Mixed Single Layer-Based Quantum Dot Light Emitting Diodes with the Substrate Temperature Variation of ZnO Layer

Mohammad Mostafizur Rahman Biswas, and Hiroyuki Okada

okada@eng.u-toyama.ac.jp
University of Toyama, Graduate School of Science and Engineering
930-8555, Toyama city, Toyama, Japan

Keywords: cadmium-free quantum dot light emitting diode, sputtered zinc oxide, mixed single layer

ABSTRACT

Mixed single layer-based quantum dot light-emitting diode (QLED) was fabricated using the sputtered ZnO layer, where the substrate temperature was varied from RT to 200℃ to improve the electrical properties and device performances. Maximum power efficiency of 5.01 lm/W, current efficiency of 5.58 cd/A, and external quantum efficiency *of 2.41% were achieved for the fabricated QLED.*

1 INTRODUCTION

Quatum dot light emitting diode (QLED) is an emerging research field for the future lighting and display applications.[1] Most common attributes of the quantum dos (QDs) are size-dependent tunable emission, wide viewing angle, and high quantum yields. [2] Cd-based QD contains heavy metal substances, which are harmful to human matters. Therefore, researchers are trying to improve the Cd-free QDs. Among them, I–III–VI type QD group, such as ZnCuInS/ZnS (ZCIS/ZnS), is the most stable QD for the application aspect.[3] There are three primary colors of QLED, such as red, green, and yellow. One of these colors, the yellow or orangish emission-based ZCIS/ZnS QLEDs, shows better performance.

Furthermore, it is common to improve the device performances in two ways, the material development of QDs itself and the device structural development of QLEDs. The device performance of QLED mainly depends on the carrier balance to generate excitons. Commonly, researchers use the inverted QLEDs for the balanced excitons generation, as it contains no additional layer between the electron transport layer (ETL) and emission layer (EML). One of the device selections, a mixed single layer (MSL), is an interesting technique. Naka et al. introduced the MSL into the fabrication of organic light – emitting diode (OLED) device in order to improve the device's performance and durability.[4] Wang et al. showed optimization of the MSL OLED device process, performance, and stability.[5]
Considering the above background, we have reported inverted MSL-QLED, where solution-processed QLED is employed in order to improve the carrier injection to the QDs covered with bipolar conduction material (BM) and ETL of organic materials.[6] In this material system, the sphere shape of QD material is covered with MSL. Therefore, a good carrier injection balance between electron and hole is achieved, and flat current efficiency (*CE*) under wide current density (*J*) variation is also realized. In this time, we have investigated the inverted MSL-QLEDs, where the metallic ZnO was used with the substrate temperature (T_d) variation between RT and 200℃.

2 EXPERIMENT

To fabricate the MSL-QLED, we have selected the commercially available ZCIS/ZnS QD with the photoluminescent quantum yield (PLQY) of 45% and particle size of 4~5 nm. The metallic ZnO film was deposited on the ITO substrate by using the radio frequency (RF) sputtering technique. The ETL of 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP), BM of 4,4'-bis(carbazolyl) biphenyl (CBP), and EM of QDs materials were used to obtain the MSL. The mixing ratio of the organic materials were BCP: QD: CBP = 40: 20: 40 is used, which was optimized in our previous work. [7] The fabrication process is as follows: Patterned ITO substrate was thoroughly cleaned using organic cleaning solvents and UV ozone chamber. After that, the pure ZnO (99.99%) was sputtered on the ITO substrate using RF sputtering technique. Base pressure below 2×10^{-6} torr and the working gas was argon (Ar, 99.999%). During the sputtering process, the T_d was varied at RT, 100℃ and 200℃ to obtain a compact texture and good electrical properties of ZnO. Then, polyethyleneimine-ethoxylated (PEIE) (4 wt%) solution was prepared with ethanol and was coated on ZnO film using the rinse method. And then, baking was carried out at 55℃ for 1 h. Next, the MSL consist of BCP, ZCIS/ZnS, and CBP was dissolved into toluene as a common solvent for all the materials. The prepared mixed solution was spin-coated inside the glove box. After then, baking was done at 80℃ for 10 min, and 110℃ for 5 min to remove the solvent. Finally, the substrates were transferred from the glove box to the evaporator immediately, where the following layers were evaporated using the vacuum evaporation technique.
The fabricated MSL-QLED structure was ITO (100 nm)/ ZnO (60 nm)/ PEIE (<1nm)/ MSL (20 nm)/ molybdenum oxide (MoO_3, 10 nm)/ Al (70 nm).
All measurements were carried out in air at RT. The fabricated device area was 0.04 cm^2.

3 RESULTS AND DISCUSSIONS

Smooth surface, compact grain boundary and higher mobility are necessary for the excellent performance of the QLEDs. Considering this situation, we have varied the T_d during the sputtering of ZnO layer. Therefore, significant improvement in device performance was observed with the improved electrical parameters of ZnO. The carrier mobility μ and carrier concentration N was observed from the Hall measurement technique. At the

T_d of 200℃ with the ZnO thickness of 60 nm, the value of μ was 24.3 cm²/Vs, and N was 4.98×10^{20} cm^{-3}, whereas μ was 15.1 cm²/Vs and 3.5×10^{20} cm^{-3} for the RT of ZnO film.

Figure 1 (a) shows the J vs. voltage (V) and luminance (L)-V characteristics of the QLED varied with the T_d of ZnO layers. The turn-on voltage (V_t) was gradually decreased with increment of T_d and minimum V_t is 2 V for the T_d = 200℃. From the L-V characteristics, the maximum luminances are 980, 1,200, and 1,090 cd/m² for the T_d at RT, 100℃ and 200℃, respectively. The power efficiency η of the QLED was 3.38, 4.15, and 5.01 lm/W for the T_d at RT, 100℃ and 200℃, respectively, as shown in Fig. 1(c). The CE and external quantum efficiency (EQE) are shown in Fig. 1(b). The device with RT, 100℃, and 200℃ of the ZnO layer exhibit steady and uniform behavior. The maximum CE and EQE were observed 3.64, 4.49, 5.58 cd/A, and 1.9, 2.13, 2.41%, respectively.
Considering the normalized EL spectrum, as shown in Fig. 1(d), the spectral changes were observed by varying the T_d of ZnO as the main peaks at around 575 nm and supplementary peaks at 430 nm. In detail, the obtained intensity peaks are 582, 574, and 571 nm for the T_d value of RT, 100℃, and 200℃, respectively.

4. CONCLUSIONS

We had investigated the substrate temperature variation of the ZnO layer for the fabrication of MSL QLED devices. The QLED at the T_d of 200℃ showed better device performance of CE = 5.58 cd/A and EQE = 2.41% with the lower V_t = 2 V. In all cases, the MSL QLEDs showed the better performances than the stacked QLED. Therefore, it can conclude that the MSL-QLED for the ZnO layer of T_d = 200℃ shows a visible impact on the device operation.

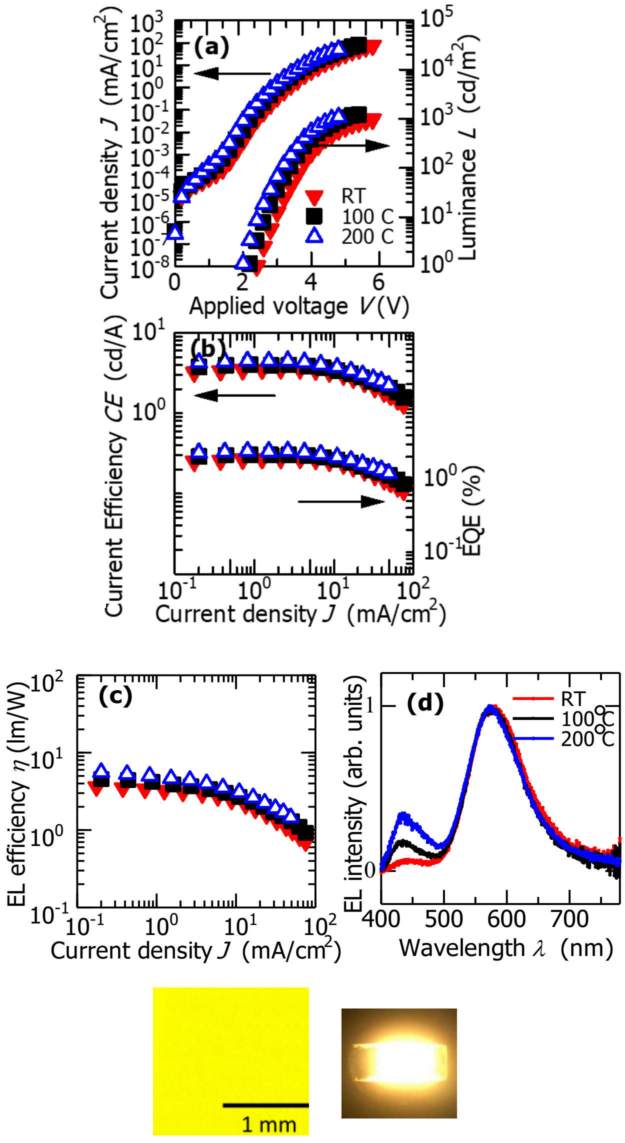

Figure 1: Characteristics curve of (a) J-V, L-V (b) CE-J, EQE-J (c) EL spectrum of fabricated QLED at different T_d.

REFERENCES

[1] J. W. Jo, Y. Kim, J. Choi, F. P. G. de Arquer, G. Walters, B. Sun, O. Ouellette, J. Kim, A. Proppe, R. Quintero-ermudez, J. Fan, J. Xu, C. S. Tan, O. Voznyy, and E. H. Sargent, "Enhanced open-circuit voltage in colloidal quantum dot photovoltaics via reactivity-controlled solution-phase ligand exchange," Adv. Mater. **29**, pp. 1703627-1-6 (2017).

[2] M. Kovalenko, L. Manna, A. Cabot, Z. Hens, D. Talapin, C. Kagan, V. Klimov, A. Rogach, P. Reiss, D. Milliron, P. Guyot-Sionnnest, G. Konstantatos, W. Parak, T. Hyeon, B. Korgel, C. Murray, and W. Heiss, "Prospects of nanoscience with nanocrystals," ACS Nano **9**, pp.1012-1057, (2015).

[3] J. Grim, L. Manna and I. Moreels, "A sustainable future for photonic colloidal nanocrystals", Chem. Soc. Rev. **44**, pp. 5897-5914, (2015).

[4] S. Naka, K. Shinno, H.Okada, H.Onnagawa and K. Miyashita, "Organic electroluminescent devices using a mixed single layer", Jpn. J. Appl. Phys. **33**, L1772-L1774 (1994).

[5] Z. Wang, H. Okada, and S. Naka, "Evaluation of reliability in rubrene-based organic light emitting devices with a mixed single layer", Jpn. J. Appl. Phys. **49**, pp. 01AA02-1-3 (2010).

[6] M. M. R. Biswas and H. Okada, "Cd-Free quantum-dot light-emitting diode with a mixed single layer to improve the flatness of current efficiency", SID2021Dig. Tech. Pap. pp. 967-970 (2021).

[7] M. M. R. Biswas and H. Okada, "Investigation of the inverted ZnCuInS/ZnS based quantum-dot light-emitting diodes with the sputtered ZnO film layers", Proc. Int'l Display Workshop, pp. 427-428 (2020).