ADVANCED FUNCTIONAL MATERIALS
www.afm-journal.de

Engineering of Mixed Host for High External Quantum Efficiency above 25% in Green Thermally Activated Delayed Fluorescence Device

*Bo Seong Kim and Jun Yeob Lee**

Highly efficient thermally activated delayed fluorescence (TADF) devices are developed by engineering mixed host materials in the emitting layer. Mixed hosts with deep highest occupied molecular orbital and high singlet energy without any exciplex formation are ideal as the host material for the TADF organic light-emitting diodes. A high external quantum efficiency of 28.6% is achieved in the green TADF organic light-emitting diodes using a mixed host of 1,3-bis(*N*-carbazolyl)benzene:1,3,5-tri[(3-pyridyl)-phen-3-yl]benzene and green emitting (4s,6s)-2,4,5,6-tetra(9H-carbazol-9-yl)isophthalonitrile TADF emitter.

1. Introduction

Thermally activated delayed fluorescence (TADF) devices have attracted great attention as next generation organic light-emitting diodes (OLEDs) because of high quantum efficiency of the TADF devices.[1] In the TADF devices, triplet excitons can be converted into singlet excitons via reverse intersystem crossing from triplet state to singlet state and the reverse intersystem crossing activates the triplet excitons for light emission. Therefore, 100% internal quantum efficiency can be theoretically obtained by the up-conversion process.

Comparing the TADF devices with phosphorescent OLEDs, both devices can achieve 100% internal quantum efficiency because both singlet and triplet excitons can be utilized for light emission. Phosphorescent OLEDs use triplet excitons for light-emission to harvest both singlet and triplet excitons,[2] while TADF OLEDs take advantage of singlet excitons for light-emission to activate both singlet and triplet excitons. Although maximum internal quantum efficiency of the TADF devices and phosphorescent OLEDs can be as high as 100%, the TADF devices can be better than phosphorescent OLEDs because of versatile design and easy synthesis of pure organic TADF emitters[3–11] compared to limited molecular structure and complicated synthesis of Ir based triplet emitters.[12–15] In addition, the singlet and triplet energies of TADF emitters are lower than those of triplet emitters if both emitters give emission at the same wavelength, which is advantageous to lower the driving voltage because narrow bandgap host materials can be applied.

Therefore, there have been several studies to develop high efficiency TADF devices and most researches were directed to design and synthesize new TADF emitting materials.[1,3–11] Adachi et al. reported a high quantum efficiency of 19.3% using (4s,6s)-2,4,5,6-tetra(9H-carbazol-9-yl) isophthalonitrile (4CzIPN) as a green TADF emitter.[1] The 4CzIPN emitter exhibited a high photoluminescence (PL) quantum efficiency of 94 ± 5% and TADF emission by reverse intersystem crossing from triplet excited state to singlet excited state. Other than this, several TADF emitters were reported although the quantum efficiency of the TADF devices is still not comparable to that of phosphorescent OLEDs.[3–11] However, there has been few papers studying host materials for TADF devices in spite of importance of the host materials. Common triplet host materials such as 4,4′-di(9H-carbazol-9-yl)biphenyl (CBP) and 2,8-bis(diphenylphosphoryl)dibenzo[b,d]thiophene were used as the host materials for the TADF devices,[1] but those host materials could not give optimized device performances because of strong hole or electron transport properties. The problem of poor charge balance of the host materials for the TADF devices can be solved by using a mixed host in the emitting layer.[16–18] The mixed host made up of a hole transport type host and an electron transport type host can balance holes and electrons in the emitting layer and improve the quantum efficiency of the device. In particular, exciplex type mixed hosts are effective to enhance the quantum efficiency of phosphorescent OLEDs.[17,18] However, the exciplex type mixed host structures developed for phosphorescent OLEDs may not be suitable for application in TADF OLEDs because of deep energy levels of TADF emitters. Therefore, more detailed study about the mixed host is strongly required to improve the device performances of the TADF OLEDs.

In this work, an optimized host structure for high efficiency TADF OLEDs was developed by combining hole transport type hosts and electron transport type host materials. It was demonstrated that a mixed host with the deep highest occupied molecular orbital (HOMO) and high singlet energy is ideal as the host material for the TADF OLEDs. A high external quantum efficiency of 28.6% was achieved in the green TADF OLEDs using a mixed host of 1,3-bis(*N*-carbazolyl)benzene (mCP):3,3″,5,5″-tetra(pyridine-3-yl)-1,1′:3′1″-terphenyl (BmPyPb) and the green emitting 4CzIPN TADF emitter. The external quantum efficiency value is better than any other data reported in the TADF devices.

B. S. Kim, Prof. J. Y. Lee
Department of Polymer Science and Engineering
Dankook University
126, Jukjeon-dong, Suji-gu, Yongin, Gyeonggi
448-701, Korea
E-mail: leej17@dankook.ac.kr

DOI: 10.1002/adfm.201303730

2. Results and Discussion

There are several requirements for the host materials of TADF devices, which include high singlet and triplet energies, large overlap of host emission with dopant absorption, the HOMO/lowest unoccupied molecular orbital (LUMO) levels to suppress exciton quenching and bipolar charge transport properties. The high singlet and triplet energies of the host materials are important to prevent exciton quenching by the host materials. In the case of the TADF emitters, triplet excitons contribute to the light emission via reverse intersystem crossing, so both the singlet and triplet energies of the host materials should be higher than those of TADF emitters. Large overlap of the host emission with dopant absorption is also needed for efficient energy transfer from the host to the TADF dopant and the HOMO/LUMO levels of the host should be suitable for suppressing exciton quenching. In particular, the HOMO level of the TADF emitters is generally deep because of strong electron withdrawing units in the molecular structure,[1] which should be carefully considered for the development of the host for the TADF emitters. In addition, bipolar charge transport properties are required to improve the recombination efficiency by balancing holes and electrons in the emitting layer.

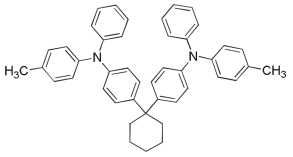
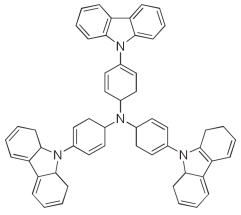
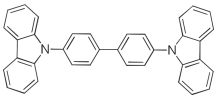
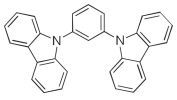
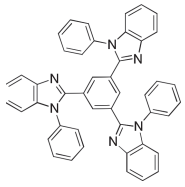
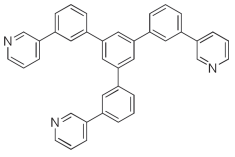
To develop an ideal mixed host for the TADF emitters, four hole transport type host materials and two electron transport type host materials were used. The hole transport type host materials were 4,4′-(cyclohexane-1,1-diyl)bis(N-phenyl-N-p-tolylaniline) (TAPC), 4,4′,4″-tris(*N*-carbazolyl)triphenylamine (TCTA), CBP and mCP which have different HOMO levels of −5.5, −5.7, −5.9, and −6.1 eV, respectively. The four host materials were selected to study the effect of HOMO levels of the hole transport type host on the exciplex formation and device performances of the TADF OLEDs. The electron transport type host materials were 1,3,5-tris(N-phenylbenzimidazole-2-yl)benzene (TPBI) and BmPyPb which have LUMO levels of −2.80 and −2.7 eV, respectively. All host materials used in this work showed higher singlet and triplet energies than the 4CzIPN dopant to suppress exciton quenching by the host itself. The HOMO, LUMO, singlet and triplet energies of the host materials are summarized in **Table 1**.

The effect of the hole transport type host materials on the photophysical properties of the mixed hosts was investigated using mixed hosts of TPBI mixed with four hole transport type host materials. Four mixed hosts of TAPC:TPBI, TCTA:TPBI, CBP:TPBI, and mCP:TPBI were prepared. The composition of the mixed hosts was 50:50 in all hosts. Exciplex formation in the mixed host was investigated by comparing photoluminescence (PL) emission of the mixed host. **Figure 1** shows PL spectra of the mixed host materials in comparison to the emission of the single host material. TAPC:TPBI and TCTA:TPBI exhibited long wavelength emission originated from exciplex formation between the hole transport type host materials and TPBI, while CBP:TPBI and mCP:TPBI did not show the long wavelength emission by exciplex. This indicates that TAPC:TPBI and TCTA:TPBI are exciplex type mixed hosts, but CBP:TPBI and mCP:TPBI are exciplex free mixed hosts. The shallow HOMO level of TAPC and TCTA originated from aromatic amine units induced the exciplex formation in the TAPC:TPBI and TCTA:TPBI, while the deep HOMO level of CBP and mCP prevented the exciplex formation in the CBP:TPBI and mCP:TPBI. The singlet energies of the TAPC:TPBI, TCTA:TPBI, CBP:TPBI, and mCP:TPBI were 2.71, 2.82, 3.00, and 3.15 eV, respectively, which were higher than 2.40 eV of 4CzIPN and the singlet emission of the mixed hosts was overlapped with the absorption of 4CzIPN. Especially, the overlap of the mCP:TPBI emission with the 4CzIPN absorption was extensive. Therefore, the TPBI derived mixed hosts may transfer emission energy to the green emitting 4CzIPN dopant.

PL spectra of the mixed hosts doped with the 4CzIPN dopant were analyzed to compare energy transfer from the mixed host to 4CzIPN. Excitation wavelength was 310 nm for host excitation as can be seen in ultraviolet-visible (UV-Vis) spectra of the mixed hosts (Figure S1 in supporting information). **Figure 2**a shows the PL spectra of 4CzIPN doped mixed hosts at an excitation wavelength of 310 nm. The PL spectra were not normalized to compare the PL intensity of the 4CzIPN emission. The PL intensity of 4CzIPN was strong in the mCP:TPBI host and the PL intensity was weakened in other hosts. In particular, the PL emission of 4CzIPN doped TCTA:TPBI and TAPC:TPBI hosts was very weak. The PL intensity of the 4CzIPN doped mixed host is related with poor energy transfer and exciton quenching by hole transport type host materials. Comparing the energy transfer of the mixed host materials, the overlap of the PL emission of the mixed hosts with UV-Vis absorption of 4CzIPN was extensive in the CBP:TPBI and mCP:TPBI hosts, but it was narrow in the TAPC:TPBI and TCTA:TPBI hosts because of exciplex formation which emits at long wavelength. Therefore, the energy transfer from the mixed hosts to 4CzIPN is efficient in the CBP:TPBI and mCP:TPBI hosts which do not form exciplex. In the case of the 4CzIPN doped TAPC:TPBI host, exciplex formation between TAPC and 4CzIPN dopant was also observed. Comparing the PL emission of TAPC:TPBI and TAPC:TPBI:4CzIPN, the 4CzIPN doped TAPC:TPBI showed exciplex emission of the TAPC:TPBI host and featureless long wavelength emission by exciplex formation between TAPC and 4CzIPN. Similar behavior was observed in the TCTA:TPBI:4CzIPN, which resulted in broad PL emission extended to long wavelength by exciplex formation between TCTA and 4CzIPN.

Energy transfer from the mixed hosts to 4CzIPN can be confirmed by the PL spectra. In the case of TAPC:TPBI, TCTA:TPBI, and CBP:TPBI hosts, host emission was clearly observed, implying incomplete energy transfer from the host to 4CzIPN. However, the host emission disappeared in the mCP:TPBI host because of efficient energy transfer. PL quantum yields of the 4CzIPN doped mixed hosts were 97, 66, 15 and 6% for mCP:TPBI:4CzIPN, CBP:TPBI:4CzIPN, TCTA:TPBI:4CzIPN, and TAPC:TPBI:4CzIPN films, respectively. The efficient energy transfer yielded high quantum efficiency in the mCP:TPBI:4CzIPN film. Förster energy transfer rate was calculated for the CBP:TPBI and mCP:TPBI mixed hosts to further study the energy transfer from the mixed hosts to 4CzIPN. Förster radii of the CBP:TPBI:4CzIPN and mCP:TPBI:4CzIPN emitting layer were 4.6 nm and 4.7 nm, respectively, according to the method reported in the literature.[19] Energy transfer rates from the Förster radii and excited state lifetime of the host materials were 1.5×10^{16} s^{-1} and 1.3×10^{16} s^{-1} for CBP:TPBI and mCP:TPBI mixed hosts.

Table 1. HOMO, LUMO, singlet energy, and triplet energy of the host materials.

Host	Chemical structure	HOMO [eV][a]	LUMO [eV][b]	Singlet energy [eV][c]	Triplet energy [eV][d]
TAPC		−5.5	−2.1	3.26	2.90
TCTA		−5.7	−2.4	3.12	2.70
CBP		−5.9	−2.6	3.00	2.60
mCP		−6.1	−2.4	3.47	2.90
TPBI		−6.1	−2.8	3.22	2.70
BmPyPb		−6.4	−2.7	3.45	2.78

[a]The HOMO was measured by cyclic voltammetry; [b]The LUMO was calculated from the HOMO and bandgap of UV-Vis absorption; [c]Singlet energy was calculated from the peak maximum of PL emission; [d]Triplet energy was calculated from the first phosphorescent peak of low temperature PL emission measured in liquid nitrogen.

Exciton quenching by hole transport type hosts can also affect the PL emission of the 4CzIPN doped mixed hosts. The HOMO level of 4CzIPN is −5.80 eV and the 4CzIPN excitons can be quenched by the host materials with the HOMO level shallower than that of 4CzIPN. The exciton quenching was studied by exciting only 4CzIPN without mixed host excitation. Figure 2b represents PL emission spectra of the 4CzIPN doped mixed hosts excited by 370 nm light source. The PL intensity was high in the 4CzIPN doped CBP:TPBI and mCP:TPBI mixed hosts, and weak PL emission was observed in the 4CzIPN doped TAPC:TPBI host. The order of PL intensity (mCP>CBP>TCTA>TAPC) was the same as the order of the HOMO level of the hole transport type host. This indicates that the hole transport type host quenches the 4CzIPN excitons due to mismatch of the HOMO levels. The PL quenching was significant in the TAPC:TPBI and TCTA:TPBI hosts due to shallow HOMO levels of TAPC and TCTA, while it was insignificant in the CBP:TPBI and mCP:TPBI hosts due to deep HOMO levels of CBP and mCP. Although both CBP:TPBI and mCP:TPBI hosts doped with 4CzIPN exhibited strong PL emission, mCP was better than CBP to suppress exciton quenching of 4CzIPN because of relatively deep HOMO level of mCP. Therefore, the low PL intensity of the CBP:TPBI:4CzIPN compared to that of mCP:TPBI:4CzIPN after excitation by 310 nm light source is due to both exciton quenching by CBP and poor energy transfer.

TADF emission of 4CzIPN in the mixed host was confirmed by measuring delayed PL emission of 4CzIPN. Delay time for the PL measurement was 0.1 ms. Delayed PL emission of 4CzIPN doped mixed hosts is shown in **Figure 3**. The CBP:TPBI and mCP:TPBI hosts doped with 4CzIPN showed delayed PL emission corresponding to TADF emission by reverse intersystem crossing from triplet excited state to singlet excited state. The delayed PL emission was strong in the mCP:TPBI:4CzIPN due to efficient energy transfer and suppressed exciton quenching, but it was weak

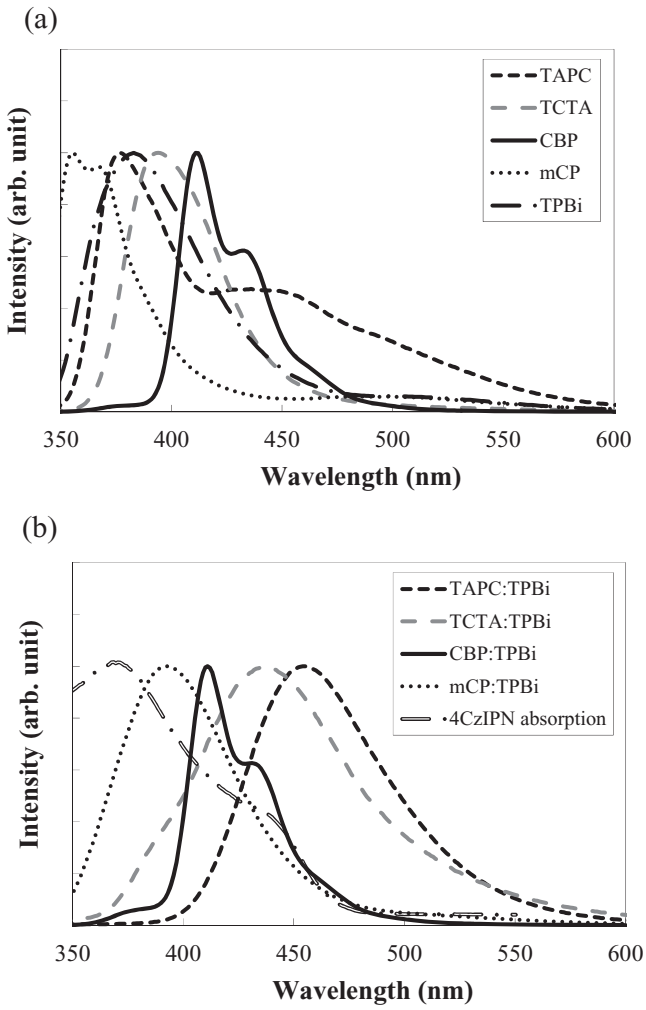


Figure 1. Solid PL spectra of a) single hosts and b) mixed hosts. UV-Vis absorption of 4CzIPN was added for comparison.

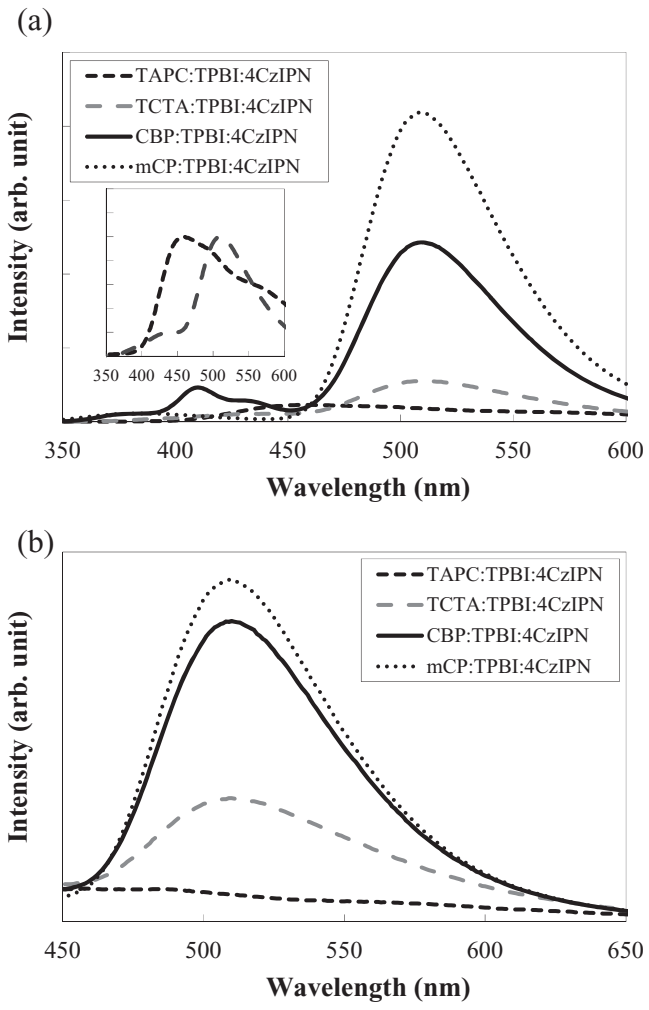


Figure 2. Solid PL spectra of 4CzIPN doped mixed hosts. Excitation wavelength was a) 310 nm and b) 370 nm.

in the CBP:TPBI:4CzIPN film. The TAPC:TPBI:4CzIPN and TCTA:TPBI:4CzIPN film did not show any delayed PL emission because of exciton quenching and exciplex formation between TAPC and 4CzIPN. This results suggests that the CBP:TPBI and mCP:TPBI mixed hosts effectively activate the TADF emission of 4CzIPN dopant, but the TAPC:TPBI and TCTA:TPBI hosts deactivate the TADF emission. In particular, the mCP:TPBI host was effective to harvest triplet excitons for singlet emission.

Transient PL measurement of the 4CzIPN doped mixed host was carried out to study the emission process of the mixed hosts. **Figure 4** shows transient PL curves of the 4CzIPN doped mixed hosts with different hole transport type host materials. TAPC:TPBI and TCTA:TPBI mixed hosts doped with 4CzIPN did not exhibit clear delayed PL decay curves, while CBP:TPBI and mCP:TPBI mixed hosts doped with 4CzIPN showed clear delayed PL emission with an excited state lifetime of 4.3 μs and 4.5 μs, respectively. The excited state lifetime for delayed PL emission was similar to that of 4CzIPN reported in Adachi's work.[1] This indicates that the CBP:TPBI and mCP:TPBI mixed hosts activate the TADF emission of 4CzIPN.

Based on the efficient TADF emission of 4CzIPN in the mixed host, TADF devices were fabricated. The device structure of the TADF devices was indium tin oxide (ITO,

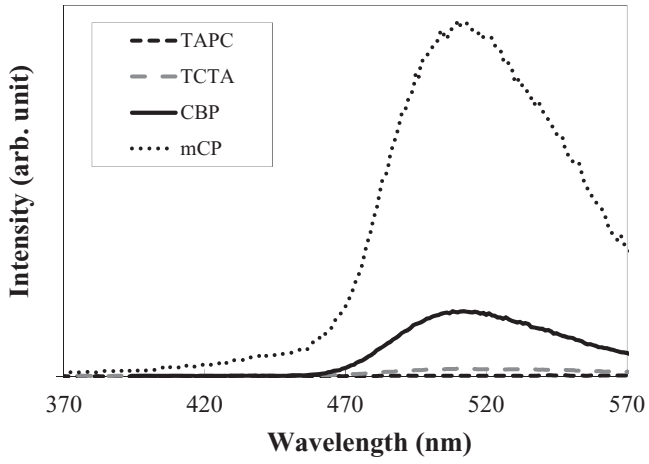


Figure 3. Delayed PL spectra of 4CzIPN doped mixed hosts (delay time 10 μs) excited by 310 nm light source.

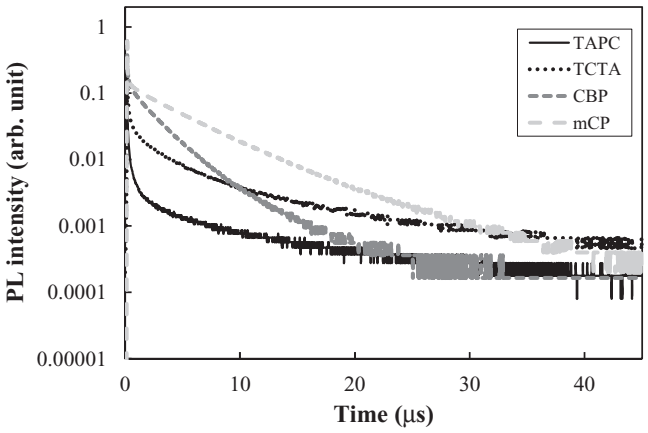


Figure 4. Transient PL decay curves of 4CzIPN doped mixed hosts.

50 nm)/poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, 60 nm)/TAPC (20 nm)/mCP (10 nm)/mixed host:4CzIPN (25 nm, 3%)/ diphenylphosphine oxide-4-(triphenylsilyl)phenyl (TSPO1, 35 nm)/LiF (1 nm)/Al (200 nm). **Figure 5** shows current density-voltage-luminance curves of the 4CzIPN doped mixed host devices. The current density of the mixed host device was high in the TAPC:TPBI and TCTA:TPBI devices, while it was low in the CBP:TPBI and mCP:TPBI devices. The high current density of the TAPC:TPBI and TCTA:TPBI devices is due to high hole mobility of TAPC and TCTA, and shallow HOMO level for efficient hole injection as reported in other works.[20,21] In the case of luminance, the luminance was high in the TAPC:TPBI and TCTA:TPBI devices at low current density, but it was reduced at high current density due to poor recombination efficiency and exciton quenching.

External quantum efficiency-luminance curves of the mixed host devices are shown in **Figure 6**. The quantum efficiency of the 4CzIPN mixed host devices was in the order of mCP:TPBI>CBP:TPBI>TCTA:TPBI>TAPC:TPBI, which agreed with the order of PL intensity of the 4CzIPN doped mixed host film because high quantum efficiency can be obtained in the device with an emitting layer having high PL quantum efficiency. The maximum quantum efficiency of the mCP:TPBI device was

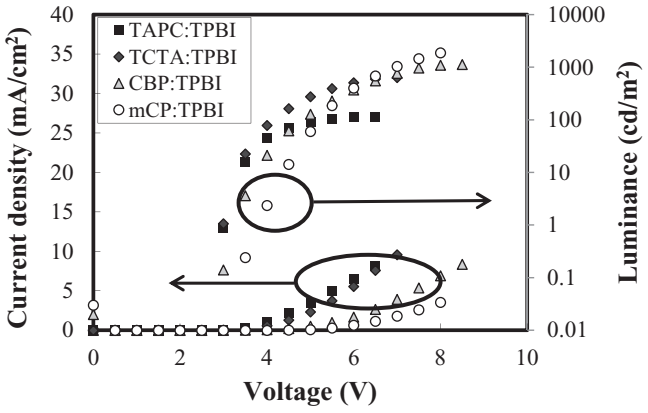


Figure 5. Current density–voltage–luminance curves of 4CzIPN doped mixed host devices.

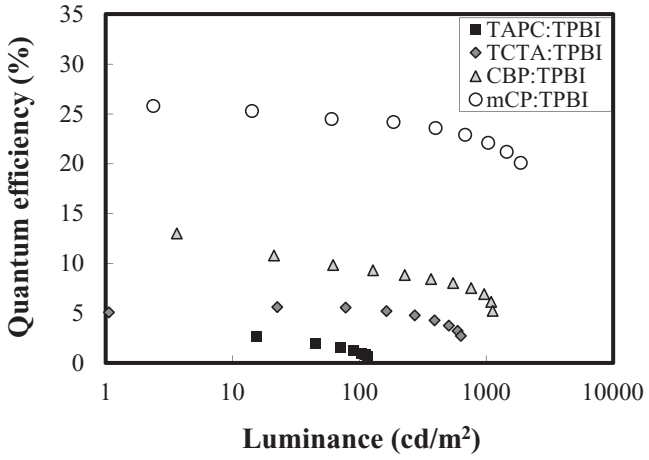


Figure 6. Quantum efficiency–luminance curves of 4CzIPN doped mixed host devices.

25.8% and the quantum efficiency at 1000 cd m^{-2} was 22.2%. The quantum efficiency obtained in this work is better than any other data reported in the TADF device. Compared with the state of the art external quantum efficiency of 19.3% in Adachi's work,[1] the quantum efficiency was improved by more than 33% in this work by engineering the mixed host in the emitting layer. There are several factors for the high quantum efficiency of the mCP:TPBI mixed host device. Firstly, the high PL quantum efficiency of the mCP:TPBI:4CzIPN emitting layer improved the quantum efficiency of the TADF device because of efficient energy transfer. Extensive overlap of PL emission of mCP:TPBI host with UV-Vis absorption of 4CzIPN facilitated the energy transfer between mCP:TPBI host and 4CzIPN. Secondly, the mCP:TPBI activated the TADF emission of 4CzIPN due to high singlet and triplet energy of the host material. The high singlet and triplet energies of the host suppressed exciton quenching, improving the quantum efficiency of the device. Thirdly, little exciton quenching of 4CzIPN by the mCP host material also contributed the high quantum efficiency owing to the deep HOMO level of mCP. The deep HOMO level of mCP prevented 4CzIPN exciton quenching and maximized light emission of 4CzIPN. Lastly, holes and electrons balance in the emitting layer by the mixed host structure increased recombination efficiency in the emitting layer, resulting in the high quantum efficiency. Therefore, it can be seen from this result that exciplex free mixed host made up of a hole transport type host with the deep HOMO level and an electron transport type host can be effectively used as the mixed host structure for TADF dopant. Only carbazole derived hosts were much better than aromatic amine type hosts because aromatic amine hosts shift the HOMO level, and induce exciton quenching and exciplex formation. This result is quite different from the experimental results about exciplex host in phosphorescent OLEDs.[17] In the case of the phosphorescent OLEDs, the exciplex host was effective to improve the quantum efficiency because the HOMO level of the hole transport type host in the exciplex host was deep enough for exciton blocking. However, the exciplex type host could not suppress exciton quenching in the 4CzIPN TADF devices, which lead to low quantum efficiency in the exciplex type host.

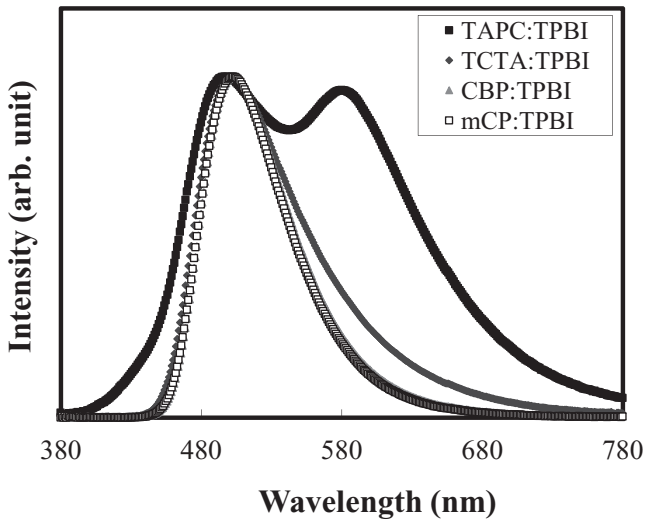


Figure 7. Electroluminescence curves of 4CzIPN doped mixed host devices.

Electroluminescence (EL) spectra of the mixed host TADF devices are shown in **Figure 7**. The CBP:TPBI:4CzIPN and mCP:TPBI:4CzIPN devices showed typical emission of 4CzIPN with a peak maximum at 502 nm without any other emission from host materials due to efficient energy transfer and charge trapping by 4CzIPN. However, the TCTA:TPBI:4CzIPN device exhibited extra tail at long wavelength in the EL spectra because of exciplex formation between TCTA and 4CzIPN. The exciplex emission between host and 4CzIPN and electromer emission of TAPC[22] were observed in the TAPC:TPBI device at a wavelength of 577 nm in addition to exciplex emission of TAPC:TPBI mixed host around 450 nm. It can be inferred from the EL spectra that the incomplete energy transfer, exciplex emission and electromer emission degraded the quantum efficiency of the TAPC:TPBI device, and exciplex emission decreased the quantum efficiency of the TCTA:TPBI device.

As it was confirmed that mCP is efficient as the hole transport type host material for the mixed hosts, other mCP based mixed hosts with different electron transport type host

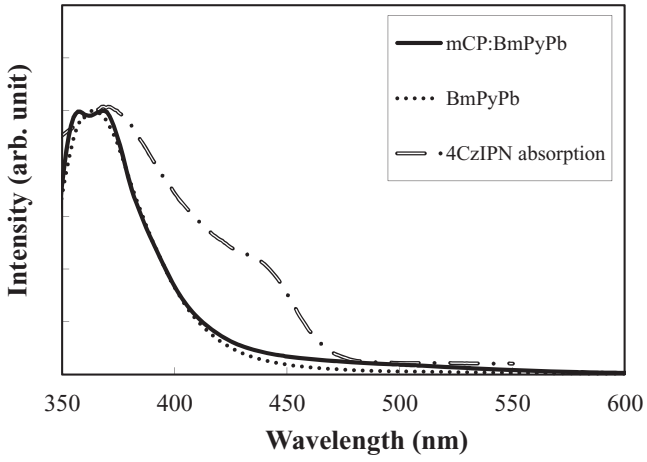


Figure 8. Solid PL spectra of single hosts and mixed host. UV-Vis absorption of 4CzIPN was added for comparison.

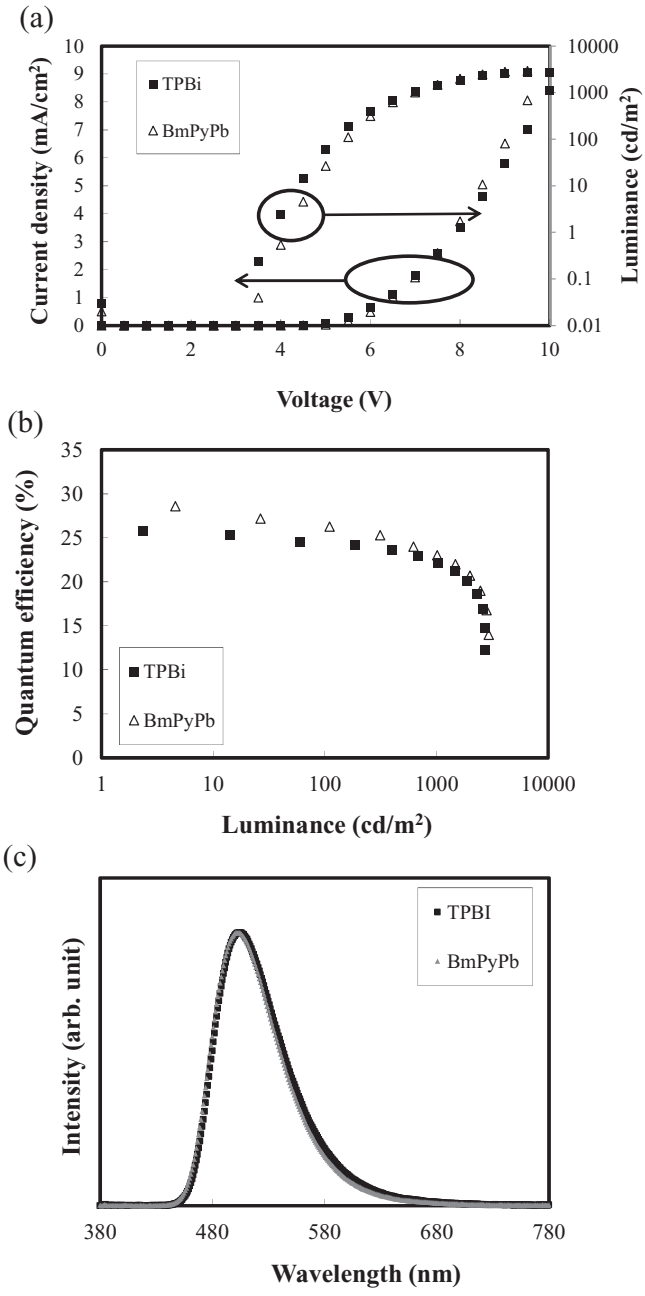


Figure 9. a) Current density–voltage–luminance, b) quantum efficiency-luminance, and c) electroluminescence curves of mCP:TPBI:4CzIPN and mCP:BmPyPb:4CzIPN mixed host devices.

materials were prepared. In addition to TPBI, an electron transport type host material, BmPyPb was mixed with mCP to produce the mixed hosts. The mCP:BmPyPb host did not form any exciplex as can be confirmed in the PL spectrum of the mixed host in **Figure 8**. There was no difference of the PL emission of the mCP:BmPyPb mixed host and that of BmPyPb or mCP. Therefore, the mCP:BmPyPb can be effectively used as the mixed host for the 4CzIPN dopant.

Current density-voltage-luminance curves of the mCP based mixed hosts are shown in **Figure 9**a. The current density of the

Table 2. Device performances of the mixed host TADF OLEDs.

Mixed host	Color index[a)]	Quantum efficiency [%]			Power efficiency [lm W^{-1}]		
		Max	100 [cd m^{-2}]	1000 [cd m^{-2}]	Max	100 [cd m^{-2}]	1000 [cd m^{-2}]
TAPC:TPBI	0.36, 0.44	3.3	1.1	–	8.2	1.4	–
TCTA:TPBI	0.27, 0.50	5.6	5.4	–	14.3	11.1	–
CBP:TPBI	0.23, 0.54	13.0	9.5	6.7	34.1	18.1	7.4
mCP:TPBI	0.22, 0.55	25.8	24.4	22.2	58.6	43.0	29.2
mCP:BmPyPb	0.21, 0.53	28.6	26.6	23.1	56.6	43.4	29.6

[a)]Color index was obtained at 100 cd m^{-2}.

mixed host was low in the mCP:BmPyPb host and the luminance also followed the same tendency as the current density.

Quantum efficiency-luminance curves of the mCP based mixed host devices are presented in Figure 9b. The external quantum efficiency was optimized in the mCP:BmPyPb device and maximum quantum efficiency of the mCP:BmPyPb device was 28.6%. The high quantum efficiency of the mCP:BmPyPb device can be explained by extensive overlap of the host PL emission with 4CzIPN absorption as shown in Figure 8. The extensive spectral overlap facilitated energy transfer from the mCP:BmPyPb host to 4CzIPN, which increased the quantum efficiency of the mCP:BmPyPb device. The quantum efficiency value achieved in this work is the best external quantum efficiency data reported in the TADF OLEDs. All device data are summarized in **Table 2**.

EL spectra of the mixed hosts are shown in Figure 9c. There was little difference of the EL spectra between mCP:TPBI and mCP:BmPyPb devices. Only pure 4CzIPN emission without any emission from charge transport materials or host materials was observed. This indicates that singlet and triplet excitons were confined in the emitting layer without any hole or electron leakage to the charge transport layers, which contributed to the high quantum efficiency of the mCP:BmPyPb device.

3. Conclusions

In conclusion, an optimized mixed host structure for high external quantum efficiency in green TADF OLEDs was developed by correlating the photophysical properties of the mixed hosts with device performances. A mixed host without any exciplex formation was suitable as the mixed host for the 4CzIPN TADF emitter because of reduced exciton quenching and efficient energy transfer. A mixed host of mCP and BmPyPb was the optimized mixed host structure and high external quantum efficiency of 28.6% was achieved in the green TADF OLEDs, which was better than any other data reported in green TADF OLEDs. From this result, it can be confirmed that the TADF OLEDs can be comparable to phosphorescent OLEDs in terms of external quantum efficiency.

4. Experimental Section

General Information: TAPC, TCTA, CBP, mCP, TSPO1 and TPBI were products of P&H Co. and BmPyPb were purchased from Daejoo Co. PEDOT:PSS was obtained from Sigma-Aldrich and was diluted with 2-propanol.

Device Fabrication and Measurements: Device architecture of the TADF OLEDs was ITO (50 nm)/PEDOT:PSS (60 nm)/TAPC (20 nm)/mCP (10 nm)/mixed host:4CzIPN (25 nm)/TSPO1 (35 nm)/LiF (1 nm)/Al (200 nm). The mixed hosts were TAPC:TPBI, TCTA:TPBI, CBP:TPBI, mCP:TPBI and mCP:BmPyPb. Mixed host composition was 50:50 in all mixed hosts and doping concentration of 4CzIPN was 3%. PL measurement of the mixed hosts and 4CzIPN doped mixed host films was carried out using fluorescence spectrophotometer (HITACHI, F-7000). Excitation wavelengths were 310 nm for host excitation and 370 nm for dopant excitation. Solid films were prepared by vacuum thermal evaporation at a thickness of 30 nm. Transient PL measurement was performed using a pulsed Nd—YAG laser (355 nm) as the excitation light source and an intensified charge-coupled device (ICCD) as a detector. Device performance measurement of the TADF OLEDs was carried out using Keithley 2400 source measurement unit and CS 1000 (Minolta Co.) spectroradiometer.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Received: November 3, 2013
Revised: February 3, 2013
Published online: March 24, 2014

[1] H. Uoyama, K. Goushi, K. Shizu, H. Nomura, C. Adachi, *Nature* **2012**, *492*, 234.
[2] M. A. Baldo, D. F. O'Brien, Y. You, A. Shoustikov, S. Sibley, M. E. Thompson, S. R. Forrest, *Nature* **1998**, *395*, 151.
[3] F. B. Dias, K. N. Bourdakos, V. Jankus, K. C. Moss, K. T. Karntekar, V. Bhalla, J. Santos, M. R. Bryce, A. P. Monkman, *Adv. Mater.* **2013**, *25*, 3707.
[4] K. Goushi, K. Yoshida, K. Sato, C. Adachi, *Nat. Photonics* **2012**, *6*, 253.
[5] Q. Zhang, J. Li, K. Shizu, S. Huang, S. Hirata, H. Miyazaki, C. Adachi, *J. Am. Chem. Soc.* **2012**, *134*, 14706.
[6] J. Li, T. Nakagawa, Q. Zhang, H. Nomura, H. Miyazaki, C. Adachi, *Adv. Mater.* **2013**, 25, 3319.
[7] S. Y. Lee, T. Yasuda, H. Nomura, C. Adachi, *Appl. Phys. Lett.* **2012**, *101*, 093306.
[8] T. Nakagawa, S.-Y. Ku, K.-T. Wong, C. Adachi, *Chem. Commun.* **2012**, *48*, 9580.
[9] G. Méhes, H. Nomura, Q. Zhang, T. Nakagawa, C. Adachi, *Angew. Chem. Int. Ed.* **2012**, *51*, 11311.
[10] H. Tanaka, K. Shizu, H. Miyazaki, C. Adachi, *Chem. Commun.* **2012**, *48*, 11392.
[11] H. Nakanotani, K. Masui, J. Nishide, T. Shibata, C. Adachi, *Sci. Rep.* **2013**, *3*, 2127.

[12] C. Ulbricht, B. Beyer, C. Friebe, A. Winter, U. S. Schubert, *Adv. Mater.* **2009**, *21*, 4418.
[13] K. Lu, H. Chou, C. Hsieh, Y. O. Yang, H. Tsai, H. Tsai, L. Hsu, C. Chen, I. Chen, C. Cheng, *Adv. Mater.* **2011**, *23*, 4933.
[14] K. S. Yook, J. Y. Lee, *Adv. Mater.* **2012**, *24*, 3169.
[15] L. Xiao, Z. Chen, B. Qu, J. Luo, S. Kong, Q. Gong, J. Kido, *Adv. Mater.* **2011**, *23*, 926.
[16] S. H. Kim, J. Jang, J. Y. Lee, *Appl. Phys. Lett.* **2007**, *91*, 083511.
[17] Y. Park, S. Lee, K. Kim, S. Kim, J. Lee, J. Kim, *Adv. Funct. Mater.* **2013**, *23*, 4914.
[18] S. Lee, K. Kim, D. Limbach, Y. Park, J. Kim, *Adv. Funct. Mater.* **2013**, *23*, 4105.
[19] T. Förster, **1949**, *4a*, 321.
[20] J. V. Grazulevicius, P. Strohriegl, *Adv. Mater.* **2002**, *14*, 1439.
[21] J. Kang, S. Lee, H. Park, W. Jeong, K. Yoo, Y. Park, J. Kim, *Appl. Phys. Lett.* **2007**, *90*, 223508.
[22] S. Yang, M. Jiang, *Chem. Phys. Lett.* **2009**, *484*, 54.