Mechanisms of operation in quantum-dot light-emitting diodes

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ABSTRACT

Mechanisms of operation in quantum-dot light-emitting diodes (QLEDs) have been investigated theoretically and experimentally. Important factors governing the current efficiency of QLED were examined using a machine learning approach. High hole injection barrier to QD is the dominant efficiency limiting factor, and the machine learning result was confirmed experimentally. A mechanism of high current efficiency even in the presence of high hole injection was discussed in terms of device simulation.

1. INTRODUCTION

Quantum-dots (QDs) are solution-processed semiconductor nanocrystals that feature narrow band emission, size-tunable band-gaps, and high photo-luminescence quantum efficiency [1]. These advantages make them good materials for use in displays. Organic light-emitting diode (OLED) displays limit themselves to smaller color gamuts because the emission spectra of OLED are still too broad. Quantum-dot light emitting diode (QLED) is a competitive alternative to organic light-emitting diodes (OLEDs) and a potential candidate for next generation display (8K TV) [2]. High-performance and solution-processed QLEDs have been extensively studied since the first report of the QLED in 1994 [3]. In general, hole injection barrier from the hole transport layer (HTL) to the QD layer is about 1 eV in solution-processed QLEDs, whereas electron injection barrier from the electron transport layer (ETL) to the QD layer is negligible. Although such high injection barrier exists in QLEDs with Cd-based QDs, high current efficiency was reported in the QLEDs [4-5]. It is therefore important to investigate efficiency limiting factors of QLEDs including hole injection mechanisms.

In this study, first, we examine efficiency limiting factors of QLEDs using a machine learning approach. We then examine the efficiency limiting factors experimentally. We also examine the origin of high current efficiency in QLEDs using device simulation by taking account of the efficiency limiting factors.

2. MACHINE LEARNING

We collected 96 published papers dealt with the device characteristics of QLEDs with Cd-based QDs and with conventional structures (transparent conductive oxide (anode)/hole injection layer (HIL)/HTL/QD/ETL/cathode). The energy levels (highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO)) and the thicknesses of HIL, HTL, QD layer, and ETL were obtained from the papers as features, and then the importance of the features with respect to current efficiency was examined in terms of random forest regression.

3. RESULTS AND DISCUSSION

The random forest regression shows that the energy level of the valence band edge of the QD layer has the highest importance and the HOMO energy levels of the HIL and the HTL have high importance. The results indicate that hole injection from the HTL to the QD layer is the limiting factors for the current efficiency of QLEDs. In addition, the energy level of the conduction band edge of the QD layer is as important as the energy level at the valence band edge of the QD layer. This is because the ETL collected from the published papers is mostly ZnO while QDs with different emission wavelengths were used as QD layers.

In order to investigate the relation between the hole injection barrier and the current efficiency experimentally, QLEDs with different hole injection barriers were fabricated. We measured the current density – luminance - voltage (*J-L-V*) characteristics and the current efficiency - current density characteristics of QLEDs with poly[9,9-dioctylfluorene-co-N-[4-(3-methylpropyl)]-di-phenylamine] (TFB) or poly-N-vinyl carbazole (PVK) used as HTL (shown in Fig. 1). CdSe QDs with red emission was used as the QD layer.

The *J-V-L* and the current efficiency - current density characteristics of QLEDs are shown in Fig. 2. The QLED with PVK as HTL exhibits higher brightness in comparison with the QLED with TFB as HTL. The maximum current efficiency of the QLED with TFB (HOMO energy level of 5.3 eV) is 1.55 cd/A, whereas that of the QLED with PVK (HOMO energy level of 5.8 eV) is 8.26 cd/A. Although we examined two QLEDs with different hole injection barriers only, the results (the QLED with lower injection barrier exhibits higher current efficiency) are consistent with the results of the machine learning study. The reduction in hole injection barrier in QLEDs is extremely important to improve the current efficiency.

Device simulation (Atlas™, SILVACO) was conducted to investigate the mechanisms of the operation of QLEDs. Figure 3 shows the band diagram of QLED at the external applied voltages of 1.0 V and 2.0 V. We found that the energy band level is shifted owing to the accumulation of holes in the HTL and electrons in the QD layer at the interface between HTL and QD layer. Such hole injection barrier lowering is the origin of high current efficiency of QLEDs in literature. High electric field at the interface between HTL and QD layers can be an origin of the degradation of QLEDs as well.

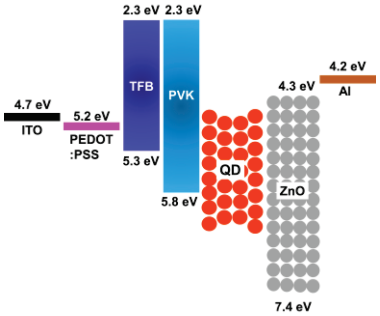

Fig. 1 Energy diagram of the QLEDs fabricated in this study

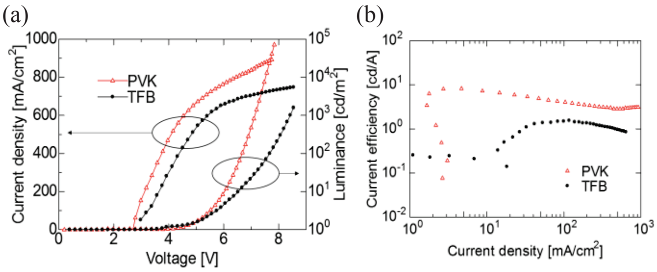

Fig. 2 (a) *J-L-V* characteristics and (b) current efficiency of QLEDs with TFB or PVK as HTL

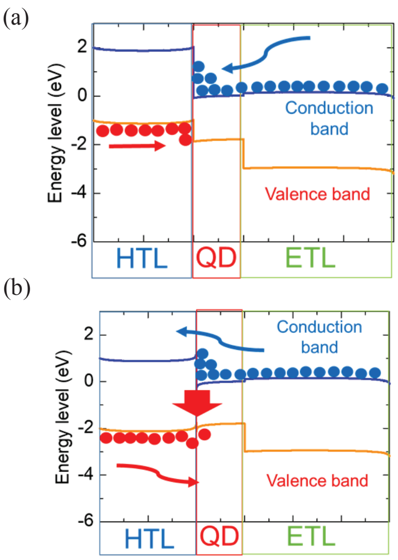

Fig. 3 Energy diagram drawn from the device simulation at the external applied voltage of (a) 1.0 V and (b) 2.0 V

Figure 4 shows the relation between the current efficiency and hole injection barrier of QLEDs with different HTLs in literature [6]. We found from Fig. 4 that the reduction in the hole injection barrier does not necessarily contribute to the improvement of the current efficiency.

We found that the electron mobility of HTL as well as the hole injection barrier is responsible for high current efficiency (since the electron mobilities of HTLs were not described in literatures, the electron mobility of HTLs was not taken into account in the present machine learning study). The relation between the electron mobility of HTL and the current efficiency was investigated by device simulation, and we found that the current efficiency was improved by reducing the electron mobility of HTL. The result shows that the electron mobility of HTL, whose importance has not been known before, is one of the efficiency limiting factors of QLEDs.

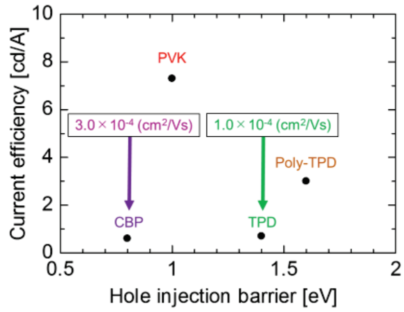

Fig. 5 Relation between the current efficiency of QLEDs and hole injection barrier in the QLEDs with different HTLs. Poly-TPD is poly(4-butyl-phenyl-diphenyl-amine), CBP is 4,4'-bis(carbazole-9-yl)-biphenyl, and TPD is N,N'-diphenyl-N,N'-bis-(3-methylphenyl)-1,1'-diphenyl-4,4'-diamine. The values shown in the figure are electron mobilities. The electron mobilities of PVK and poly-TPD have not been reported mainly because the electron mobilities of the HTLs are immeasurably small.

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