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# Enhancement of EL performance and flexibility of Perovskite-light emitting diodes

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In this thesis, I mainly focus on the improvement of the performance of PeLED using amorphous oxide semiconductor (AOS) as an ETL. To accomplish this objective, the following five chapters were established.

Chapter 1 described a brief background on perovskite light emitting diodes (PeLEDs) and technical issues to be resolved. The characteristics of amorphous oxide semiconductors were pointed out as promising electron transport layers (ETL) for next generation display. Based on these backgrounds, motivations and objectives of this thesis were addressed.

In Chapter 2, the feasibility of AOS as an ETL in PeLED was examined. Recently, there are many reports about low-dimensional perovskite emission layer (EML) to realize high photoluminescent quantum yield. However, high PLQY material always does not guarantee high EL performance because low-D materials possess poor charge transport nature resulted from their highly localized electronic structure. I discovered that PL property of 3D materials was significantly governed by adjacent charge transport layer owing to non-radiative recombination arisen from small exciton binding energy. This finding enables us to exploit AOS based ETL with proper electronic structure to not only confine exciton and realize effective charge injection into 3D EML. This strategy demonstrates the significance of high charge transport property of 3D materials for highly efficient PeLED. The proposed AOS ETL, amorphous zinc silicate (a-ZSO) possesses not only sufficiently shallow electron affinity (~3.2 eV) to confine excitons but also high electron mobility (~0.8 $cm^2/V$ s) to effectively transport electrons. Thanks to the synergy effect of a-ZSO ETL and 3D perovskite EML, very low operating voltage of 2.9 V at 10,000 $cd/m^2$ and high efficiency of 33 lm/W were attained. The obtained ultra-high brightness of ~500,000 $cd/m^2$ at 5 V also exhibited the validity of the proposed strategy. It was also extended into 3D $CsPbBrI_2$ (red) and 3D $CsPbBrCl_2$ (blue) PeLEDs, and realized a record high brightness of 20 000 $cd/m^2$ for the red PeLED.

Based on the results of Chapter 2, an extended study was conducted to realize PeLED with high efficiency and excellent mechanical robustness. For highly flexible PeLEDs, simple device structure comprised of minimal layers is a key point because achieving high flexibility and performance of all layers and interfaces are challenging. In Chapter 3, I propose a simplified device structure by applying the concept of core-shell structure into transparent conductive electrode (TCE) and perovskite EML. For the TCE, tri-layer structure of ZSO (ETL) /Ag/ZSO ETL is adopted to utilize the intrinsic characteristic of a-ZSO ETL such as good charge transport property and ohmic contact with Ag layer. Moreover, Ag layer is fully embedded by ZSO layers. Thus, this edgeless structure enables to prevent the chemical degradation of Ag layer from chemical precursor solution and humidity and to solve the issue of weak adhesion of Ag to oxide. On the other hand, mesh-like perovskite EML is fabricated by adding poly ethylene oxide (PEO) into perovskite precursor. Furthermore, leakage current in the area w/o EML is suppressed by large energy barrier (PN junction) formed at ETL/HTL interface. Thus, core shell structured EML can realize both high EL performance and excellent flexibility. Consequently, PeLED comprised by only 4 layers and fabricated on 50 um thick plastic substrate attains highly stable performance for 100,000 times bending stress at 4 mm bending radius, demonstrating the validity of our strategy.

In previous chapters, we confirmed that oxide ETL with superior electrical property exhibits high compatibility with PeLED. Thus, various applications of oxide ETL in perovskite optoelectronics are expected by combining with 3D organic–inorganic hybrid perovskite EMLs. However, there were critical problems to prevent the utilization of oxide ETL. In Chapter 4, it was revealed that mutual chemical reaction occurs between metal oxides and hybrid perovskite EMLs. It was discovered that PL property of EMLs significantly degrade after heat treatment and a new phase is formed between the two layers. On the other hand, it was also revealed that employing interlayers used in recent hybrid PeLEDs such as LiF, inhibit the chemical reaction showing no PL degradation and smooth interface. Nevertheless, the interlayer method involves another problem of lowering EL efficiency due to the subsequent increase in operating voltage. Based on this background, a new method to suppress the aforementioned chemical reaction without an interlayer was explored. Consequently, it was found that the use of 18-crown 6-ether (18C6) as an additive in the precursor solution achieves (i) suppression of the chemical reaction and (ii) a low operating voltage via efficient charge injection (3.2 V at 10,000 cd/$m^2$).

In Chapter 5, I summarized the major conclusions obtained in this thesis.