An Exciplex Forming Host for Highly Efficient Blue Organic Light Emitting Diodes with Low Driving Voltage

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The exciplex forming co-host with phosphorescent dopant system has potential to realize highly efficient phosphorescent organic light emitting didoes (PhOLEDs). However, the exciplex forming co-host for blue phosphorescent OLEDs has been rarely introduced because of higher triplet level of the blue dopant than green and red dopants. In this work, a novel exciplex forming co-host with high triplet energy level is developed by mixing a phosphine oxide based electron transporting material, PO-T2T, and a hole transporting material, *N*,*N′*-dicarbazolyl-3,5-benzene (mCP). Photo-physical analysis shows that the exciplexes are formed efficiently in the host and the energy transfer from the exciplex to blue phosphorescent dopant (iridium(III)bis[(4,6-difluorophenyl)-pyridinato-*N*,C2′]picolinate; FIrpic) is also efficient, enabling the triplet harvest without energy loss. As a result, an unprecedented high performance blue PhOLED with the exciplex forming co-host is demonstrated, showing a maximum external quantum efficiency (EQE) of 30.3%, a maximum power efficiency of 66 lm W^{-1}, and low driving voltage of 2.75 at 100 cd m^{-2}, 3.29 V at 1000 cd m^{-2}, and 4.65 V at 10 000 cd m^{-2}, respectively. The importance of the exciton confinement in the exciplex forming co-host is further investigated which is directly related to the performance of PhOLEDs.

1. Introduction

Highly efficient blue organic light emitting diodes (OLEDs) have been desired to realize high performance OLEDs for flat-panel displays and solid-state lightings. Over the past two decades, a lot of efforts have been made to demonstrate blue phosphorescent OLEDs (PhOLEDs) with both high external quantum efficiency (EQE) and power efficiency (PE) by tuning the electrical balance, confining the excitons in the emitting layer (EML), and reducing the operating voltage of the devices. For example, a large number of host materials with higher triplet (T_1) level than that of a blue emitter have been synthesized for a blue EML,[1–23] and the mixed host systems or dual EMLs have been utilized to achieve good charge balance and exciton confinement.[24–29] New electron or hole transporting materials with high T_1 level adjacent to an EML have also been introduced to confine excitons in the EML.[5,8,18,30–34] Moreover, the electrical doping in the injection layers and synthesis of organic materials with high mobility and proper energy level were reported to reduce the operating voltage.[14,17,23,31,32,34] As a result, the highly efficient blue PhOLED with a maximum EQE of 30% was reported,[22] but the device showed large efficiency roll-off at high luminance and high driving voltage. There are still challenging issues to realize blue PhOLEDs with high EQE, PE, and low efficiency roll-off at the same time.

Use of an exciplex forming co-host is a promising approach to resolve the issues. The efficient energy transfer from an exciplex forming co-host to phosphorescent dye resulted in high efficiencies approaching the theoretical limits, low driving voltages, and low efficiency roll-offs, simultaneously, with a simple structured device.[34–40] Green, orange and red PhOLEDs and a fluorescent OLED with EQEs over 30% have been reported,[35–42] but these exciplex forming systems are not applicable to blue dopants because of the higher T_1 level of blue dopants than the T_1 level of exciplexes.

The following is required for an exciplex forming co-host for efficient phosphorescent OLEDs: 1) T_1 level of an exciplex has to be lower than those of the consisting molecules in order to confine the excitation energy in the exciplex state, not to be transferred to the consisting molecules, and 2) T_1 level of an exciplex has to be higher than that of a phosphorescent dopant to utilize the energy transfer from the exciplex to a blue dopant. However, seeking for an ideal exciplex system meeting the requirements for a blue dopant seemed to be a challenging issue.[43–45]

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Herein, an efficient exciplex forming host system for blue dopant is reported by mixing a phosphine oxide based electron transporting material, PO-T2T, and a hole transporting material, *N,N′*-dicarbazolyl-3,5-benzene (mCP). This exciplex system satisfies the two requirements described before, that is, the T_1 level of the exciplex (2.64 eV) is lower than the T_1 levels of the consisting molecules of mCP (2.94 eV) and PO-T2T (2.99 eV) and higher than iridium(III)bis[(4,6-difluorophenyl)-pyridinato-*N*,C2′]picolinate (FIrpic) (2.63 eV) so that the exciplex system well confines the excitation energy in the exciplex state, followed by energy transfer to a blue dopant of FIrpic, which was confirmed by the time resolved photoluminescence (PL) measurement. As a result, a high performance blue PhOLED was realized using the exciplex forming co-host doped with FIrpic with a maximum EQE of 30.3%, PE of 66 lm W^{-1}, and low driving voltage of 2.75 at 100 cd m^{-2}, 3.29 V at 1000 cd m^{-2}, and 4.65 V at 10 000 cd m^{-2}, respectively. The results were attributed to novel exciplex forming system as well as PO-T2T with low-lying lowest unoccupied molecular orbital (LUMO) level of 3.5 eV which boosts the efficient electron injection and transport under the low external voltage bias.

2. Results and Discussions

2.1. Molecular Structures and Energy Levels of Organic Materials

Figure 1a show the molecular structures of PO-T2T and mCP. mCP was purchased from Nichem Fine Technology and utilized as received without further purification. PO-T2T was synthesized and the detailed information on the synthesis, optical and electrical properties are described in the previous report.[46] The energy level of PO-T2T was additionally measured by using ultraviolet photoelectron spectroscopy and inverse photoelectron spectroscopy shown in Figure 1b, respectively. The ionization potential (I.P.) or highest occupied molecular orbital (HOMO) level of PO-T2T was determined to be –7.5 eV and the electron affinity (E.A.) to be –2.9 eV, respectively. The measured energy gap of 4.6 eV corresponds to the transport gap, E_g^T, which is defined as the minimum energy to form a separated electron-hole pair and the value is 0.6 eV larger than the optical band gap, E_g^{opt}, of 4.0 eV due to the exciton binding energy under optical excitation. The difference between E_g^T and E_g^{opt} is reasonable in comparison with the reported one.[47,48] In this work, the LUMO level of the material was assigned as –3.5 eV based on E_g^{opt}. It is expected that the low E.A. and/or low-lying LUMO level of PO-T2T boosts easy electron injection from the electrode under low external voltage bias and the high electron mobility of PO-T2T, ~10^{-3} cm^2 V^{-1} s^{-1},[44] contributes to reduce driving voltage and improve electrical balance in the devices. The HOMO and LUMO level of mCP were known to be –6.1 eV and –2.4 eV, respectively.[13,18,26]

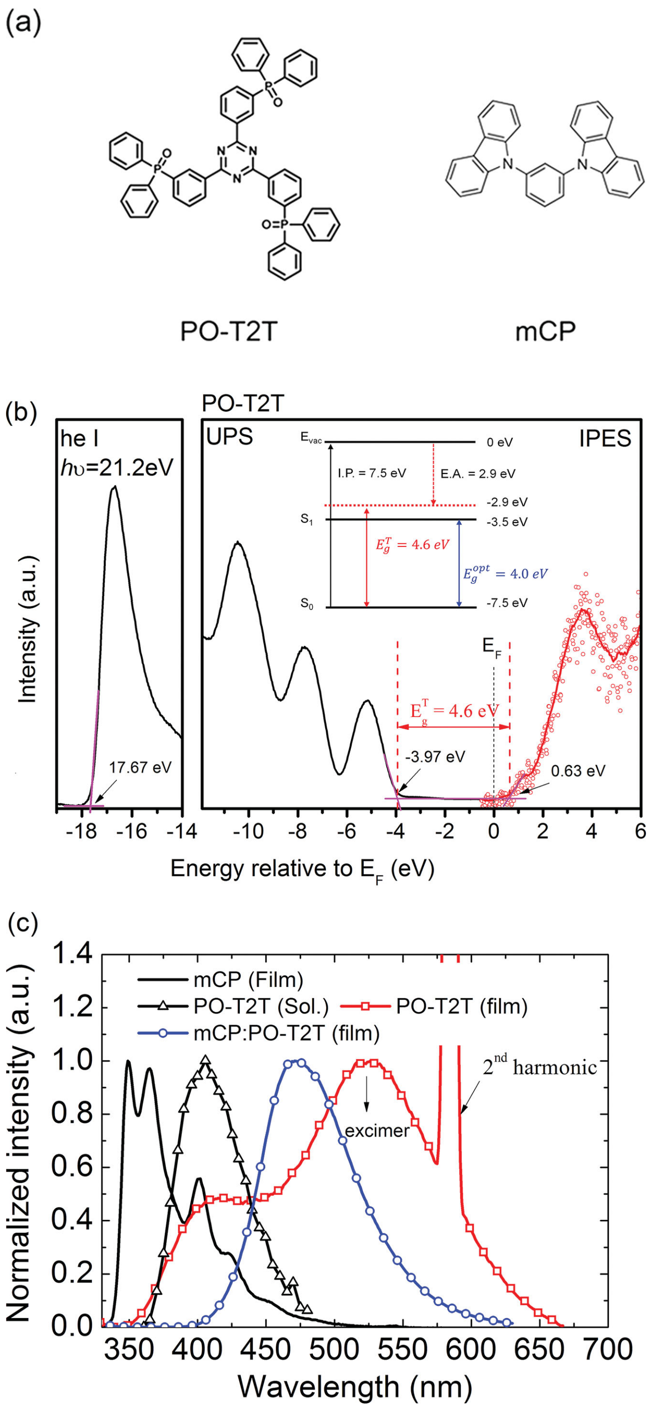

Figure 1. a) Molecular structure of PO-T2T and mCP, b) ultraviolet photoelectron spectroscopy (UPS) and inverse photoelectron spectroscopy (IPES) spectra of PO-T2T thin film and energy level of PO-T2T, and c) Normalized PL spectra of the mCP film (solid line), PO-T2T solution (line with triangle), PO-T2T film (line with square), and mCP:PO-T2T co-deposited film (line with circle).

2.2. Photophysical Analysis of the Exciplex with and without FIrpic

The formation of the exciplex in the mCP:PO-T2T film (1:1 molar ratio) was confirmed from the PL spectra of the mCP, PO-T2T and mCP:PO-T2T (molar ratio 1:1) co-deposited films shown in Figure 1c. The PL spectrum of the mCP film was consistent with the reported one,[28] but that of the PO-T2T was different from the reported solution PL.[46] The PL spectrum of the PO-T2T film showed two peaks except another peak near 580 nm originating from 2nd harmonic of the excitation source. The peak at 410 nm is from the singlet emission of PO-T2T which coincides with the peak from the PO-T2T solution. The other peak at 525 nm is assigned as the excimer emission of the PO-T2T film from the red shifted and featureless broad emission spectrum which is not observed in the PO-T2T solution. Interestingly, the mCP:PO-T2T co-deposited film showed a featureless PL spectrum with the peak position at 470 nm (2.64 eV) red shifted from those of mCP and PO-T2T. The value of 2.64 eV is close to the energy difference between the HOMO level of mCP and the LUMO level of PO-T2T, indicating that the emission is from the exciplex formed between the mCP and PO-T2T molecules under the photo excitation. No excimer emission from PO-T2T indicates that mCP and PO-T2T are well mixed in the co-deposited film.

Figure 2 shows the time resolved PL spectra of the mCP:PO-T2T co-deposited film at different delay times, and at two different temperatures of 300 K and 35 K, respectively. At 300 K, the peak position of the PL spectra progressively shifted to longer wavelength from 464 nm to 477 nm as the delay time increases as shown in Figure 2a. The delayed emission was clearly observed even after 10 μs since the exciplex has broad energy distributions and small energy splitting between the singlet and triplet charge transfer (CT) exciplex states, enabling the long delayed emission through efficient intersystem crossing (ISC) and reverse intersystem crossing (RISC) between the singlet and triplet charge transfer exciplex states.[49–54] The singlet and triplet CT states simultaneously contributed to the delayed emission so that it is difficult to distinguish their contribution to the PL spectra. The time dependent peak shift of the PL spectra in the exciplex might be related to the relaxation of the exciplexes to a stable geometry.[51] At 35 K, the smaller peak shift with delay time was observed than the peak shift at 300 K as shown in Figure 2b due to the reduction of the molecular movements at low temperature. No other peaks observed even at long delayed times and at low temperature suggest that the excitation energy is well confined in the exciplex due to lower T_1 level (2.64 eV) of the exciplex than that of mCP (2.93 eV) and PO-T2T (2.99 eV). Triplet emission from the host molecule(s) will appear at low temperature if the T_1 level of the host molecule(s) is lower than the exciplex triplet level due to the energy transfer from the exciplex to the host molecules.[43,52,54,55]

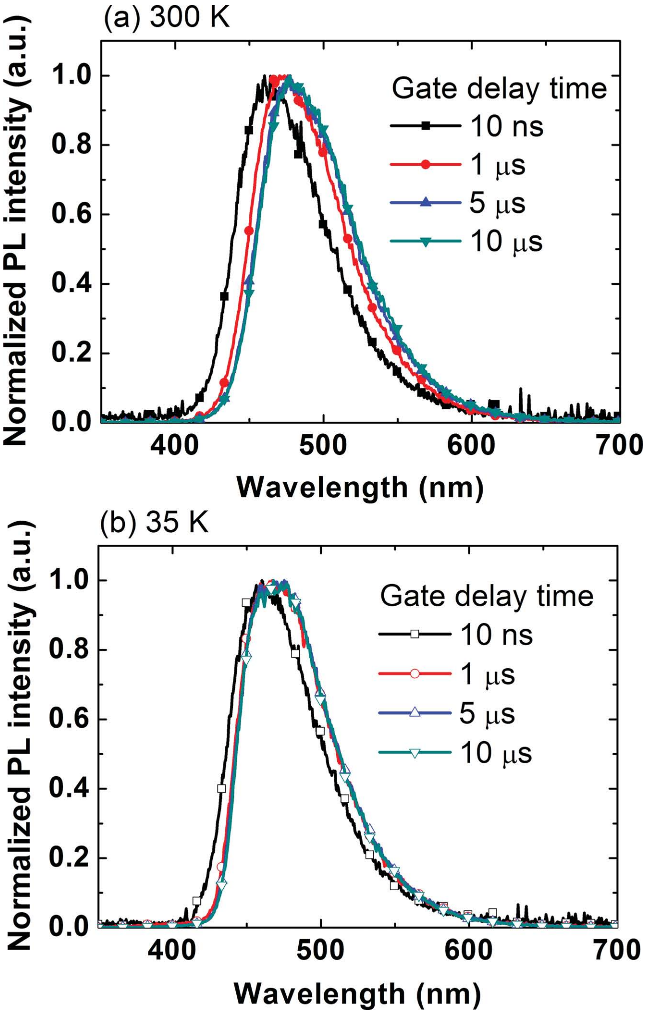

Figure 2. Time resolved photoluminescence (PL) spectra of the mCP:PO-T2T co-deposited film at different gate delay time from 10 ns to 10 μs a) at 300 K and b) 35 K, respectively.

The importance of the exciton confinement in the exciplex is more clearly manifested when a blue dopant, FIrpic, is doped in the exciplex host. **Figure 3** show the time resolved PL spectra of two films, mCP:PO-T2T:10% FIrpic and mCP:B3PYMPM:10% FIrpic on a quartz substrate. The energy levels of the consisting materials and the exciplexes are displayed in Figure 3a. The energy level of the mCP:B3PYMPM exciplex is higher than that of the mCP:PO-T2T exciplex due to the higher LUMO level of B3PYMPM (−3.2 eV) than PO-T2T (−3.5 eV). Despite of the energy level differences between the two systems, the similar PL spectra were observed at 300 K independent of the delayed time as shown in Figure 3b and 3c. It seems that the PL spectra are only originated from the FIrpic emission, resulting from the energy transfer from the host to FIrpic because of the lowest T_1 level of FIrpic (2.63 eV) among those of the monomers (2.93 eV of mCP, 2.99 eV of PO-T2T, and 2.7 eV of B3PYMPM, respectively) and exciplexes (2.64 eV of mCP:PO-T2T and 2.9 eV of mCP:B3PYMPM).

However, apparently different PL spectra were obtained between the two systems at 35 K as shown in Figure 3d,e. The FIrpic doped mCP:PO-T2T film showed the PL spectra originated from the FIrpic emission without other contribution independent of the delay time, indicating the efficient energy transfer from the mCP:PO-T2T host to FIrpic. In contrast, the time dependent PL spectra of the mCP:B3PYMPM:10% FIrpic film (Figure 3e) showed an additional peak at 420 nm at the delay times of 2 and 100 ns. The intensity of the extra PL peak gradually decreased as the delay time increased from 2 ns to 100 ns and disappeared after 100 ns. Comparison of the spectrum with the exciplex emission of mCP:B3PYMPM clearly indicates that the peak at 420 nm originates from the exciplex emission. The other two long wavelength peaks in the low temperature PL spectrum of mCP:B3PYMPM are from the triplet emission of B3PYMPM which is attributed to the energy transfer from mCP:B3PYMPM to B3PYMPM due to

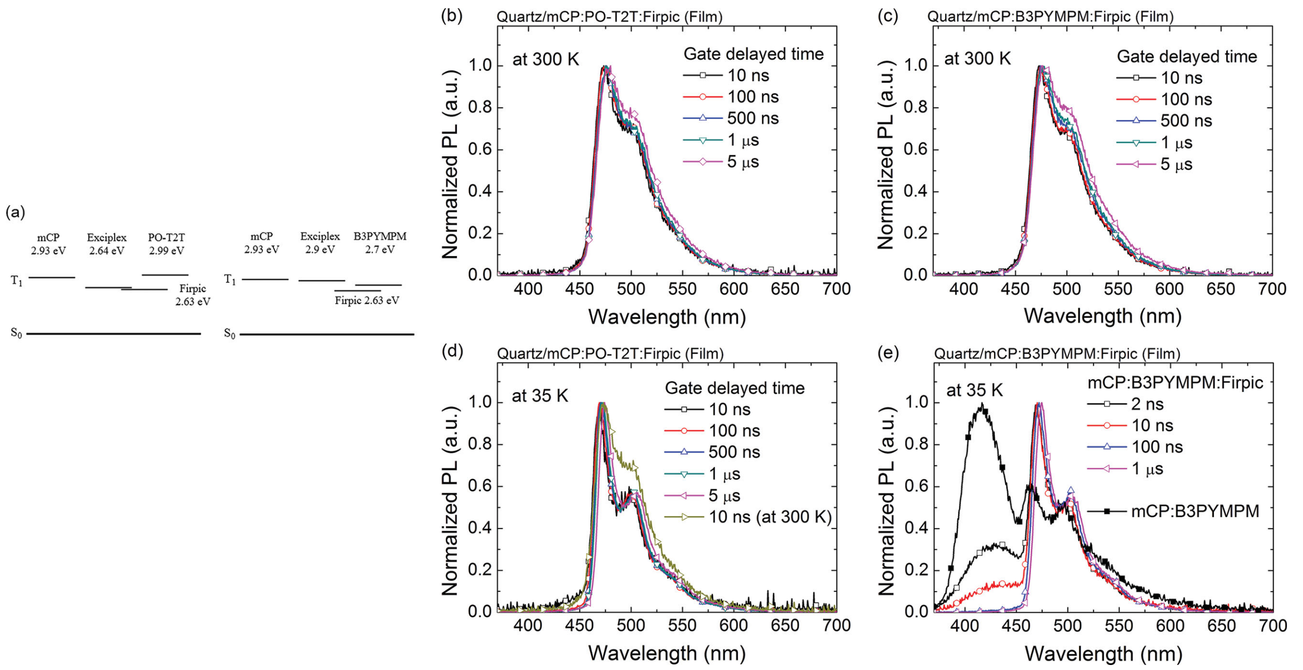

Figure 3. a) Comparison of triplet (T_1) energy level of mCP, PO-T2T, B3PYMPM, and the exciplexes, respectively. Time resolved photoluminescence (PL) spectra of b) the mCP:PO-T2T:10% FIrpic film at 300 K, c) the mCP:B3PYMPM:10% FIrpic film at 300 K, d) the mCP:PO-T2T:10% FIrpic film at 35 K, e) the mCP:B3PYMPM:10% FIrpic film at 35 K at different gate delay time.

the lower T_1 level of B3PYMPM than that of exciplex as shown in Figure 3a. Namely, the different behaviors between the mCP:PO-T2T and mCP:B3PYMPM can be understood based on the relative magnitude of the T_1 levels of the exciplex and the energy levels of the consisting materials. In the mCP:PO-T2T:FIrpic system, the T_1 levels are mCP ≈ PO-T2T >> exciplex > FIrpic. Therefore, the exciplex energy will be transferred to FIrpic if the energy transfer is efficient and there is no emission from the host materials. In contrast, in the mCP:B3PYMPM:FIrpic system, the T_1 levels are mCP > exciplex > B3PYMPM > FIrpic. There is a chance now for the exciplex energy to be transferred to B3PYPM as well as to FIrpic. Hence, it is not surprising to observe the PL emission from B3PYMPM in the mCP:B3PYMPM:FIrpic system. The energy transfer from the host to FIrpic does not seem to be very efficient in this system. Understanding of the energy transfer processes in the systems require further experiments in a picosecond or femtosecond time scale. It will be reported separately after investigating various systems. Given the time resolved PL emission spectra in Figure 3, it is inferred that all excitation energy in the host is not perfectly transferred to FIrpic in the mCP:B3PYMPM:FIrpic film. In contrast, the excitation energy is well confined in the exciplex composed of mCP:PO-T2T and singlet as well as triplet exciplexes are efficiently transferred to FIrpic without energy loss. Therefore, a stable and high performance blue OLED is anticipated using the mCP:PO-T2T host.

2.3. Blue Emitting Phosphorescent OLED

Figure 4a shows a schematic diagram of the blue PhOLED structure with the energy levels of the organic layers of the optimized blue PhOLED. The optimized device structure of the blue PhOLED is as follows: ITO (70 nm)/6% ReO_3:mCP (45 nm)/mCP (15 nm)/mCP:PO-T2T:10% FIrpic (30 nm)/PO-T2T (20 nm)/4% Rb_2CO_3:PO-T2T (25 nm)/Al (100 nm). The optimized structure was selected based on the previous report.[43] The current density–voltage–luminance (*J*–*V*–*L*) characteristics of the blue PhOLED are displayed in Figure 4b. The current injection started at low voltage of 2.0 V and the device was turned on at 2.4 V at 1 cd m^{-2} and reached 100, 1000 and 10000 cd m^{-2} at 2.75 V, 3.29 V and 4.65 V, respectively. The driving voltages of the device are the lowest voltages among the FIrpic-based blue PhOLEDs over 25% of EQE,[22,25,34,43,44] reported up to now and the turn on voltage is lower than the photon energy of the emitted light devided by the unit charge, attributing to the diffusion current of the device from the metal reservoir to organic layers under the low external voltage bias.[38] The low turn–on and driving voltages indicate that the charge injection in the device is almost Ohmic. The reduction of charge injection barrier by the exciplex forming co-host as well as the low-lying LUMO level of PO-T2T (3.5 eV) also contributed to easy electron injection from the electrode reducing the driving voltage of the device. Angle dependent emission intensity distribution is shown in the inset of Figure 4b which is narrower than the Lambetian distribution by the factor of 0.94. The calibrated EQEs and PEs of the device using the emission pattern are shown in Figure 4c. The device exhibited a high maximum EQE of 30.3% and low efficiency roll-offs with the EQE of 29.4% at 1000 cd m^{-2}, 24.1% at 5000 cd m^{-2} and 19.9% at 10,000 cd m^{-2}. The detailed characteristics of the device are listed in **Table 1**. The maximum EQE value is very close to the simulation result based on the classical dipole model performed under the assumption of no electrical loss.[43] This good match indicates

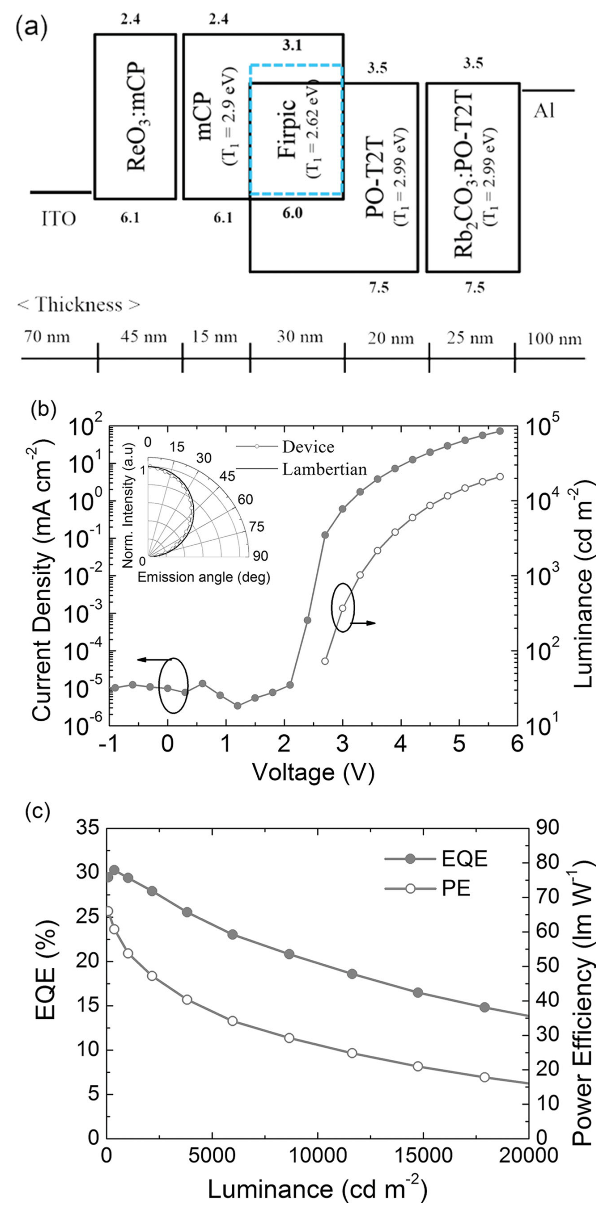

Figure 4. a) Schematic diagram of the device structure with the energy levels of the organic layers, b) Current density–voltage–luminance (*J*–*V*–*L*) characteristics, and c) Calibrated external quantum efficiency (EQE) and power efficiencies of the optimized blue PhOLED as a function of luminance.

that almost all the injected charges are recombined without electrical loss in the device. The low driving voltage and high EQE lead to high PEs with high maximum PE of 66 lm W^{-1} at 72.4 cd m^{-2}, 54.1 lm W^{-1} at 1000 cd m^{-2}, and 27.3 lm W^{-1} at 10 000 cd m^{-2}, which is the highest values among the FIrpic based blue PhOLEDs.

3. Conclusion

In summary, a high performance blue PhOLED showing low driving voltages of 2.75 V, 3.29 V and 4.65 V at 100, 1000 and 10 000 cd m^{-2}, respectively, a high maximum EQE of 30.3%, and maximum PE of 66 lm W^{-1} was demonstrated using an exciplex forming co-host composed of mCP and PO-T2T which has low-lying LUMO level of −3.5 eV. The time resolved PL spectra confirmed that the exciplex was formed by mixing mCP:PO-T2T and the excitation energy was well confined in the exciplex state. In addition, the energy transfer from the exciplex to FIrpic was also efficient without energy loss. All the combined effects resulted in the high performance OLEDs.

4. Experimental Section

A blue PhOLED with exciplex forming co-host and FIrpic were fabricated by thermal deposition under a base pressure of < 5×10^{-7} Torr. Organic layers and metal cathode were deposited onto 70 nm-thick indium tin oxide (ITO) pre-patterned glass substrates. Prior to the deposition, the ITO patterned substrates were cleaned by isopropyl alcohol and acetone, and then the substrates were exposed to a UV-ozone flux for 10 min. The PL spectra of organic films on a quartz substrate were measured by using a spectrofluorometer (Photon Technology International, Inc.) incorporated with a monochromator (Acton Research Co.) and a monochromator-attached pulsed xenon flash lamp. The wavelength of 290 nm and 325 nm were selected to excite the organic layer, respectively. For the time resolved PL measurement, a pulsed Nd:YAG laser (355 nm) was used as the excitation source and an intensified charge-coupled device (iCCD) was utilized as the optical detector. The low-temperature PL spectra of organic films were obtained by concentrating the light emitted from the film in a cryostat (SHI Cryogenics, Inc.) with a cooling system. The current density–voltage–luminance (*J*–*V*–*L*) characteristics were measured using a Keithley 2400 programmable source meter and SpectraScan PR650 (Photo Research) radiometer as well as a Minolta CS100. The EQE and power efficiency of the OLEDs were calculated by measuring the emission pattern of angle-dependent electroluminescent intensity.

Table 1. Summary of the performance of the blue PhOLED.

			At 100 cd m^{-2}	At 1000 cd m^{-2}	At 5000 cd m^{-2}	At 10 000 cd m^{-2}
Experimental results	Driving Voltage [V]	2.4 (Turn-on)[a]	2.75	3.29	4.07	4.65
	Power Efficiency [lm W^{-1}]	66.0 (Maximum)	65.6	54.1	37.0	27.3
	EQE [%]	30.3 (Maximum)	29.6	29.4	24.1	19.9
Simulation result[48]	EQE [%]	29.9[b]				

[a] Operating voltage at 1 cd m^{-2}; [b] The simulation result of the EQE value was obtained using the measured horizontal dipole ratio of 76 ± 1% and the photoluminescence quantum yield (PLQY) of 97 ± 3% under the assumption of no electrical loss.[48]

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