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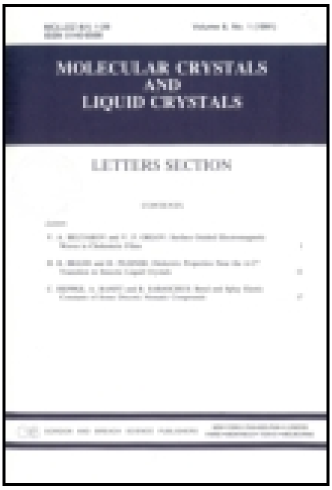

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Enhanced Hybrid Blue Organic Light Emitting Diodes with a Multi-Emitting Layer Using Fluorescent and Phosphorescent Emitters

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Enhanced Hybrid Blue Organic Light Emitting Diodes with a Multi-Emitting Layer Using Fluorescent and Phosphorescent Emitters

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We investigated efficient blue organic light emitting diodes (OLEDs) containing fluorescent and phosphorescent emitters in the emitting layer (EML). We fabricated several hybrid blue OLEDs with different thicknesses or switching positions for the fluorescent and the phosphorescent EMLs using the fluorescent deep blue emitter 1,6-Bis(N-phenyl-p-CN-phenylamino)-pyrenes and the phosphorescent blue emitter iridium(III)bis[(4,6-difluorophenyl)-pyridinato-N,C^2]picolinate. The fabricated device E stacked a 20 nm thickness of phosphorescent EML and a 10 nm thickness of fluorescent EML. Hybrid blue OLEDs have many advantages in terms of their electrical characteristics, as they have a lower efficiency roll-off than phosphorescent OLEDs and a higher efficiency than fluorescent OLEDs.

Keywords Organic light emitting diode; triplet harvesting; hybrid; fluorescence; phosphorescence; efficiency roll-off

Introduction

Organic light-emitting diodes (OLEDs) are drawing tremendous attention and are important because of their wide commercial applications in next-generation displays and solid-state lighting because they are thin, light weight, and eco-friendly [1, 2]. In terms of materials, phosphorescent organic light emitting diodes (PHOLEDs) have achieved high efficiency because both the singlet and triplet excitons can be harvested for light emission [3]. Therefore, many studies on the efficiency of PHOLEDs, with an adequate interlayer, a double emitting layer (EML), and multiple quantum well structures, have been suggested [4–6]. However, until now, a high efficiency blue phosphorescent emitter with an acceptable operational lifetime had not been developed [7]. To solve the problems of blue PHOLEDs, a

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combination of fluorescent and phosphorescent EMLs was introduced to a hybrid OLED, and triplet excitons can be harvested from the blue fluorescence to phosphorescence [8]. The triplet harvesting is primarily related to the fact that triplet excitons are usually lost in a fluorescent emitter system [9]. The concept here is to transfer these triplets to a phosphorescent emitter, where they can decay radiatively. For an efficient transfer, the triplet energy level of the phosphorescent emitter needs to be lower than that of the fluorescent emitter.

In this work, we reported the fabrication and characterization of hybrid blue OLEDs with blue fluorescent and phosphorescent emitters doped in different host materials. Hybrid blue OLED structures are controlled thickness or by switching the position of the fluorescent and phosphorescent EMLs. The triplet harvesting for the hybrid blue OLED, which combines a blue fluorescent emitter with a blue phosphorescent emitter, can be achieved with a higher efficiency than with fluorescent OLEDs and with a reduced roll-off when compared with PHOLEDs [9, 10].

Experiment

To fabricate the hybrid blue OLEDs, we used indium-tin-oxide (ITO)-coated glass substrates, which have a sheet resistance of 10 Ω/sq and a thickness of 180 nm. The cleaning procedure included sonication in acetone, methyl alcohol, distilled water, and ethyl alcohol at room temperature for 15 min, before depositing the organic layers. The pre-cleaned ITO-coated glass substrates were kept for 24 h in ethyl alcohol and dried in a convection oven at 110°C for 10 min. The ITO substrates were then treated with O_2 plasma at 2×10^{-2} Torr at 125 W for 2 min. Organic layers were deposited by thermal evaporation through a shadow mask in a high vacuum (5×10^{-7} Torr). The deposition rates were 1.0 to 1.1 Å/s for all organic materials and 0.1 Å/s for lithium quinolate (Liq). After the deposition of the organic layers without a vacuum break, an aluminum (Al) cathode was deposited at a rate of 10 Å/s. The deposition rates and the doping concentration of the dopants were controlled with a quartz crystal monitor. The fabricated devices were sealed simultaneously in a N_2-purged glove box with H_2O at a concentration below 1 ppm. Barium-oxide powder was used to absorb the residue moisture in the encapsulated device. The current density–voltage (J-V) characteristics of the OLEDs were measured with a source measure unit (Keithley 238). The electroluminescence (EL) performance of the devices including the luminance, luminous efficiency (LE), external quantum efficiency (EQE), Commission International de L'Éclairage coordinates, and EL spectra, was analyzed using CHROMA METER CS-1000A instruments. The half-decay lifetime was measured using a lifetime tester (Polaronix M6000).

Results and Discussion

Figure 1(a) shows the chemical structures of the key materials, with 1,6-Bis(N-phenyl-p-CN-phenylamino)-pyrenes (Pyrene-CN) and iridium(III) bis[(4,6-difluorophenyl)-pyridinato-N,C^2]picolinate (FIrpic) as the fluorescent and phosphorescent blue dopants, and 2-methyl-9,10-di(2-naphthyl)anthracene (MADN) and N,N′-dicarbazolyl-3,5-benzene (mCP) as the fluorescent and phosphorescent blue hosts. Pyrene-CN is a deep blue fluorescent emitter with CIE coordinates of $x = 0.14$ and $y = 0.10$, and MADN has multifunctional characteristics, including both hole transporting and electron transporting [11,12,13]. In addition, FIrpic is a well-known electron transporting blue emitter and mCP is a p-type host material based on carbazole [14,15,16]. Figure 1(b) shows energy level diagrams of for

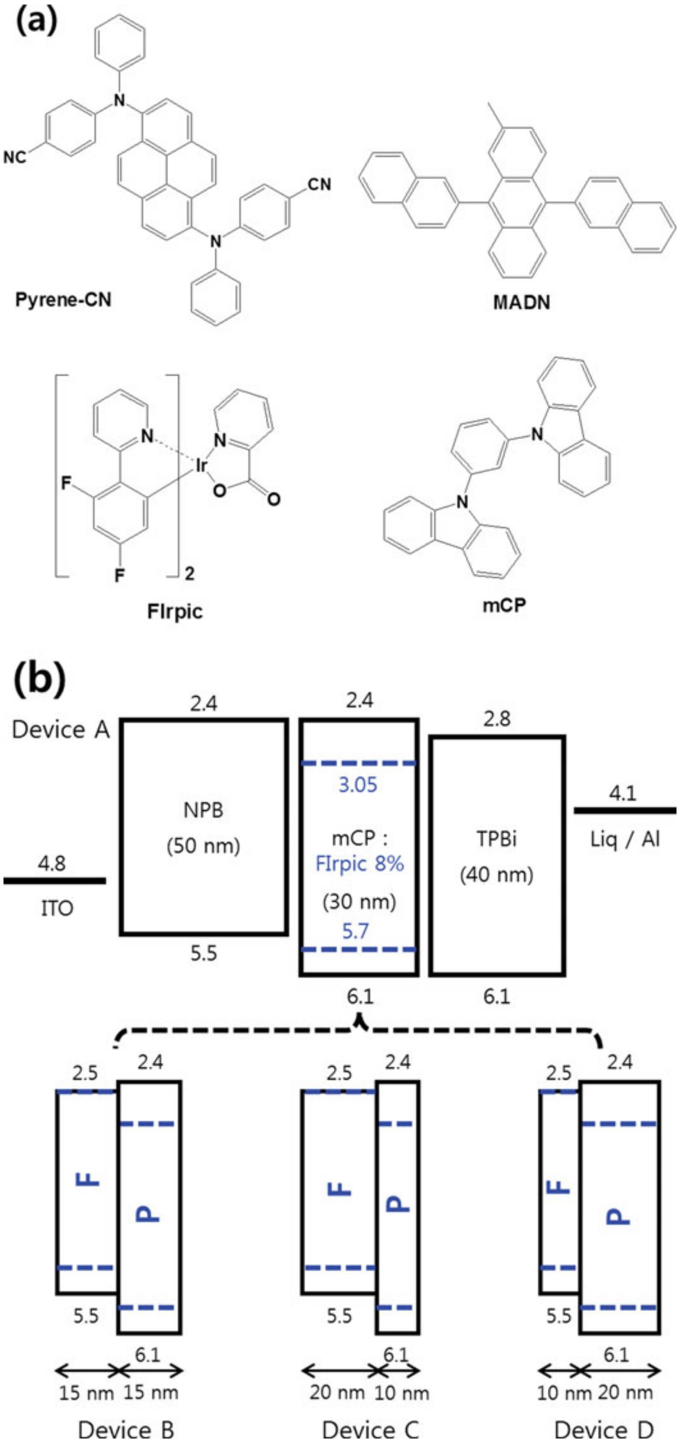

Figure 1. (a) Chemical structure of the blue fluorescent and phosphorescent emitting materials. (b) Energy level diagrams of devices A, B, C, and D (F: Pyrene-CN doped in MADN of 10 wt.%, P: FIrpic doped in mCP of 8 wt.%).

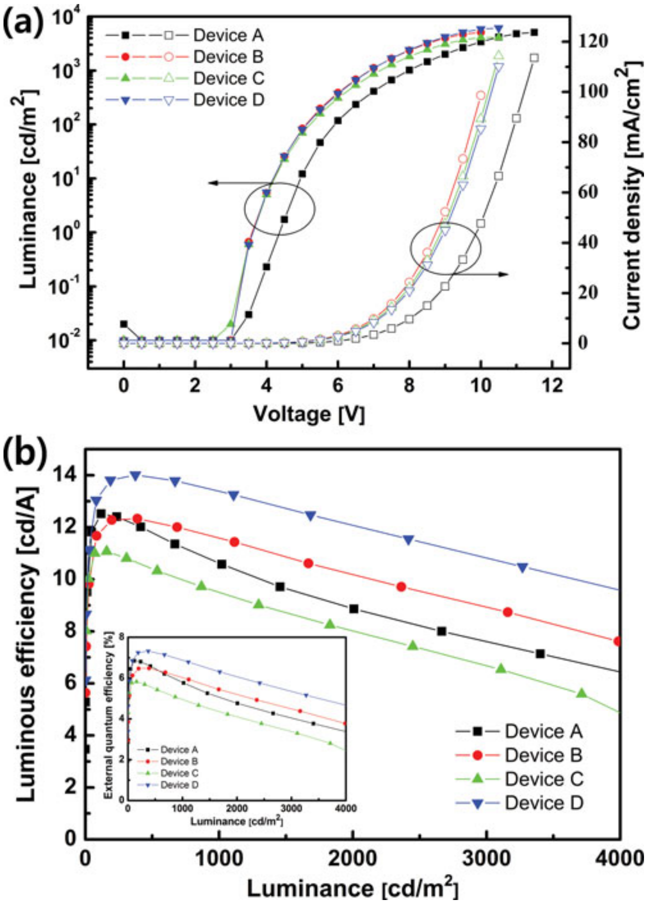

Figure 2. (a) Current density–voltage–luminance characteristics. (b) Luminous efficiency-luminance characteristics and (inset) external quantum efficiency-luminance characteristics of hybrid blue devices A, B, C, and D.

hybrid blue devices A, B, C, and D, which have the following structure: ITO as an anode / N,N′-bis-(1-naphyl)-N,N′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB, 50 nm) as a hole transporting layer (HTL) / EMLs (30 nm, Devices A, B, C, and D) / 2,2′,2″-(1,3,5-benzenetryl)tris(1-phenyl)-1H-benzimidazol (TPBi, 40 nm) as an electron transporting layer (ETL) / Liq (2 nm) as an electron injection layer (EIL) / Al (100 nm) as a cathode. Device A is a blue phosphorescent device used as a reference device, and hybrid blue devices B, C, and D have fluorescent (10 wt.%) and phosphorescent emitters (8 wt.%) doped individually into different hosts with different EML thicknesses. Devices B, C, and D were fabricated with 15 nm, 20 nm, and 10 nm thicknesses of fluorescent EML and 15 nm, 10 nm, 20 nm of phosphorescent EML, respectively.

Figure 2(a) shows the current density–voltage–luminance (J-V-L) characteristics of the fabricated hybrid blue OLEDs with different EMLs. The current density and luminance for devices A, B, C, and D were 47.71, 98.65, 89.35, and 85.29 mA/cm^2, and 3403, 5086, 4108, and 5850 cd/m^2 at 10 V, respectively. The current density and luminance values of device A were lower than those of devices B, C, and D within the whole voltage range. We

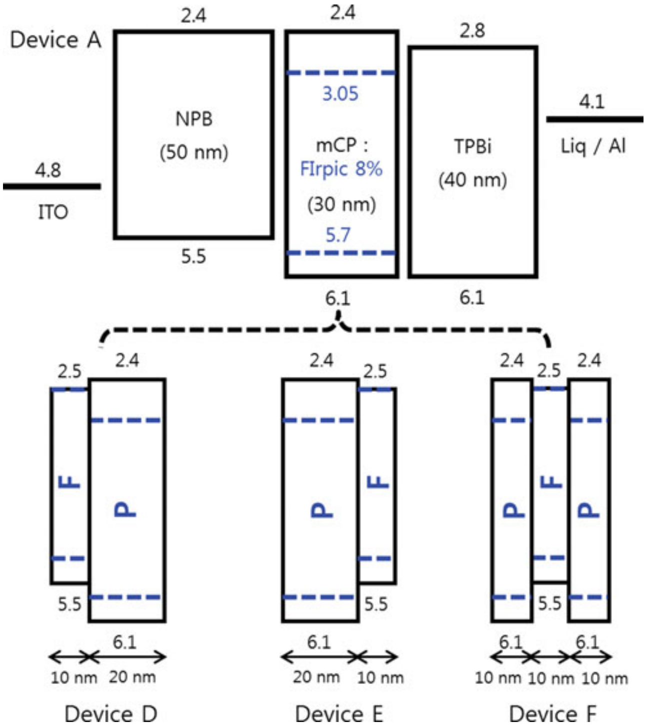

Figure 3. Energy level diagrams of the hybrid blue OLEDs (F: Pyrene-CN doped in MADN of 10 wt.%, P: FIrpic doped in mCP of 8 wt.%).

have also determined the LE-luminance characteristics of hybrid blue OLEDs, as shown in Figure 2(b), and the EQE-luminance characteristics of the hybrid blue devices as shown in the inset of Figure 2(b). The maximum LE and EQE of devices A, B, C, and D were 12.52, 12.33, 11.07, and 14.01 cd/A and 6.85, 6.48, 5.83, and 7.32%, respectively. The LE and EQE of device D were higher than those of the other blue devices. Typically, the use of a fluorescent emitter significantly reduces the efficiency of the device when compared to similar phosphorescent emitter based devices because of the waste of most of the triplet excitons from the fluorescent recombination case. Therefore, we developed efficient blue OLEDs based on mixed emission using fluorescence and phosphorescence that operated the triplet harvesting mechanism [8,9]. In order to do this mechanism, we used the fluorescent emitter with a higher triplet energy level than the FIrpic (phosphorescent emitter). The triplets of the Pyrene-CN can be transferred to the triplet level of a FIrpic. At the same time, the singlet transfer from Pyrene-CN to FIrpic needs to be hindered. This can be achieved through a spatial separation of the fluorescent emitter Pyrene-CN and the phosphorescent emitter FIrpic. Therefore, the three hybrid blue devices had higher current densities, luminance, and efficiency values than device A.

In addition, we compared devices A, D, E, and F, which had different thicknesses and positions for the fluorescent and phosphorescent EMLs. Figure 3 shows energy level diagrams for the hybrid blue devices, which have various different structures and different positions for the fluorescent and phosphorescent EMLs. Devices E and F were fabricated with a 10 nm fluorescent EML close to the ETL, and the center of the EML maintaining a thickness of 30 nm for the whole EML. Figure 4(a) shows the J-V-L characteristics of the

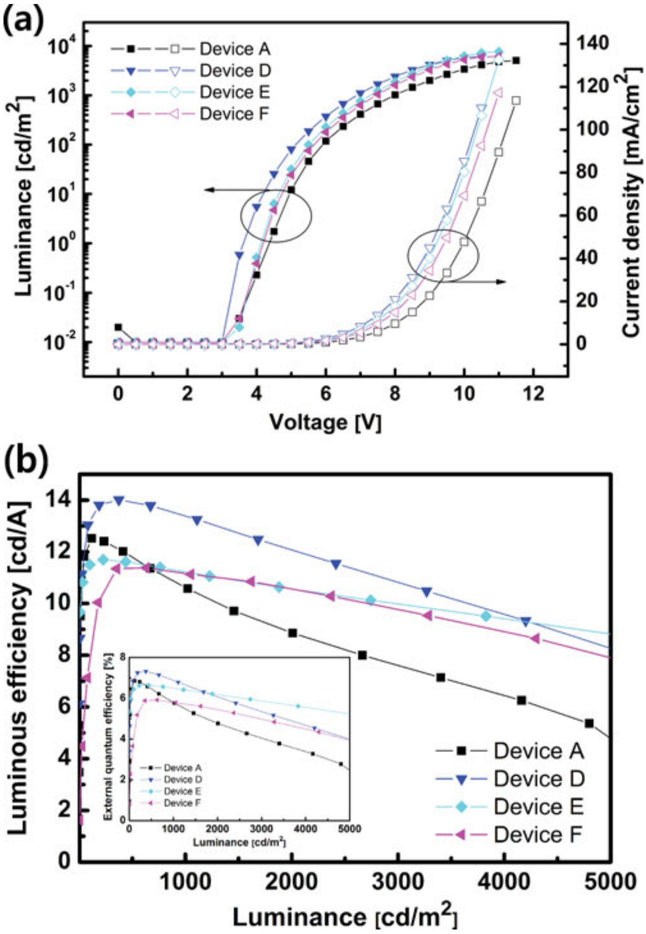

Figure 4. (a) Current density–voltage–luminance characteristics. (b) Luminous efficiency-luminance characteristics and (inset) external quantum efficiency-luminance characteristics of hybrid blue devices A, D, E, and F.

hybrid blue OLEDs (devices A, D, E, and F) for carrier and recombination balance. The current density and luminance of devices E and F were 80.42 and 69.31 mA/cm^2 and 6258 and 5274 cd/m^2 at 10 V, respectively. The EQE-luminance properties of the hybrid blue devices E and F are shown in Figure 4(b), and the LE-luminance properties of the devices are shown in the inset of Figure 4(b). The blue devices A, D, E, and F exhibited the LE and EQE values of 12.52, 14.01, 11.69, and 11.38 cd/A, and 6.85, 7.32, 6.62, and 5.18% at 6 V, respectively. Therefore, device D was high efficiency at a range of low voltage, but device E could be maintained at a high voltage; the EQE values were 4.76, 4.53, 5.61, and 4.83% at 9 V, respectively. The difference in roll-off for the EQE values between 6 V and 9 V were 30.5, 38.1, 15.2, and 6.8% for devices A, D, E, and F, respectively. These results can be explained by the different recombination zones. The half-decay lifetime properties for all the hybrid blue devices are shown in Figure 5(a) to support trend of devices. The half-decay lifetime value of device E was higher than for the other devices, while devices A, B, C, and D had almost the same values. Moreover, Figure 5(b) shows the EL spectra of the hybrid

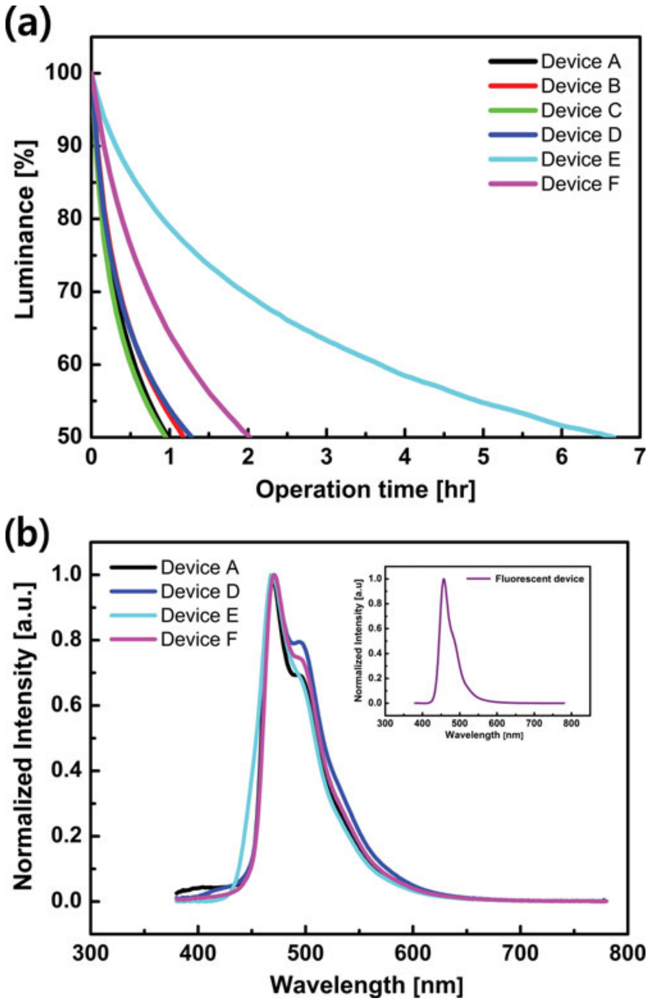

Figure 5. (a) Half-decay lifetime properties for all hybrid blue OLEDs. (b) The electroluminescent (EL) spectra of the hybrid blue OLEDs.

blue OLEDs (devices A, D, E, and F) as a function of the applied voltage at 8 V and the EL spectra of the reference device using the fluorescent dopant as Pyrene-CN in the inset of Figure 5(b). Therefore, the recombination zone was influenced by the emitter properties in the EML close to the ETL. As a result, device E had reduced efficiency roll-off, improved efficiency at a high voltage, and a high half-decay lifetime.

Conclusions

In summary, we have presented hybrid blue OLEDs using Pyrene-CN as fluorescent emitter and FIrpic as phosphorescent emitter based on different hosts in the EML. This structure shows an improved LE, EQE, and roll-off efficiency with a 10 nm thickness of fluorescent EML close to the ETL. The corresponding device E can maintain high electrical and optical properties at whole range of voltages, such as LE, EQE, and reduced efficiency roll-off as well. The hybrid blue OLED device E is caused by the triplet harvesting mechanism between the fluorescence and phosphorescence. These mechanism approaches are expected to yield OLEDs with enhanced efficiency and increased lifetimes because of their complete triplet systems.

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References

[1] D'Andrade, B., Thompson, M., & Forrest, S. (2002). *Adv. Mater.*, *14*, 147.
[2] D'Andrade, B., & Forrest, S. (2004). *Adv. Mater.*, *16*, 1585.
[3] Baldo, M. A., O'Brien, D. F., & You, Y. (1998). *Nature*, *395*, 151.
[4] Lee, J., Lee, J.-I., Song, K.-I., Lee, S. J., & Chu, H. Y. (2008). *Appl. Phys. Lett.*, *92*, 203305.
[5] Eom, S.-H., Zheng, Y., Chopro, N., Lee, J., So, F., & Xue, J. (2008). *Appl. Phys. Lett.*, *93*, 133309.
[6] Lee, S. J., Koo, J. R., Hyung, G. W., Lim, D. H., Lee, D. H., Lee, K. H., Yoon, S. S., Kim, W. Y., & Kim, Y. K. (2012). *Nanoscale Res. Lett.*, *7*, 23.
[7] Seo, J. H., Lee, S. J., Seo, B. M., Moon, S. J., Lee, K. H., J. K. P., Yoon, S. S., & Kim, Y. K. (2010). *Org. Electron.*, *11*, 1759.
[8] Zheng, T., & Choy, W. C. H. (2010). *Adv. Funct. Mater.*, *20*, 648.
[9] Schwartz, G., Reineke, S., Rosenow, T. C., Walzer, K., & Leo, K. (2009). *Adv. Funct. Mater.*, *19*, 1319.
[10] Zheng, C.-J., Wang, J., Ye, J., Lo, M.-F., Liu, X.-K., Fung, M.-K., Zhang, X.-H., & Lee, C.-S. (2013). *Adv. Mater.*, *25*, 2205.
[11] Wee, K. R., Ahn, H. C., Son, H. J., Han, W. S., Kim, J. E., Cho, D. W., & Kang, S. O. (2009). *J. Org. Chem.*, *74*, 8472.
[12] Seo, J. H., Seo, B. M., Koo, J. R., Lee, K. H., You, J. N., Yoon, S. S., & Kim, Y. K. (2011). *Curr. Appl. Phys.*, *11*, S356.
[13] Ho, M. H., Wu, Y. S., Wen, S. W., Lee, M. T., & Chen, T. M. (2006). *Appl. Phys. Lett.*, *89*, 252903.
[14] Adachi, C., Baldo, M. A., Thomposon, M. E., & Forrest, S. R. (2001). *J. Appl. Phys.*, *90*, 5048.
[15] Kim, S. H., Jang, J., & Lee, J. Y. (2007). *Appl. Phys. Lett.*, *90*, 223505.
[16] Su, S. J., Gonmori, E., Sasabe, H., & Kido, J. (2008). *Adv. Mater.*, *20*, 4189.