Enhanced Performance of I-III-VI Type Quantum Dot-Light-Emitting Devices through Introducing Alloyed Oxide Nanoparticle-Based Electron Transport Layer

Chang-Yeol Han, Jong-Hoon Kim, Ki-Heon Lee, Dae-Yeon Jo, and Heesun Yang**
Dept. of Materials Science and Engineering, Hongik University, Seoul 121-791, Korea
Tel.:82-2-320-3039, E-mail: hyang@hongik.ac.kr

Quantum dot-light-emitting diode (QLED) is typically based on the multilayered configuration consisting of hole injection layer (HIL), hole transport layer (HTL), quantum dot (QD) emitting layer (EML), and electron transport layer (ETL). Keeping the energy levels of QDs *versus* surrounding charge transport layers (CTLs) in mind, various HTL/ETL combinations have been attempted to improve the device performance. A breakthrough in dramatically enhancing the performance of QLED has been made by adopting hybrid CTLs of organic HTL and inorganic solution-processed ETL with TiO_2 sol or ZnO nanoparticle (NP) dispersion. Holloway's group reported the fabrication of hybrid-type QLED comprising poly(*N,N'*-bis(4-butylphenyl)-*N,N'*-bis(phenyl)benzidine) (poly-TPD) HTL and ZnO NP ETL, showing a maximum external quantum efficiency (EQE) of 1.8% [1]. Later, with uniquely designed, thick-shelled QDs, a much higher EQE of 12.6% could be achieved by our group from a similar hybrid device with poly(9-vinylcarbazole) (PVK) HTL and ZnO NP ETL [2].

Herein, three types of I–III–VI QDs with ternary Cu-In-S (CIS) and quaternary Cu-In-Ga-S (CIGS) and Zn-Cu-In-S (ZCIS) are utilized for the formation of QD EML of solution-processed, multilayer-structured QLEDs. While PVK HTL is commonly applied in those devices, inorganic ETLs of a series of colloidal $Zn_{1-x}Mg_xO$ (x=0, 0.05, 0.1) NPs are unprecedentedly introduced (see Figure 1a and b for device schematic and transmission electron microscopic (TEM) image, respectively). Increase in band gap of metal oxide NP by MgO alloying produces an upshift of its conduction band minimum (CBM) level, beneficially leading to a closer proximity in energy between CBMs of QD EML and ZnMgO ETL and consequently rendering the electron injection to QD region facilitated, as shown in Figure 1c. As a result, the device with ZnMgO NP ETL is superior to that of ZnO one with respect to luminance and efficiency. As presented in Figure 1d, for all I–III–VI QLEDs having CIS, CIGS, and ZCIS QDs, the consistent trend of a strong dependence of device performance on the type of ETL is indeed observed, proving an efficacy of such alloyed ETL in improving the device performance.

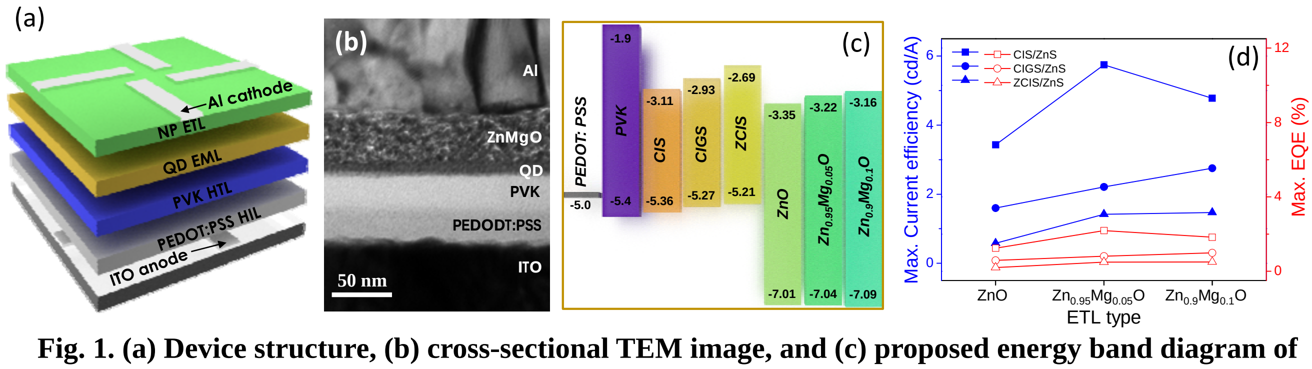

Fig. 1. (a) Device structure, (b) cross-sectional TEM image, and (c) proposed energy band diagram of solution-processed, multilayered QLED. (d) ETL type-dependent maximum values in current efficiency and EQE of CIS/ZnS, CIGS/ZnS, and ZCIS/ZnS QLEDs.

Acknowledgment

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIP) (No. 2013R1A2A2A01068158).

References

1. L. Qian, Y. Zheng, J. Xue, and P. H. Holloway, *Nat. Photonics*, 5, 543 (2011).
2. K. H. Lee, J. H. Lee, H. D. Kang, B. Park, H. Ko, C. Lee, J. Lee, and H. Yang, *ACS Nano*, 8, 4893 (2014).