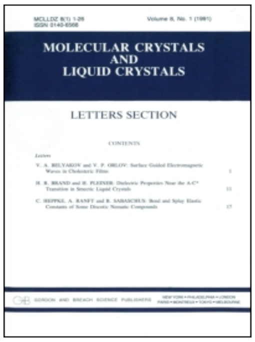

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Fabrication of hybrid white OLEDs with an inorganic color conversion layer

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ABSTRACT
We report hybrid white organic light-emitting diodes (WOLEDs) made by combining FIrpic-based blue-emitting OLEDs and a printable red-emitting inorganic phosphor. The color conversion can be easily controlled by adjusting the thickness of the red phosphor. FIrpic-based phosphorescent OLEDs, with a 15-μm-thick phosphor layer, show warm white light emission leading to a current efficiency of 11.4 cd/A, a power efficiency of 4.9 lm/W, and a CIE coordinate of (0.33, 0.41) at a luminance of 1000 cd/m^2. Moreover, the output spectra and CIE coordinates of the WOLED show no significant change over a wide range of current densities.

KEYWORDS
color conversion; white organic light-emitting diodes; inorganic phosphor

Introduction

The study of white organic light-emitting devices (WOLEDs) is of great importance because of their various advantages, including their low power consumption, large viewing angle, high contrast ratio, and fast response time. WOLEDs can be used in full color displays, backlighting, and for solid-state lighting [1–4]. Typically, white light is obtained by mixing either the three primary colors (red, green, and blue) or two complementary colors. To date, many methods have been developed for the realization of WOLEDs. These approaches have employed a single emissive layer that has been simultaneously doped with different color dopants [5–10], combinations of multiple-color light emissive layers [11–13], or the stacking of several OLED units that emit different colors [14–15].

Although efficient WOLEDs have been realized by adopting these methods, the fabrication process is somewhat complicated. The exciton distribution profile and the doping concentration must be carefully tuned to achieve a balanced exciton distribution in each emissive layer [16–17]. Moreover, these WOLEDs suffer spectra changes upon different applied voltages and/or operation times, which are likely due to changes in the carrier recombination region, exciton distribution, and/or differential aging of the different emitting materials [18, 19, 22]. WOLEDs made by combining blue OLEDs with a color down-conversion layer have been proposed to produce white emission with improved color stability and a simpler

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fabrication process [20–25]. In principle, when utilizing the down-conversion phosphors approach, a part of the blue light emitted from an OLED is passed through a yellow or red phosphor layer, resulting in white light emission. More specifically, white emission can be generated by the superposition of the non-absorbed blue light emitted by an OLED and the red emission that is emitted by red phosphorus due to absorption of the blue light from the OLED [27–29]. The physical processes in the phosphor layer of a down-conversion device can be described by processes where the photon emitted by the blue OLED is absorbed by the phosphor and re-emitted at a longer wavelength [30].

In this paper, we demonstrated organic-inorganic hybrid WOLEDs that are fabricated by combining highly efficient bis(3,5-difluoro-2-(2-pyridyl)phenyl-(2-carboxypyridyl) iridium(III) (FIrpic) based blue phosphorescent OLED and red inorganic phosphor layers using a down-conversion method. A blue phosphorescent OLED is coupled with a down-converting red inorganic phosphor layer. The phosphor layers absorb emissions from the blue OLED and emit red light according to their intrinsic properties [26]. By combining the unabsorbed blue emission with the red emission, the resulting WOLEDs exhibit broadband spectra with 1931 Commision International de L'Eclairage (CIE) coordinates of (0.33, 0.41). These devices also demonstrate high color stability over a wide range of driving voltages and possess a current efficiency of 11.4 cd/A and a power efficiency of 4.9 lm/W at a luminance of 1000 cd/m^2. This method can be implemented with an easy fabrication method and provides improved color stability and device stability.

Experimental details

The hybrid WOLEDs consist of a vacuum-deposited FIrpic-based phosphorescent OLED (on an indium tin oxide (ITO)-coated glass substrate) and a surface color conversion layer that contains an inorganic phosphor that is applied to the outside surface of the ITO glass, as a shown in Figure 1. Figure 1(a) shows the schematic diagram of the blue phosphorescent OLEDs (as a reference). The WOLED with the down-color conversion layer (i.e., a red inorganic phosphor) is shown in Figure 1(b). The red phosphor layer (GLLEPE CO., LTD, red phosphor ink) was fabricated via spin coating on the outside of the ITO glass substrate. This was cured in an oven at 80°C for 30 min. The thickness of the cured layers were measured with a 2D surface profiler (KLA_Tencor, P-16+). The photoluminescence (PL) and absorption spectra of the phosphor layer were recorded with a spectrofluorometer (Horiba NanoLog-C) and a UV-visible spectrophotometer (Dong-il SHIMADZU Corp., UV-2600), respectively. The OLED devices were fabricated on plasma-treated, ITO-coated glass substrates. The substrates were then brought into a thermal evaporation chamber (SUNICEL 200Plus), which

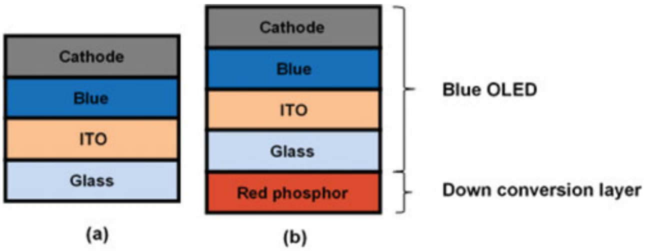

Figure 1. Schematic structures of (a) blue OLEDs as a reference and (b) hybrid WOLEDs with a down-color conversion layer (red phosphor layer).

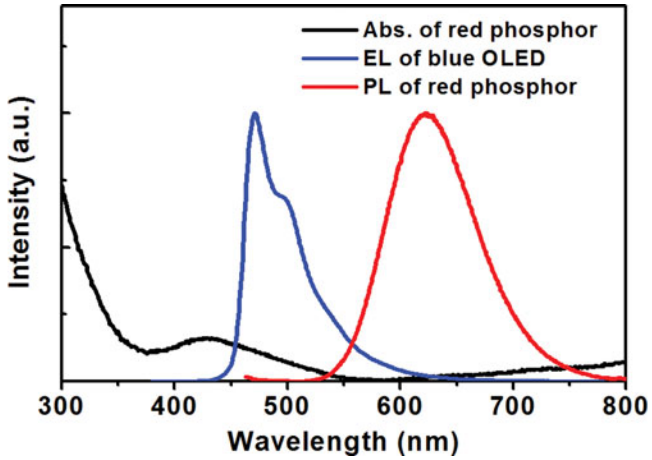

Figure 2. Normalized EL spectrum of a blue OLED and the absorption/PL spectra of the color conversion layer (red phosphor ink).

was enclosed within a glove box filled with N_2 gas. Vacuum deposition was done under high vacuum (~7 × 10^{-7} torr). The blue OLED device structures had the following sequence: glass substrate / ITO(150 nm) / HAT-CN (10 nm) / NPB (50 nm) / TCTA (5 nm) / mCBP : FIrpic (10 wt.%, 30 nm)/TmPy PB (60 nm) / Liq (1 nm) / Al (100 nm). Dipyrazino[2,3-f:2′,3′-h]quinoxaline-2,3,6,7,10,11- hexacarbonitrile (HAT-CN) was used as the hole-injection layer, N,N0-bis-(1-naphyl)-N,N0 -diphenyl-1,10-biphenyl-4,40-diamine (NPB) was used as the hole-transport layer,tris(4-car bazoyl-9-ylphenyl)amine (TCTA) was used as the electron-blocking layer, 4,40-bis(3-methyl carbazol-9-yl)-2,20-biphenyl (mCBP) : bis(3,5-difluoro-2-(2-pyridyl)phenyl-(2-carboxypyridyl) iridium(III) (FIrpic) was used as the emitting layer, 3,3′-[5′-[3-(3-Pyridinyl)phenyl][1, 1′:3′,1″-terphenyl]-3,3″-diyl]bispyridine (TmPyPB) was used as the electron-transport layer, 8-hydroxy-quinolinato lithium (Liq) was used as the electron injection layer, and Al was used as the top cathode. The current density–voltage (*J-V*) and luminance–voltage (*L-V*) characteristics of the devices were measured by using a source-measure unit (Keithley 236) and a calibrated photodiode (Photo Research Inc., PR-670).

Results and discussion

The absorption and photoluminescence (PL) spectra (excited at 621 nm) of the inorganic phosphor are shown in Figure 2. The electroluminescence (EL) spectrum of the blue OLED is also shown in Figure 2. The inorganic phosphor shows an absorption that matches well with the emission of the blue OLEDs, which allows the inorganic phosphor to absorb the blue photo energy and effectively convert it to a red emission via the PL process.

The normalized EL spectra of devices with different phosphor thicknesses at a current density of 10 mA/cm^2 are shown in Figure 3. The blue emission spectra experience a red-shift as the thickness of the phosphor layer increases. The EL spectra of the WOLEDs also contain a red component (from the inorganic phosphor), which absorbs a part of the blue electrophosphorescence of the FIpic. This is then converted to a red fluorescence. The WOLEDs emit white light due to the final mixture of the color-converted red light and the unabsorbed blue light. The red emission from the phosphor grows in relation to the blue emission as the red phosphor thickness is increased [22]. By simply varying the phosphor thickness, the color of the WOLEDs can be adjusted significantly. The CIE coordinates and the corresponding

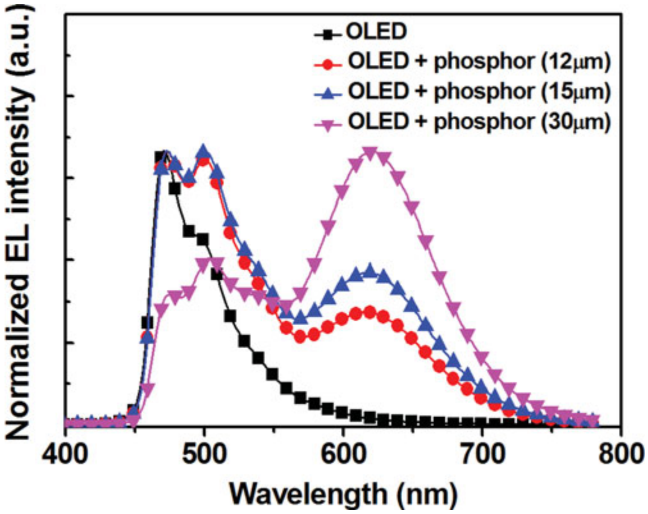

Figure 3. Normalized EL spectra as a function of the phosphor thickness at a current density of 10 mA/cm^2.

photographs of the operating devices are shown in Figure 4. Figure 4(a) shows changes of the CIE color coordinates according to different adjustments to the phosphor layer. The blue OLED shows only the blue emission of FIrpic (with peaks at 471 nm and 495 nm) and has CIE coordinates near (0.16, 0.32). The CIE coordinates of the WOLEDs shift from (0.30, 0.41), which is a bluish white, to (0.45, 0.42), which is a reddish white, as the thickness of the phosphor layer is increased from 12 to 30 μm. As shown in Figure 4(b), by combining the red phosphor layer with the blue phosphorescent OLEDs, the blue emission is modified to yield a uniform white emission. The WOLED device with a 15-μm-thick phosphor layer shows a warm white light emission with CIE coordinates of (0.33, 0.41). The normalized EL spectra of the WOLEDs with a 15-μm-thick red phosphor layer, at different current densities, are shown in Figure 5. The spectra corresponding to current densities between 4 and 50 mA/cm^2 did not change significantly in the devices, demonstrating the excellent color stability of WOLEDs.

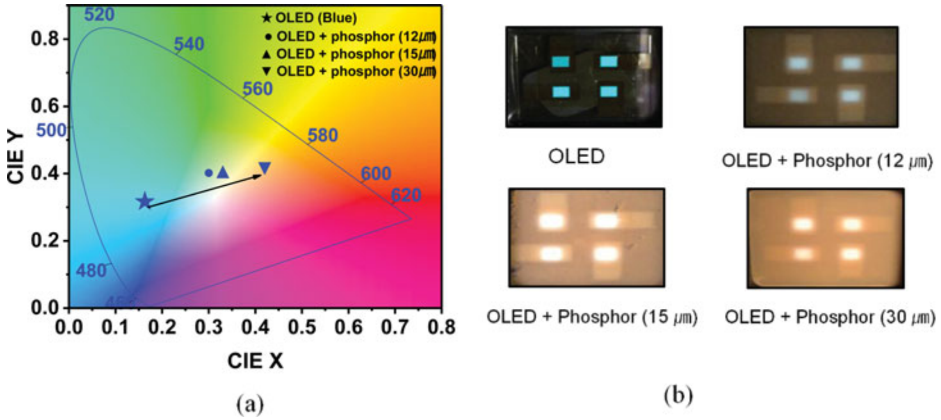

Figure 4. (a) CIE color coordinates of the phosphor as a function of the thickness and (b) photographs of the light-emitting OLED devices as a function of the phosphor thickness.

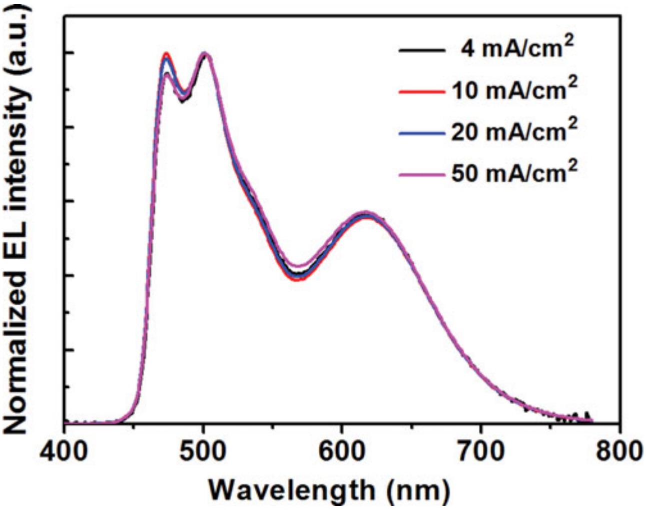

Figure 5. Normalized EL spectra of the WOLEDs with a 15-μm-thick red phosphor at different current densities.

Figure 6(a) shows the *J*-*V*-*L* characteristics of the blue phosphorescent OLEDs (as a reference) and the white OLEDs with different phosphor layer thicknesses. Since the phosphor layer does not alter the electrical characteristics of the device, OLEDs with and without the phosphor layer show nearly identical current densities. All devices exhibit a turn-on voltage at around 4 V for 1 cd/m^2. However, as a shown in Figure 6(b), WOLEDs with a larger thickness show slightly lower luminance compared to the blue OLEDs. For example, the blue phosphorescent OLED (as a reference) has a luminance 1107 cd/m^2 at 6.9 V. However, the WOLEDs with phosphor layers with thicknesses of 12, 15, and 30 μm exhibited luminance of around 780, 760, and 553 cd/m^2, respectively, at 6.9 V. Due to the conversion loss, the luminance decreases as the phosphor layer thickness increases [24].

The device efficiencies, as a function of the current density, are shown in Fig. 6(c). The blue phosphorescent devices show a luminous (power) efficiency of 26.6 cd/A (12.1 lm/W) at 1000 cd/m^2. The WOLEDs that possess color conversion layers containing 12, 15, and 30-μm-thick phosphor layers show luminous (power) efficiencies of 22.6 cd/A (9.9 lm/W), 11.4 cd/A (5.0 lm/W), and 11.0 cd/A (4.6 lm/W), respectively. The performance of these devices is summarized in Table 1.

Summary

In summary, we demonstrated a facile fabrication method for hybrid white OLEDs that are made by combining blue-phosphorescent OLEDs and a printed red phosphor. WOLEDs with an appropriate color conversion layer exhibit a luminous (power) efficiency of 11.4 cd/A (4.9 lm/W) with a CIE coordinate of (0.33, 0.41) at a luminance of 1000 cd/m^2. By employing phosphor coating, the thickness can be adjusted to manipulate the color of the white light. The proposed method yields devices that possess good lifetimes under normal conditions; this is accomplished due to the fact that we use an inorganic phosphor instead of an organic phosphor. This approach can be implemented by using easy fabrication techniques and provides improved color stability in the resultant devices. We expect to be able to control the spectra in organic-inorganic hybrid OLEDs by using this down-conversion method.

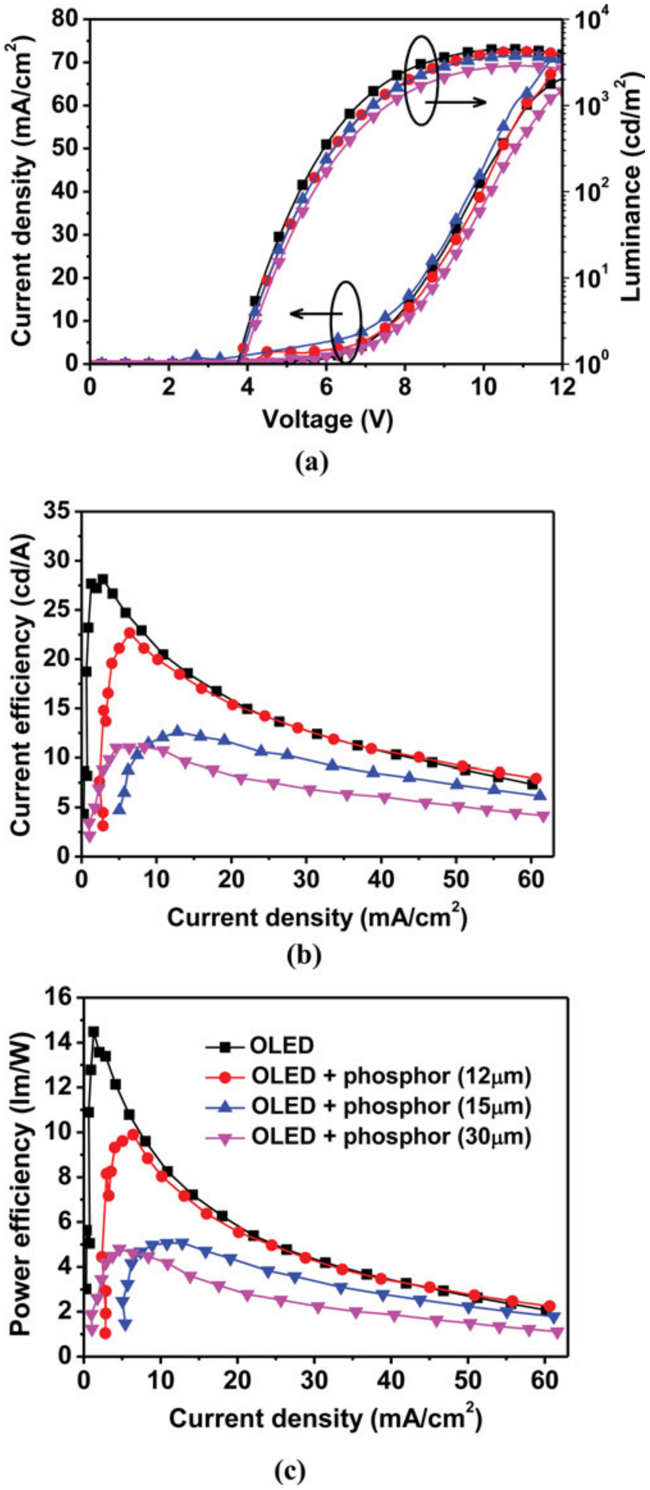

Figure 6. (a) Current density-voltage-luminance characteristics, (b) current density-current efficiency characteristics, and (c) current density-power efficiency characteristics of the OLEDs devices.

Table 1. Device performance at 1000 cd/m^2.

Device	Current efficiency (cd/A)	Power efficiency (lm/W)	Quantum efficiency (%)	CIE coordinate	
				X	Y
OLED (reference)	26.6	12.1	13.4	0.16	0.32
OLED+phosphor (12 μm)	22.6	9.9	10.8	0.3	0.41
OLED+phosphor (15 μm)	11.4	5.0	5.5	0.33	0.41
OLED+phosphor (30 μm)	11.0	4.6	5.8	0.45	0.42

Acknowledgments

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