Solution-processed small molecular phosphorescent organic light emitting devices with a mixed single layer

Zhaokui Wang, Shigeki Naka and Hiroyuki Okada

Univ. Toyama, 3190 Gofuku Toyama, Japan
Phone: +81-76-445-6731 E-mail: zhaokuiwang@hotmail.com

1. Introduction
Phosphorescent organic light emitting devices (PHOLEDs) that have the potential to achieve internal quantum efficiency close to 100% have attracted considerable research interest.[1,2] Previous attempts of solution processing phosphorescent devices have been achieved using phosphorescent polymers and dendrimers with high efficiency.[3-5] However, satisfactory fabricating cost and device performance has not yet been achieved. In the same manner, sprayer solution-processed PHOLEDs were reported.[6] Fortunately, OLEDs with a mixed single layer structure have attracted wide attention because they have potential for improving the device performances.[7-9] Moreover, it is possible to use simple wet-process due to its simplest architecture. In this paper, we have reported the highly simplified single layer PHOLED.

2. Experiments
Mixed organic layer was consist of electron transport material 1,3,5-tris[2-N-phenylbenzimidazolyl]benzene (TPBi), phosphorescent dopant fac-tris (2-phenylpyridine) iridium (Ir(ppy)_3) and host materials 4,4‘-bis (N-carbazolyl) biphenyl (CBP). Mixed organic layer (MOL) was spin-coated using 1.0 wt% chloroform solution by mixing TPBi, CBP and Ir(ppy)_3 in weight. The optimized structure of single layer PHOLED was ITO/ PEDOT (50 nm)/MOL (100 nm)/LiF (1 nm)/Al (70 nm). For comparison, vacuum-processed mixed single layer PHOLED was also fabricated by thermally evaporating. Hole-dominated devices (HDDs) with a structure of Au (30 nm)/MoO_3 (20 nm)/MOL (100 nm)/MoO_3 (20 nm)/Au (30 nm) were fabricated by solution- and vacuum-processed means for comparing charge transporting properties.

3. Results and Discussion
Figure 1 (a), (b) and (c) shows the current density vs voltage (J-V), power efficiency \( \eta_p \) and external quantum efficiency (EQE) characteristics of PHOLEDs fabricated by solution- and vacuum-processed means, respectively, where mixing ratio of organic materials in single layer is kept with an optimized ratio CBP: Ir(ppy)_3: TPBi = 100:5:40. Leakage current in solution-processed devices is larger than that in vacuum-processed devices. We ascribe it to a wet process being included in solution-processed device fabrication. It is identifiable that an almost identical luminescence characteristic was achieved between devices by two processing methods. Obtained maximum power efficiency \( \eta_p \) of 11.5 lm/W corresponding to external quantum efficiency (EQE) of 9.6% for solution-processed device. At lower current densities, \( \eta_p \) and EQE of solution-processed devices are lower than that of vacuum-processed devices, which is related to the large leakage current in solution-processed devices as observed in J-V characteristics. Whereas, at higher current densities, \( \eta_p \) and EQE of solution-processed devices are lower than that of vacuum-processed devices. As different glass transition temperature of three organic materials and instable evaporating rate, it is difficult for vacuum process to ensure the formation of uniform distribution of all organic materials in mixed organic layer. However, the solution processing can do it easily due to its wet-processing merit. It suggests that a solution-processed device is excellent for uniform material distribution in mixed organic single layer.

Fig. 1 Device characteristics of mixed single layer PHOLEDs fabricated by solution- and vacuum-processed techniques.
Figures 2 (a) and (b) show the temperature dependent of $J$-$V$ characteristics in solution- and vacuum-processed HDDs, respectively. For solution-processed device, two distinct regions were seen in the $J$-$V$ curves in a double logarithmic representation. The slope in the low bias region was about 2 and temperature dependence of current density was obvious in this region. We ascribed the hole injection to be a Schottky thermal emission mechanism with a barrier height of 0.38 eV. The slope in the high applied bias region showed a great change with temperature from 6.4 to 9.8 upon cooling from 273 to 233 K. It suggests that hole transport is bulk trapped conduction in higher bias condition. For vacuum-evaporated device, $J$ increases smoothly with $V$ in whole measured voltage range. The obvious temperature dependence of $J$-$V$ characteristics implies that the hole injection could also be ascribed to Schottky thermal emission and a injection barrier 0.28 eV is obtained after fitting.

The above results demonstrate that the charge injection and conduction mechanism is different between solution-processed and vacuum-evaporated devices. The difference implies that some changes were taken place at the interfaces of metals/MOL between these two-processed devices. We suppose that circumstance effect, chemical interaction and interfacial state were the main reasons causing the large difference of charge conduction between two-processed devices. In the case of solution-process, such as spin coating, solvents may remain in the organic film even after baking. Furthermore, the formation of a thin insulating layer between metal and organic layer is probable when solution process is carried out in the device fabrication. It largely affects the creating of surface-state charge at interface and space charge in organic layer, and the interfacial conditions will be determined by the redistribution of these generated charges. In addition, the high reactivity of the vaporized hot metal atom often leads to a chemical reaction at the interface when the metal is deposited on an organic layer by evaporation. These chemical reactions will depend sensitively on the morphology and chemistry conditions of interfaces. In contrast, the chemical interaction situation in vacuum process is usually milder than that in solution process.

4. Conclusions

In summary, we have demonstrated a highly simplified small molecular PHOLED with the maximum $\eta_P$ 11.5 lm/W corresponding to EQE 9.6%. This result is achieved by mixing all host and dopant into one layer with a wet process without using multilayer structure for avoiding the complexities of vacuum processing, which greatly simplifies the device design. The simplified devices are also shown to have comparable device performance to vacuum-processed single layer PHOLEDs. This simplified device design strategy represents a pathway toward large area, low cost and high efficiency OLEDs in the future. Further investigation of charge transport and their dynamics at metal/organic interfaces in organic devices fabricated using solution- and vacuum-processed methods is necessary.

References