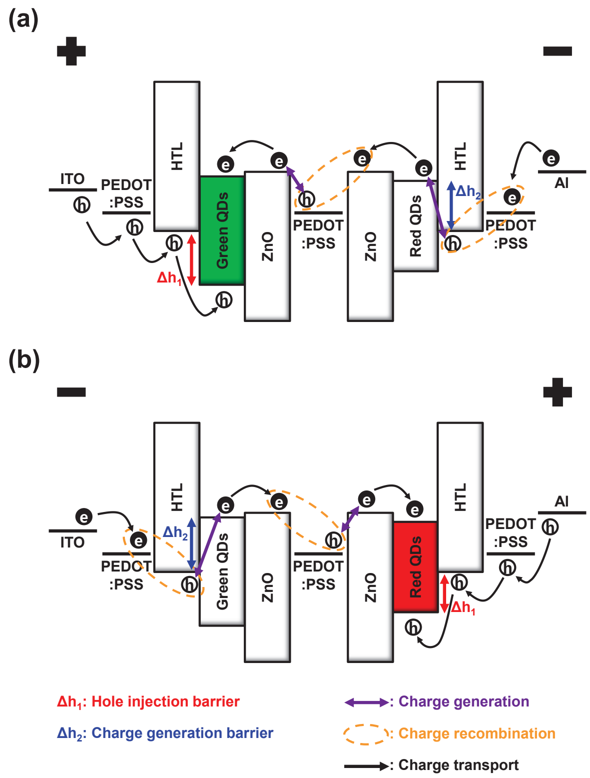

Fig. 3 Carrier transport paths of hole-only tandem QD-LED under (a) positive bias and (b) negative bias.

Table 1. Summary J-V-L characteristics of the tandem QD-LEDs with two different HTLs depending on the polarity of the applied voltage.

Bias	HTL	V_T [V]	L_{max} [cd/m^2]	EQE_{max} [%]
Positive	Poly-TPD	14.1	36	0.005
	PVK	31.0	2	0.008
Negative	Poly-TPD	23.3	65	0.156
	PVK	26.7	30	0.336