

Improvement of power efficiency of hybrid white OLEDs based on p-i-n structures

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ABSTRACT

In this article, hybrid white organic light-emitting diodes (WOLEDs) under p-i-n structures have been investigated in terms of power efficiency. By using tris(8-hydroxy quinolinato) aluminum (Alq_3) doped with 8-hydroxy-quinolinato lithium (Liq) as an n-type and WHI112 doped with molybdenum trioxide (MoO_3) as a p-type, the typical device structure of ITO/WHI112: 20 wt.% MoO_3 (55 nm)/HTG-1 (10 nm)/UBH15: 3 wt.% EB502 (10 nm)/EPH31: 3 wt.% EPY01 (25nm)/3TPYMB (10 nm)/ Alq_3 : 33 wt.% Liq (25 nm)/Al (150 nm) was fabricated. It has been found that the p-i-n device based device showed the lowest driving voltage and highest power efficiency among the undoped and n-type devices. At the current density of 20 mA/cm^2 , the roll-off of the efficiency in the p-i-n device was much smaller than the n-type and the undoped devices. The current and power efficiency of the p-i-n device were maintained with 17.2 cd/A and 5.1 lm/W at 100 mA/cm^2 , it was reduced to 7.5 % and 21 %, respectively. In contrast, the n-type device exhibited the significant reduction of efficiency (14.4 cd/A and 3.8 lm/W at 80 mA/cm^2), it was reduced to 20 % and 39.6 %, respectively. The superior performances of the p-i-n structure based device were attributed to the high hole injection ability of WHI112: MoO_3 and high electron mobility of Alq_3 :Liq, leading to high power efficiency and low driving voltage. A better balance of electrons and holes could contribute to a good current efficiency for the device. These findings strongly indicated that carrier injection ability and balance showed significant affects on the performance of OLED.

Keywords: Highly efficient OLEDs, hybrid white OLEDs, p-i-n OLED.

1. INTRODUCTION

Since the discovery of the efficient organic light-emitting diodes (OLEDs) considerable interest has been increased in developing OLEDs with high efficiency, low operating voltage for display applications is concerned [1]. Much effort has been expanded for improving the OLED performance by modifying its structure to achieve the effective and balanced carrier injection. The carrier injections from electrodes are dependent on the energy barrier height at the interfaces between the electrodes and organic layers [2,3]. Reasonable charge carrier control in the OLED emitting layers (EMLs) is a key factor in OLED low driving voltage and high efficiency structure design. Two approaches are most frequently used to overcome the driving voltage problem. The first approach involves with inserting a thin layer as an anode buffer layer between the indium tin oxide (ITO) and hole transport layer (HTL). This buffer layer reduces the energy barrier and enhances the charge injection at the interface and ultimately reduces the driving voltage improving the device power efficiency [4-7]. The second method involves the use of strong electron acceptor and donor materials as dopants in the organic HTL and electron transport layer (ETL) [8,9]. Great efforts have been carried out to enhance n-doping electron transport conductivity [10]. DING et al. [11] have demonstrated the significantly enhanced device performance by combination of Lithium hydride (LiH) doped 8-hydroxyquinoline aluminum (Alq_3) as an electron transport layer (ETL). However, a few studies have been reported using 8-hydroxy-quinolinato lithium (Liq) as the electron injection layer [12]. The p-doping of HTL for enhancing the hole injection and lowering the drive voltages in OLEDs has attracted much attention [13]. The p-doping HTL is typically made by co-evaporating the hole transporting materials with a strong electron acceptor like the molybdenum trioxide (MoO_3) [14] and the tetrafluoro- tetracyano-quinodimethane ($\text{F}_4\text{-TCNQ}$) [15]. The p-doping could also achieve the ohmic conductivity to minimize the voltage drop across

the ITO/HTL interface. Judicious control of doping levels can also lead to the efficient carrier injection by the tunneling [16]. It is very difficult to balance the holes and electrons in the emitting layer, because hole mobility is generally faster than the electron mobility in organic materials. In order to solve this problem, several kinds of HTL, ETL, hole block layer and electron block layer have been studied [17,18].

In this paper, we have demonstrated hybrid white organic light-emitting diode (WOLEDs) devices based on the p-i-n structure with Liq doped into Alq₃ as the n-doping layer and MoO₃ doped into WHI112 as the p-doping layer. In order to reduce the driving voltage and improve efficiency, we use MoO₃ doped into WHI112 as the p-doping layer and Liq doped into Alq₃ as the n-doping layer, power efficiency and carrier balance have been overwhelmingly improved. The electrical engineering and hybrid WOLED charge balance are developed based on these experimental results. The mechanism of such improvement is also discussed on these experimental results.

2. MATERIALS AND METHODS

Glass coated with indium-tin oxide (ITO) was used as the starting substrate. The substrate was immersed sequentially in acetone and isopropyl alcohol under the ultrasonic bath for 15 min each, following by rinsing in DI water. The substrates were dried with nitrogen gas. Then the samples were treated with the oxygen plasma for 1 min. prior to use. The devices were prepared by the vapor deposition onto the ITO coated glass substrate. Firstly, the series of electron-only devices were fabricated in order to obtain some data on the electron transport ability of Alq₃ doped with Liq layers. The structures of electron-only devices were as follows: ITO/Alq₃: x wt.% Liq (30 nm)/Al (130 nm), where x was 0 wt.% for device E-1, 10 wt.% for device E-2, 33 wt.% for device E-3, and 50 wt.% device E-4 (Table 1). Secondly, the study of hole-injection ability of WHI112 doped with MoO₃ layer, the series of hole-only devices were fabricated. Hole-only device got the following structures: ITO/ WHI112: y wt% MoO₃ (50 nm)/ HTG-1 (15 nm)/Al (130 nm), y was 0 wt.% for device H-1, 10 wt.% for device H-2, 20 wt.% for device H-3, and 30 wt.% for device H-4 (Table 2).

Table 1: The parameters of electron-only devices.

DEVICE	ANODE	n-TYPE		CATHODE
		Alq ₃	Liq	Al
E-1	ITO	30 nm	0%	130 nm
E-2	ITO	30 nm	10%	130 nm
E-3	ITO	30 nm	33%	130 nm
E-4	ITO	30 nm	50%	130 nm

Table 2: The parameters of hole-only devices.

DEVICE	ANODE	p-TYPE		HTL	CATHODE
		WHI112	MoO ₃	HTG-1	Al
H-1	ITO	50 nm	0%	15 nm	130 nm
H-2	ITO	50 nm	10%	15 nm	130 nm
H-3	ITO	50 nm	20%	15 nm	130 nm
H-4	ITO	50 nm	30%	15 nm	130 nm

Finally, hybrid WOLEDs devices were fabricated with undoped, n-type and p-i-n structures. HTG-1 was used as HTL, blue host UBH15 doped with blue fluorescent dopant EB-502 as blue EML, yellow host EPH-31 doped with yellow phosphorescent dopant EPY01 as yellow EML, and tris(2,4,6-trimethyl-3-(pyridin-3-yl)phenyl) borane (3TPYMB) was used as hole blocking layer (HBL), while LiF and Al were used as electron injection layer and cathode, respectively. The energy band diagrams and molecular structures were displayed in Fig. 1. The structures of undoped devices was ITO/WHI112 (55 nm)/HTG-1 (10 nm)/UBH15: 3 wt.% EB502 (10 nm)/EPH31: 3 wt.% EPY01 (25nm)/3TPYMB (10 nm)/Alq₃ (25 nm)/LiF (0.8 nm)/Al (150 nm). When we added Alq₃: 33 wt.% Liq (25 nm) and WHI112: 20 wt.% MoO₃ (55 nm) onto the undoped devices, we obtained n-doped layer and p-doped layer (Table 3), respectively. All materials were purchased from e-Ray Optoelectronics Technology Co., Ltd., Taiwan (R.O.C.). The organic layer and

the cathode layer were deposited under the ultrahigh vacuum chamber at 4×10^{-6} Torr. The active area of the devices was $5 \times 5 \text{ mm}^2$. The thickness of the organic layers was monitored by using quartz-crystal monitor. Current-voltage characteristics were measured with the computer-controlled Keithley 2400 Source Meter and electroluminescence (EL) spectra was measured with the Spectrascan PR650 photometer. All the measurements were carried out at room temperature and atmosphere.

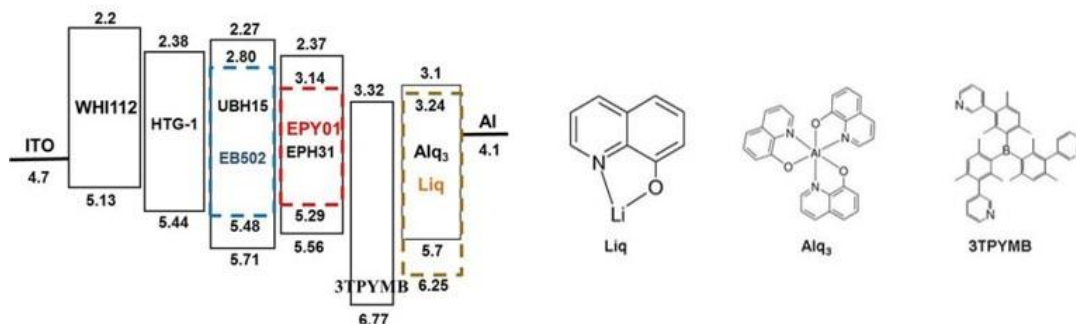


Figure 1: Energy band diagrams and molecular structures of the tested materials.

Table 3: The parameters of undoped, n-doped and p-i-n devices.

DEVICE	HIL	P-DOPANT	HTL	BLUE EML	YELLOW EML	HBL	ETL	n-DOPANT	EIL		
	WH112	MoO ₃	HTG-1	UBH15	EB502	EPH31	EPY01	3TPYMB	Alq ₃	Liq	LiF
undoped		-								-	0.8 nm
n-doped	55 nm	-	15 nm	10 nm	3%	25%	3%	10nm	25 nm	33%	-
p-i-n		20%									

3. RESULTS AND DISCUSSION

3.1 Characteristics of electron-only and hole-only devices

In electron-only device, the current density as a function of voltage (J - V) characteristics at the various Liq doped into Alq₃ ratios are shown in Fig. 2. In electron-only device, a rapid increasing in the device current occurs when Liq is doped into the Alq₃ layer. Increasing the current density is seen when a small 10 wt.% Liq doping concentration is introduced into the Alq₃ layer, as compared with the control device. The J - V characteristics of electron-only device are strongly dependent on the doping ratio in the electron transport layer. At the same voltage, the current density increases along with increasing the doping ratio. The highest current density is observed at 33 wt.% Liq doping ratio. The E-1, E-2, E-3 and E-4 device driving voltage at 100 mA/cm² are 8, 5.3, 2.6 and 6.7 V, respectively. These electron-only device J - V characteristics suggest that a certain Liq to Alq₃ doping ratio could improve the co-deposited layer electron transport ability. The advantage of using Alq₃:Liq as the ETL is explained using the electron hopping exchange along with their Lowest Unoccupied Molecular Orbital (LUMO). In a single host device, electrons hop along the LUMO in Alq₃. Since the LUMO-LUMO difference between Alq₃ (3.1 eV) and Liq (3.24 eV) is negligible, subject to their similar LUMO, transport manifolds along with their LUMO are expected to exhibit a certain extent of overlapping after a mixing ratio goes beyond 33 wt.% Liq. Therefore, it is likely that a large energetic disorder between Alq₃ and Liq contributes to the electron hopping, implying that electron hopping among Alq₃ and Liq sites is favorable [19]. The high electron conductivity of Alq₃:Liq might originate from the short electron transport hopping length as compared with the pure Alq₃ ETL. However, the current conduction is reduced dramatically as the doping ratio is further increased to 50 wt.% in the device E-4. This result is attributed to the carrier quenching and deflection effects.

In hole-only device, J - V characteristics at various MoO₃ ratios doped into WH112 are shown in Fig. 3. The J - V characteristics of hole-only device are strongly dependent on the hole transport layer doping ratio.

Comparing with the undoped device, we could see that the low doping strikingly decreases the driving voltage and the J - V characteristics are strongly dependent on the hole injection layer doping ratio. At the same voltage, the current density increases along with increasing MoO_3 doping ratio. The highest current density is observed at 20 wt.% MoO_3 doping ratio, indicating that the p-doping HIL layer conductivity increases due to MoO_3 doping into WHI112. The H-1, H-2, H-3 and H-4 device driving voltages are 5.4, 4.3, 3.8 and 4.2 V, respectively. The results indicate that doping MoO_3 reduces the potential barrier for the hole injection at the ITO interface [20]. The hole-only device current enhancement is attributed to the reduction of resistivity and activation energy, leading to decreased ohmic losses.

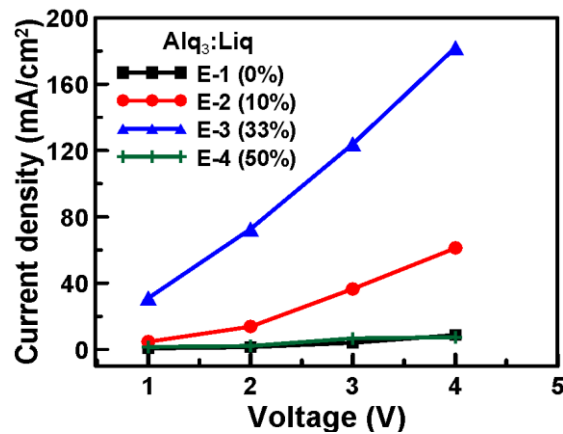


Figure 2: Electron-only J - V characteristics of devices E-1 to E-4.

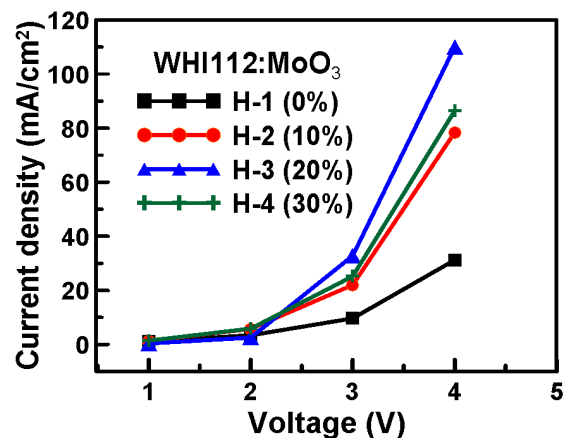


Figure 3: Hole-only J - V characteristics of devices H-1 to H-4.

It is possible that the high holes are transferred from the Highest Occupied Molecular Orbitals (HOMO) in the WHI112: MoO_3 matrix into the HTG-1 HOMO. The MoO_3 (5.3 eV) and HTG-1 (5.4 eV) HOMO levels have closely energetic positions making a charge transfer and energetically favorable process. The hole transfer results in increased charge carrier concentration in the bulk HTL which increases the film conductivity and reduces HTL ohmic losses during the device operation. Through the increased bulk conductivity process, the current density is expected to increase with increasing the doping concentration. However, our devices demonstrate the reduced performance at higher 20% MoO_3 to WHI112 concentration. It is likely that because the heavy doping MoO_3 molecules saturate the layer and escape into the HIL (WHI112: MoO_3)/HTL (HTG-1) interface. This thin MoO_3 layer creates a dipole barrier at the interface with HTL which increases the necessary device driving voltage. It is possible that a high MoO_3 concentration might lead to significant dopant diffusion through the HTL into the EML, causing electroluminescence (EL) quenching in the emissive region. This suggests that the aggregations tend to degrade the device performance.

3.2 Comparison between undoped and n-type devices of hybrid WOLEDs

Hybrid WOLEDs are attracting significant attention due to their unique large-scale fabrication merits for the

solid-state lighting sources. The yellow phosphorescent with the blue fluorescent emitter combination might result in a good compromising among the high efficiency hybrid systems. We know that in conventional devices, such as undoped device, the number of the holes is much greater than the number of the electrons. A surplus of the holes at the HTL/EML interface increases the probability that EML cations are formed, leading to the device degradation rapidly. From the electron-only device, the further experiments should focus on a device with Alq₃: 33 wt.% Liq layer as the n-doping. The n-type device with 33 wt.% Liq doped into Alq₃ as the electron carrier for hybrid WHOLEDs is therefore studied. It is very important to balance out the current supply to the emission zone. This is carried out with 3TPYMB as the electron-blocking and HTG-1 as hole-blocking layers nearby the emission zone. These layers create an additional barrier for the carriers to be injected. The result shows that the recombination and/or emission zone is clearly separated from the area with the high carrier concentration. The structure of this device is ITO/WHI112 (55 nm)/HTG-1 (10 nm)/UBH15: 3 wt.% EB502 (10 nm)/EPH31: 3 wt.% EPY01 (25nm)/3TPYMB (10 nm)/Alq₃: 0 or 33 wt.% Liq (25 nm)/with or without LiF (0.8 nm)/Al (150 nm), where Liq is 0% with LiF for undoped device and Liq is 33 wt.% without LiF for n-type device. Figure 4 shows *L-V* and *J-V* (inset) characteristics of undoped and n-type devices. The n-type device shows the lower operational voltage and higher current density and luminance slopes than the undoped device. It is clearly seen that under the same current density, the n-type device produces higher emissions than the undoped device. This represents the lower electron injection barrier and higher efficiency from the n-type device. Therefore Alq₃:Liq produces the higher electron injection efficiency and higher luminance than LiF. The power efficiency of the two devices is shown in Fig. 5. The power efficiencies of the undoped and n-type devices are 6.4 and 7.81 lm/W at 5 mA/cm², respectively. The driving voltage of the n-type device at 5 mA/cm² is 7.2 V, which is reduced, as compared with the undoped device (11 V). This significant enhancement of performance is attributed to the improved transport conductivity of the n-doping Liq doped into Alq₃ layer. This shows that Liq incorporation into Alq₃ materials could improve device performance, by increasing the electron concentration in Alq₃ films and moving the Fermi level close to the LUMO of Alq₃ [19].

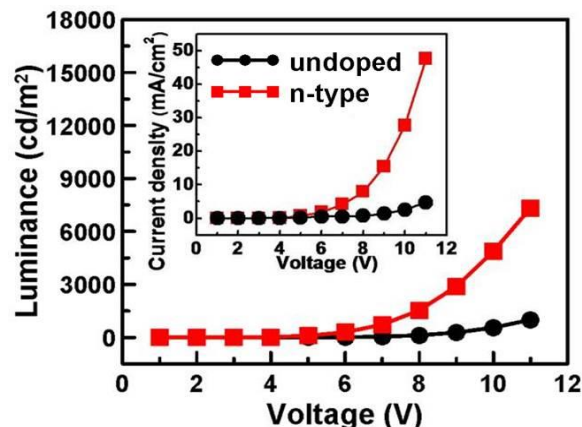


Figure 4: *L-V* and *J-V* (inset) characteristics of undoped and n-type devices.

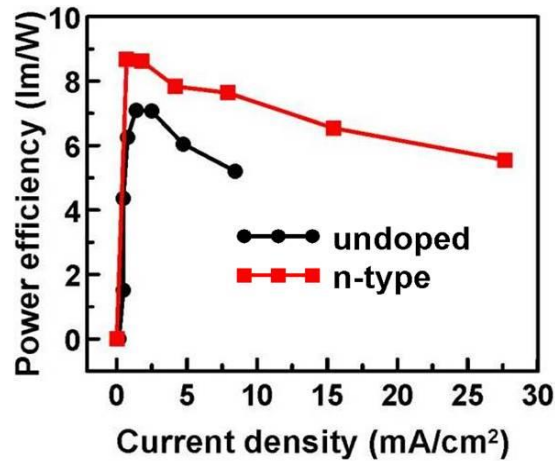


Figure 5: Power efficiency-current density characteristics of undoped and n-type devices.

3.3 Comparison between n-type and p-i-n devices of hybrid WOLEDs

The power efficiency depends on the carrier injection, the transportation and the carrier balance. The p-i-n device is fabricated in which the WH112 layer is doped with 20 wt.% MoO₃ in the ITO/WH112: 20 wt.% MoO₃ (55 nm)/HTG-1 (10 nm)/UBH15: 3 wt.% EB502 (10 nm)/EPH31: 3 wt.% EPY01 (25nm) Alq₃: 33 wt.% Liq (25 nm)/Al (150 nm) configuration. The n-type device and p-i-n device characteristics are displayed in Figs. 6-8.

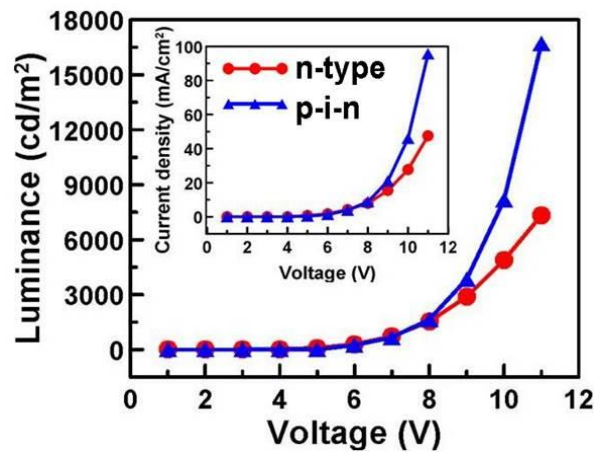


Figure 6: L-V and J-V (inset) characteristics of hybrid WOLEDs of n-type and p-i-n devices.

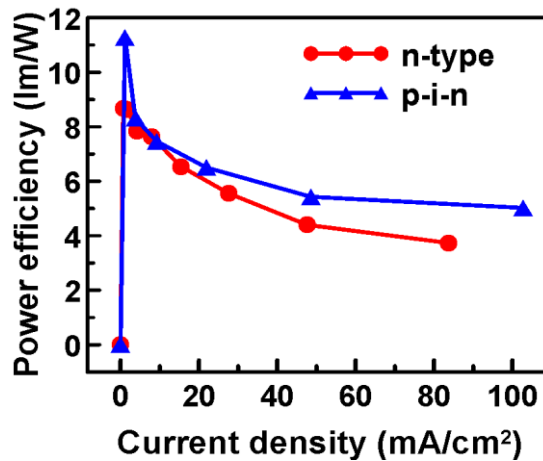


Figure 7: Power efficiency-current density characteristics of hybrid WOLEDs of n-type and p-i-n devices.

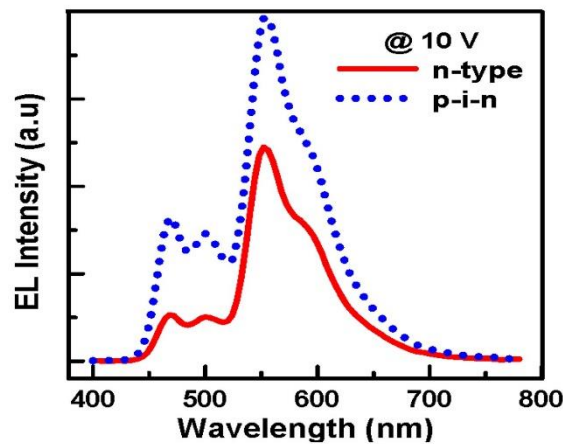


Figure 8: Electroluminescence (EL) spectra of hybrid WOLEDs of n-type and p-i-n devices.

As compared with the n-type device, the *L-V* and *J-V* (inset) curves of the p-i-n device are significantly enhanced, indicating that the device conductivity is improved using the p-i-n structure, as shown in Fig. 6. It is clear that the power efficiency of the p-i-n device is considerably increased as compared with the n-type device as shown in Fig. 7. This indicates the improvement of the p-i-n device conductivity. Table 4 summarizes the data for both devices obtained from the n-type device and p-i-n device at 20 mA/cm². The p-i-n device current efficiency, power efficiency and voltage are improved at 18.6 cd/A, 6.5 lm/W and 8.9 V at 20 mA/cm², respectively, as compared with the n-type device at 18 cd/A, 6.3 lm/W and 9.4 V, respectively.

Table 4: Performance of hybrid WOLED devices at 20 mA/cm².

DEVICES	VOLTAGE	LUMINANCE	YIELD	POWER EFFICIENCY	CIE (X,Y)
n-type	9.4 V	3600 cd/m ²	18.0 cd/A	6.3 lm/W	0.45,0.50
p-i-n	8.9 V	3720 cd/m ²	18.6 cd/A	6.5 lm/W	0.39,0.48

However, the roll-off of the efficiency in the p-i-n device is much smaller than the n-type device. The current and power efficiency of the p-i-n device are maintained with 17.2 cd/A and 5.1 lm/W at 100 mA/cm², it is reduced to 7.5 % and 21 %, respectively. In contrast, the n-type device exhibits the significant reduction of efficiency (14.4 cd/A and 3.8 lm/W at 80 mA/cm²), it is reduced to 20 % and 39.6 %, respectively. On the other hand, for the p-i-n device, the combination of the hole-transport character of 20 wt.% MoO₃ doped into

WHI112 and the electron-transport property of 33 wt.% Liq doped into Alq₃ is contributed to the controlling the holes and the electrons in the light-emitting layer and results in the stable efficiency roll-off hybrid WOLED. Figure 8 shows the EL spectra of the n-type device and p-i-n device. The EL spectrum shows the difference of the peak between these two devices. It is interesting to note that the EL spectrum of the p-i-n device is high as compared with the n-type device. The blue peak spectrum appeared as the electron injection is increased and it shifts the recombination into the blue emission layer. This indicates that the p-i-n device plays a major impact on the hybrid WOLED optical characteristics. The CIE coordinates of the n-type and p-i-n devices are (0.45, 0.50) and (0.39, 0.48), respectively, as shown in Table 1. As the result, the p-i-n device contributes to a certain degree to the good hole-electron balance in the light-emitting layer.

4. CONCLUSIONS

We have presented the hybrid WOLEDs based on p-i-n structure of novel Alq₃:Liq and WHI112:MoO₃ as n-type and p-type, respectively. Current efficiency of 18.6 cd/A, power efficiency of 6.5 lm/W, and driving voltage 8.9 V at a current density of 20 mA/cm² in p-i-n hybrid WOLEDs were obtained. The roll-off of the efficiency in the p-i-n device was much smaller than the n-type and undoped devices. The current and power efficiency of the p-i-n device were maintained with 17.2 cd/A and 5.1 lm/W at 100 mA/cm², it was reduced to 7.5 % and 21 %, respectively. In contrast, the n-type device exhibited the significant reduction of the efficiency (14.4 cd/A and 3.8 lm/W at 80 mA/cm²), it was reduced to 20 % and 39.6 %, respectively. The superior performance was attributed to the high hole and electron ability of WHI112:MoO₃ and Alq₃:Liq, leading to low driving voltage and better electron and hole balance, contributing to enhanced efficiency even at the high current density. Effective carrier balance between the holes and electrons was achieved from the enhanced transport layer conductivity, leading to the device enhanced efficiency.

5. ACKNOWLEDGMENTS

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