



Soluble processed low-voltage and high efficiency blue phosphorescent organic light-emitting devices using small molecule host systems

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ABSTRACT

We report low voltage driving and highly efficient blue phosphorescence organic light emitting diodes (PHOLEDs) fabricated by soluble process. A soluble small molecule mixed host system consisting of hole transporting 4,4',4'' tris(N-carbazolyl)triphenylamine (TCTA) and bipolar carrier transporting 2,6-bis(3-(carbazol-9-yl)phenyl)pyridine (26DCzPPy) exhibits high solubility with smooth surface properties. Moreover, this small molecule host shows the smoothest morphological property similar to a vacuum deposited amorphous film. A low driving voltage of 5.4 V at 1000 cd/m² and maximum external quantum efficiency 14.6% obtained in the solution processed blue PHOLEDs are useful for large area low cost manufacturing.

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1. Introduction

Solution processed phosphorescence organic light emitting diodes (PHOLEDs) have been a great interest for displays and lighting applications due to their simple and low cost fabrication process and large area display applications [1,2]. Good device performances of PHOLEDs by solution process had been reported by several groups [3–5]. Although several solution processed PHOLEDs with very good performances were reported, high driving voltage, low efficiency, and short lifetime still remain crucial issues. Consequently, investigations on soluble materials, film characteristics of solution process, and device structures are very important and still open issues.

In order to achieve a low driving voltage and high efficiency in solution processed PHOLEDs, the solution processed emitting layer (EML) is more vital than other layers like carrier transport layers since carriers movements are mainly limited by the host-dopant situation. Usually, the host materials should have wide band gap and high triplet state energy for exciton confinement [6]. Also, the balanced

hole and electron movement within an emitting layer is very important for the good charge balance [7]. In addition to these general requirements, solubility and amorphous-like film characteristics without crystallization are also critical factors to get the high efficiency device.

Many groups made attempts to explore the ways for improving the performance of solution processed blue PHOLEDs [8–10] in recent years. Mixed host system [3,11,12] such as a polyvinylcarbazole (PVK) hole transporting and 1,3-bis[(4-tert-butylphenyl)-1,3,4-oxadiazolyl]phenylene (OXD-7) electron transporting host system is a well known approach for good charge balance in an EML. In mixed host system, the balanced carrier transportation, minimization of carrier trapping by dopants, and decreased crystallization by mixing two hosts are of great beneficial to solution processed PHOLEDs.

Based on this approach, the high efficiency blue PHOLEDs over 15% external quantum efficiencies (EQEs) by solution processing were reported [3,5]. Typically, non-conjugated polymer of PVK has high triplet energy and good film formation characteristics. Hence it used as hole transporting host due to a reasonable HOMO value of 5.8 eV. However, the PVK host always shows high driving voltage owing to its low carrier mobility [13]. Other small

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Table 1
Performance summary of soluble processed blue PHOLEDs.

Host materials	Driving voltage (V) @1000 cd/m ²	EQE (%)	Current efficiency (cd/A)	Power efficiency (lm/W)	CIE coordinates	Ref.
PVK: OXD-7	7.5	–	22.0(max)	14.5(max)	0.17,0.37	[11]
PVK: OXD-7	8.0	9.03(max)	20.3(max)	–	–	[12]
PVK: OXD-7	10.0	15.5(max)	28.3(max)	–	0.14,0.28	[3]
TBCPF: OXD-7	12.4	–	12.5(max)	–	0.22,0.44	[8]
DTCFPB	10.5	–	24.0(max)	8.8(max)	0.14,0.23	[9]
PVK/SPP013	7.5	14.1(max)	30.4(max)	–	0.17,0.42	[4]
SimCP2	6.5	15.8(at 1000 cd/m ²)	31.1(at 1000 cd/m ²)	–	–	[5]
PTC: OXD-7	7.5	11.9(max)	25.7(max)	–	0.16,0.38	[10]

Table 2
Performance summary of soluble processed Devices A–D.

Devices	Driving voltage (V)	EQE (%)	Current efficiency (cd/A)	Power efficiency (lm/W)	CIE coordinates
Device A	6.0(1000 cd/m ²)	12.0(max)	21.0(max)	13.0(max)	0.16,0.33
Device B	5.6(1000 cd/m ²)	12.1(max)	21.8(max)	12.7(max)	0.17,0.34
Device C	5.4(1000 cd/m ²)	14.6(max)	25.1(max)	15.7(max)	0.16,0.34
Device D	6.0(1000 cd/m ²)	9.2(max)	15.9(max)	8.8(max)	0.15,0.32

molecule host systems with both hole and electron carrier transporting characteristics were also tried to address this issue. However, the solution processed blue PHOLEDs still have high driving voltage due to its too wide band-gaps of host materials. Table 1 shows the reported driving voltages and efficiencies in soluble processed blue PHOLEDs. All reported blue PHOLEDs devices have driving voltages of more than 6.5 V.

In this paper, low driving voltage and high efficiency soluble blue PHOLEDs were fabricated by using the small molecule mixed host system. The mixed host with a blue dopant, which possesses a good solubility and morphological uniformity without crystallization issues, has an energetically minimized charge trapping situation by dopant molecules and high carrier mobility characteristics for holes and electrons. As a consequence, we achieved a driving voltage as low as 5.4 V (at 1000 cd/m²) with a maximum current efficiency of 25.1 cd/A and a maximum quantum efficiency of 14.6% in the optimized solution processed blue PHOLEDs.

2. Experimental details

We have fabricated solution processed blue PHOLEDs using 1,3,5-tri[(3-pyridyl)-phen-3-yl]benzene (TmPyPB) and 2,2',2''-(1,3,5-benzinetriyl)-tris(1-phenyl-1-H-benzimidazole (TPBI) electron-transporting materials. 4,4',4''-tris(carbazol-9-yl)-triphenylamine (TCTA) and 2,6-bis(3-(9H-carbazol-9-yl)phenyl)pyridine (26DCzPPy) were used as the single or mixed host with bis(3,5-difluoro-2-(2-pyridyl)phenyl-(2-carboxypyridyl)iridium(III) (Flrpic) as the blue dopant for an EML. Fig. 1 shows the molecular structure with the energy levels of materials.

To fabricate the blue PHOLEDs, a patterned indium-tin oxide (ITO) with a 150 nm thickness glass substrate having an emission area of 4 mm² with a sheet resistance of 10–12 Ω/□ were used. A line pattern of ITO and an insulating

layer to make active areas were formed by the photolithography process. The ITO glass substrate was cleaned by sonication in isopropyl alcohol (IPA) and then rinsed in deionized water. Subsequently, the ITO-coated glass substrate was subjected to UV-ozone treatment prior to device fabrication. The PEDOT:PSS (CLEVIOSTM AI4083 (H.C. Stack)) as a hole injection layer was spin-coated on the ITO substrates and dried using a hot plate at 120 °C for 20 min to remove solvent. The emission layer dissolved in chloro-benzene was spin-coated and dried at 120 °C for 10 min. In case of mixed host, a blending ratio was fixed as 1:1 [14]. All solution depositions were performed in a glove box with nitrogen ambient at room temperature. After coating EML, TmPyPB or TPBI as an electron transporting layer (ETL) was thermally deposited at a base pressure of 10^{−7} torr and then LiF and Al cathode was deposited without breaking the vacuum.

The current density–voltage (*J*–*V*) and luminance–voltage (*L*–*V*) characteristics of the fabricated devices were collected using a Keithley 2635A and Minolta CS-100A luminance-meter. Also, the electroluminescence (EL) spectra and CIE color coordinates were obtained using a Minolta CS-1000A. For investigation of film surface morphology, atomic force microscopy (AFM) of Park systems XE-100 was used.

For the measurement of photoluminescence (PL) quantum yields, TCTA, 26DCzPPy, and TCTA: 26DCzPPy films doped with 10% Flrpic were made by spin coating and vacuum deposition process, respectively. Each layer thickness was adjusted to be 40 nm. UV–vis absorption spectra of these films were recorded with a Sinco S-3100 UV–vis spectrophotometer. Relative photoluminescence (PL) quantum yields of these films were measured by Jasco FP-6000 spectrophotometer. As a standard ($\Phi = 85\%$) sample [15], 1,3-bis(9-carbazolyl)benzene (mCP) film doped with 6% Flrpic by vacuum deposition process was used. Excitation wavelength of each film was 337 nm.

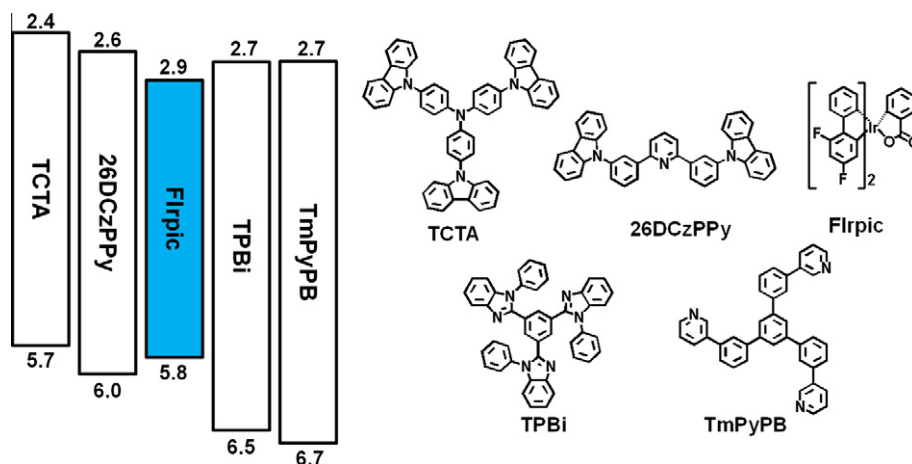


Fig. 1. Molecular structures and energy level diagrams of used materials in blue PHOLEDs.

3. Result and discussion

The TCTA is a well known hole transporting material with a good hole mobility of $3.0 \times 10^{-4} \text{ cm}^2/\text{Vs}$ and suitable triplet energy of 2.8 eV for blue PHOLEDs. The 5.7 eV HOMO energy of TCTA is most suitable for hole transportation in the EML [16]. On the other hand, 26DCzPPy was reported as a good host material in vacuum deposited blue PHOLEDs with good triplet energy of 2.7 eV. It has enough hole and electron transport capabilities with $2.0 \times 10^{-5} \text{ cm}^2/\text{Vs}$ hole mobility and $2.0 \times 10^{-5} \text{ cm}^2/\text{Vs}$ electron mobility [17]. Based on suitable host materials, we designed four kinds of simple structure PHOLEDs as follows:

Device A: ITO/PEDOT: PSS (60 nm)/TCTA: Flrpic (10 wt.%, 60 nm)/TmPyPB (30 nm)/LiF (1 nm)/Al (100 nm).

Device B: ITO/PEDOT: PSS (60 nm)/26DCzPPy: Flrpic (10 wt.%, 60 nm)/TmPyPB (30 nm)/LiF (1 nm)/Al (100 nm).

Device C: ITO/PEDOT: PSS (60 nm)/TCTA: 26DCzPPy: Flrpic (10 wt.%, 60 nm)/TmPyPB (30 nm)/LiF (1 nm)/Al (100 nm).

Device D: ITO/PEDOT: PSS (60 nm)/TCTA: 26DCzPPy: Flrpic (10 wt.%, 60 nm)/TPBi (30 nm)/LiF (1 nm)/Al (100 nm).

Device A has the TCTA as hole dominated host whereas the Device B uses the 26DCzPPy as bi-polar type host. Usually, the wide energy gap host materials characterized by the high singlet–triplet exchange energies of $>0.5 \text{ eV}$ were used to make the blue PHOLEDs [19]. In such cases, significant differences in the HOMO and/or LUMO energy levels between dopant and host materials occur. Thus, the dopant molecules act as deep traps site for electrons and holes in the EML, causing an increase in the drive voltage of the PHOLED [18]. Consequently, the selection of suitable host candidates was a critical issue in fabricating high efficiency blue PHOLEDs. In order to decrease the driving voltage value and achieve the wider emission zone in the device, TCTA: 26DCzPPy mixed host system was considered for

Device C. Our mixed host system has an energetically negligible charge trapping situation by dopant molecules. Device D was designed with well known TPBi electron transporting layer for better comparison to the effectiveness of TmPyPB material. The HOMO and LUMO levels of TmPyPB at 6.7 and 2.7 eV, respectively, are suitable for hole blocking with efficient electron injection and transportation. In addition, the triplet energy (2.9 eV) of the TmPyPB is higher than that of the Flrpic (2.6 eV) and is high enough to suppress the triplet exciton quenching of the Flrpic by the TmPyPB [19]. On the other hand, TPBi has inadequate triplet energy as about 2.7 eV to confine triplet excitons at EML [20]. Triplet energy levels (2.7–2.9 eV) of all materials used in this study except PEDOT:PSS and TPBi seem to be excellent for exciton confinement of Flrpic (2.6 eV). In blue PHOLEDs, the emission zone was reported to be located at near the position of electron transporting layer. The HTL interlayer approach with high triplet energy materials such as TCTA was also studied and evaluated. However, no big differences as far as the device performances in these four devices due to no effectiveness of emission zone shift were noticed.

Fig. 2a shows the luminance–voltage characteristic of fabricated blue PHOLEDs. All device performances are summarized in Table 2. The driving voltages (at $1000 \text{ cd}/\text{m}^2$) of 6.0, 5.6, 5.4, and 6.0 V in Devices A–D are reported, respectively. The driving voltage of the 26DCzPPy host device (Device B) is lower than that of the TCTA host device (Device A). As shown in Fig. 1, hole trapping at the dopant is not serious as the HOMO gap between host and dopant is within 0.2 eV. The LUMO gap between TCTA and Flrpic is 0.5 eV while 26DCzPPy and Flrpic has 0.3 eV. Thus, the electrons could more easily move in 26DCzPPy host system, resulting lower driving voltage in Device B. Among three types of host systems, mixed host device (Device C) showed the lowest driving voltage because of the utilization of appropriate energy levels for hole and electron transportation with good hole and electron mobility of mixed system. The higher driving voltage of Device D than Device C is the consequence of the low electron mobility of TPBi ($\sim 10^{-5} \text{ cm}^2/\text{Vs}$) [20] than TmPyPB ($\sim 10^{-3} \text{ cm}^2/\text{Vs}$) [19].

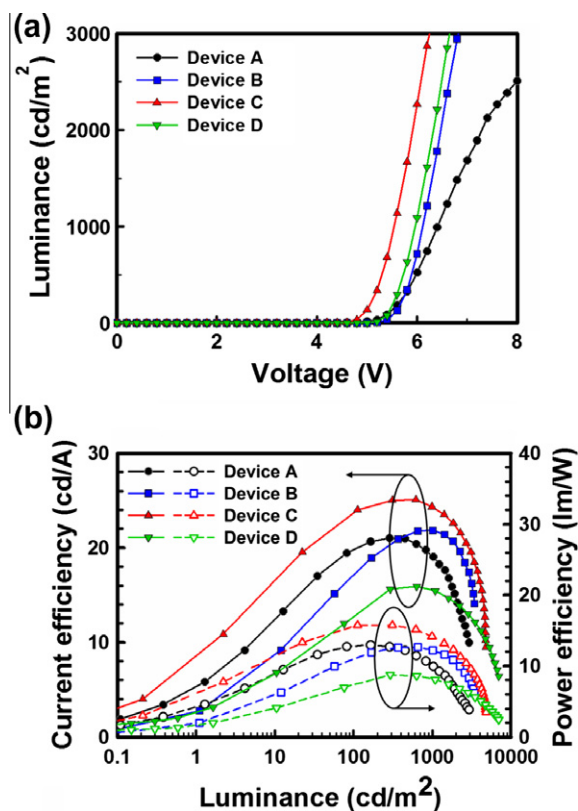


Fig. 2. (a) Luminance versus voltage characteristics and (b) current efficiency and power efficiency versus luminance characteristics of fabricated blue PHOLEDs devices.

The current and power efficiencies curves of blue PHOLEDs are shown in Fig. 2b. The current and power efficiencies at luminance of 1000 cd/m² of single host Devices A and B are 19.0, 21.8 cd/A and 10.0, 12.2 lm/W, respectively. The current efficiency of Device B was higher than that of Device A. Bipolar property of the 26DCzPPy with good HOMO and LUMO values enabled the fabrication of device with high power efficiency. The Device A have relatively high current efficiency roll-off characteristic. The unbalanced hole and electron currents make the recombination zone narrower, resulting triplet–triplet exciton quenching due to local high density of triplet excitons. The 26DCzPPy host provides a wide recombination region by facile movement of charge carriers, which results in better current efficiency roll-off. The Device C fabricated using mixed host showed higher efficiency than the Devices A and B. The maximum current efficiency of 25.1 cd/A and maximum power efficiency of 15.7 lm/W are achieved. The high efficiency of the Device C can be explained as good charge balance in the emitting layer. Holes and electrons can be efficiently injected to the TCTA:26DCzPPy mixed host emitting layer. The efficient hole and electron injection improves the charge balance in the emitting layer, resulting in the best value of efficiency. The Device D shows the lowest efficiency due to the exciton quenching issue by very similar triplet energies of Flrpic and TPBI.

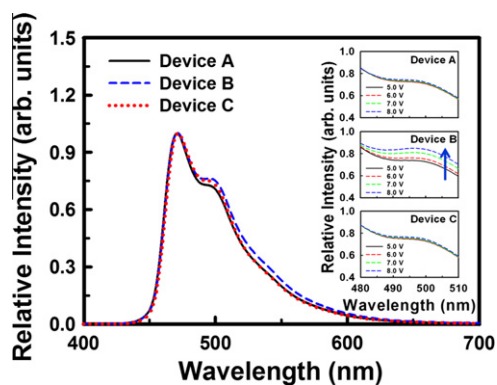


Fig. 3. The normalized EL spectra of Devices A–C at 3 mA/cm². The inset shows the EL spectra with variation of voltage from 5 to 8 V.

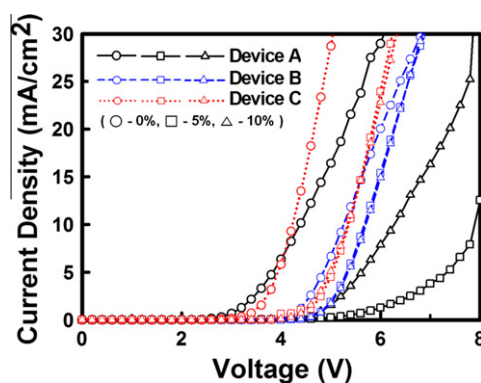


Fig. 4. Current–voltage characteristics of Devices A–C with 0%, 5%, and 10% doping concentrations.

As shown in Fig. 3, all devices exhibit a Flrpic emission centered at 470 nm with a shoulder peaks at long wavelength. In blue PHOLEDs, the emission zone was reported to be located at near the position of electron transporting layer and the intensity of shoulder peak is varied depending on emission zone location [14]. The emission shoulder around 500 nm in the EL spectrum is enhanced when charge recombination zone shifts to HTL side. The emission zone of Device A may be close to the electron transport layer due to hole dominated property of the EML, leading to weak shoulder peak than Device B. On the other hands, the Device B shown rather strong shoulder peak than Device A due to little movement of emission zone toward HTL side through the better electron injection and transportation. Moreover, Device B shows that intensity of shoulder peak is increased with increasing applied voltage. Electron mobility in general electron type host materials has large variation depending on the applied electrical field [21]. The same tendency was observed in our devices. The color coordinate of the Devices A and B change from (0.16,0.33) to (0.16,0.33) and from (0.17,0.34) to (0.18,0.37) with variation of applied voltage from 5 to 8 V, respectively. The EL spectra of Devices C and D are almost same and their spectrum does not change significantly during variation of luminance from 5 to 8 V (CIE coordinates (0.16,0.34) at 1000 cd/m²).

Indeed, low-driving voltages and high efficiencies in solution processed blue PHOLEDs were demonstrated by using several small molecule host systems. For more accurate understanding on our low driving voltage characteristics, charge trapping situation at the EML was investigated by observation of J - V characteristics depending on doping concentrations in Devices A–C. Fig. 4 shows J - V characteristics of Devices A–C with 0%, 5% and 10% doping conditions. When TCTA is used as a host, current conduction is significantly decreased with doping of Flrpic. With increasing doping concentration from 5% to 10%, current density is improved a certain amount. This decrease current conduction with doping could be understood that deep electron trapping sites by dopants delay electron carrier movement. The LUMO gap between TCTA and Flrpic is 0.5 eV, which could easily generate deep trapped charges in the EML. This deep trap situation could be improved with increasing doping concentration because sufficient dopants can make carrier connection pathways through dopant molecules. Similar results were reported by several groups [18,22]. On the other hand, 26DCzPPy host (Device B) and mixed

host (Device C) has no big energetic difference for holes and electrons. The HOMO difference and LUMO difference between the host and dopant materials are within 0.3 eV. The J - V variation with varying doping concentration is negligible and there is a small decrease of current conduction by doping, which indicates that there is only shallow charge trapping situation in the EML in Devices B and C. Such shallow charge trapping could overcome easily by applying voltage in the device. Lower driving voltages in Devices B and C than Device A are understood by this shallow charge trapping situation in the EML. In case of mixed host system, fast hole carrier movement by TCTA additionally influences to the voltage reduction of our device because TCTA hole mobility ($3.0 \times 10^{-4} \text{ cm}^2/\text{Vs}$) is one order higher than that of 26DCzPPy ($2.0 \times 10^{-5} \text{ cm}^2/\text{Vs}$). Hence, the lowest driving voltage in Device C is observed. These results are well correlated with emission zone results obtained from emission spectra.

In order to understand morphological effects in our host dopant system, the atomic force microscopy (AFM) images of solution and vacuum deposition films were investigated

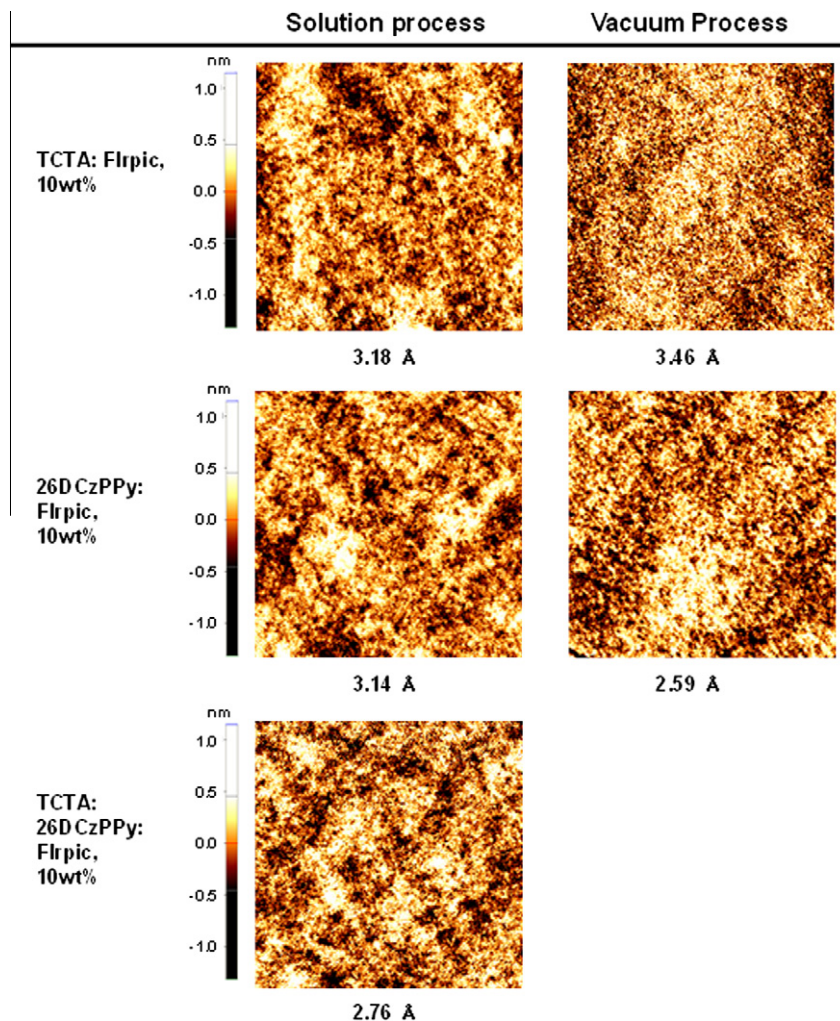


Fig. 5. AFM images of host-films by spin coating and vacuum deposition process.

Table 3

Photophysical properties of emitting layer films made by spin coating and vacuum deposition process.

Emitting layer films (doped with 10% Flrpic)	Abs λ_{\max} (nm)		PL λ_{\max} (nm)		Φ (%) ^a	
	Solution process	Vacuum process	Solution process	Vacuum process	Solution process	Vacuum process
TCTA: Flrpic	297 330	298 332	472	474	57.6	63.2
26DCzPPy	297 344	297 344	472	474	61.9	69.1
TCTA:26DCzPPy	297 328 343	297 329 342	472	474	79.6	74.0

^a Quantum yield values were obtained from the standard ($\Phi = 85\%$) sample of vacuum deposited mCP doped Flrpic 6% film [15].

as shown in Fig. 5. The TCTA, 26DCzPPy, and TCTA: 26DCzPPy (50:50) films doped with 10 wt.% of Flrpic on PEDOT: PSS were prepared by solution spin coating process and vacuum deposition process. The root-mean-square-roughness (rms) values of each host films are about 3.18 Å, 3.14 Å and 2.76 Å for solution processed TCTA, 26DCzPPy and TCTA: 26DCzPPy films and 3.46 Å, 2.59 Å for vacuum deposited TCTA and 26DCzPPy films, respectively. The TCTA: 26DCzPPy film by vacuum deposition process could not measure due to very brittle and smooth surface property. The solution processed single host films show somewhat high rms value compared to vacuum deposited films. However, such a small higher value of rms in solution processed films than vacuum deposited could not be the hindrance in making sufficient good amorphous films for the solution processed OLED. Photoluminescence quantum yields for solution and vacuum deposited films were investigated to acquire the additional information about film quality. Measured photophysical properties of EML films made by spin coating and vacuum deposition process are summarized in Table 3. The measured photoluminescence quantum yields for solution processed TCTA, 26DCzPPy, and TCTA: 26DCzPPy films are 57.6%, 61.9%, and 79.6%, respectively, compared with 85.0% standard sample (mCP film doped with 6% Flrpic) [15]. While vacuum deposited films show 63.2%, 69.1%, and 74.0% of photoluminescence quantum yields, respectively. Observation of a little bit higher quantum efficiency in the solution processed mixed host film indicate that the film quality is quite comparable to that of vacuum deposition process. The mixed small molecule host film by solution process in our system has not only acceptable rms value for the device fabrication but also smooth morphology. A low-driving voltage of 5.4 V and a high current efficiency of 25.1 cd/A in Device C are the consequences of such good morphological effect in mixed host system.

4. Conclusion

We have successfully demonstrated a mixed host system for a low-driving voltage and high efficiency solution processed blue PHOLEDs. The low driving voltage and high efficiencies are attributed to the mixed host which possesses high carrier mobility with bi-polar transport

characteristics and good morphological properties. High triplet energy and deep HOMO level of electron transporting layer can confine all triplet excitons inside the EML. The low driving voltage of 5.4 V at luminance of 1000 cd/m², maximum current efficiency of 25.1 cd/A and maximum quantum efficiency of 14.6% in a solution-processed blue PHOLED are paramount for large area low cost manufacturing.

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