Energy transfer process in white organic light-emitting devices based on carbazole/thioxanthene-S,S-dioxide host material

YOUNG PYO JEON,1 DONG HYUN PARK,2 KEON-HO YOO,3 AND TAE WHAN KIM2,4,*

1The Research Institute of Industrial Science, Hanyang University, Seoul 04763, South Korea
2Department of Information Display Engineering, Hanyang University, Seoul 04763, South Korea
3Department of Physics and Research Institute for Basic Sciences, Kyung Hee University, Seoul 02447, South Korea
4Department of Electronics and Computer Engineering, Hanyang University, Seoul 04763, South Korea
*twk@hanyang.ac.kr

Abstract: Phosphorescent white organic light-emitting devices (Ph-WOLEDs) fabricated utilizing carbazole/thioxanthene-S,S-dioxide (EBCz-ThX) host material were investigated to enhance their efficiency by understanding the energy transfer process from the host to the dopant materials. Time-resolved photoluminescence (PL) spectra at room and low temperatures demonstrated that the energy transfer occurs between the EBCz-ThX host and the bis(2-benzothiophen-2-ylpyridine)(acetylacetonate)-iridium(III) (Ir(bt)2acac) orange dopant or the (bis(2-(4,6-difluorophenyl)pyridyl-N,C2′)iridium(III) picolinate (FIRpic) blue dopant. The energy transfer process was inferred from the PL decay curves of Ir(bt)2acac doped EBCz-ThX with doping concentrations of 5, 10, and 20%. The quantum efficiency in the Ph-WOLEDs with 5% Ir(bt)2acac doped EBCz-ThX is the highest since its energy transfer process from EBCz-ThX to Ir(bt)2acac involves the charge transfer states and is most efficient. The quantum efficiency of the Ph-WOLEDs with 5% Ir(bt)2acac doped EBCz-ThX emission layers at 1,000 cd/m² showed the highest value of 10.2%, and the commission internationale de l'Eclairage (CIE) coordinates were (0.45, 0.46), indicative of the warm white emission.

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1. Introduction

Organic light-emitting devices (OLEDs) have currently emerged as excellent candidates for promising applications in wearable/flexible displays and light sources for portable devices and automobiles due to their high flexibility and superior surface emitting property [1–4]. The prospect of white OLEDs (WOLEDs) has led to substantial research and development efforts to fabricate high performance WOLEDs with various structures utilizing the three primary colors, complementary colors, or single white-light emitting materials [5–9]. Due to a wide emission advantage of phosphorescent emissive materials, the research on the phosphorescent WOLEDs (Ph-WOLEDs) based on complementary colors has been widely reported with great performance with high coverage of white emission region [9,10]. However, Ph-WOLEDs based on complementary color still have some issues that need to be addressed, such as exciton quenching, lifetime, and color stability. Recently, a research on WOLEDs based on the delayed fluorescence has been reported to solve these problems by improving the management of singlet and triplet exciton [11]. However, these problems can be also solved by well-understanding the energy transfer process and emission process of excitons participating in the Ph-WOLED emission process.

Because more than two emissive dopants are essential to fabricate Ph-WOLEDs, engineering the energy transfer and the band-gap alignment between the emissive dopants and the host materials is difficult. In particular, the efficiency of Ph-WOLEDs tends to be low due to the exciton quenching in the presence of two or three dopants in a host material, resulting in the distortions of the molecules in the emission layer (EML) [12, 13]. If the exciton energy transfer from the host to the emissive dopants is inefficient, the non-transferred energy appears as a thermal energy, also resulting in distortions of the molecules on the EMLs [14, 15]. Low glass transition temperature ($T_g$) of host materials leads to the easy crystallization, making the morphological stability poor, which reduces device reliability [16]. Although few studies on the WOLEDs with a host material doped two dopants have been reported [17], energy transfer process based on continuous-wave (cw) and time-resolved photoluminescence (PL) spectra from the host and dopants have not been performed.

In this research, Ph-WOLEDs based on complementary colors using carbazole/thioxanthene-S, S-dioxide (EBCz-ThX), an amorphous host material designed to have a large optical bandgap, a high-lying highest occupied molecular orbital (HOMO) energy level, and a high-$T_g$ [18], were fabricated, and their electrical and optical characteristics were investigated to identify the energy transfer process from the EBCz-ThX host to blue or orange dopant materials and to enhance the efficiency of WOLEDs. The cw and time-resolved PL spectra of the EBCz-ThX with various concentrations of orange emissive dopants at room and low temperatures were measured to clarify the energy transfer process between the host and the dopants.
2. Experimental details

Figure 1(a) shows a schematic diagram for the device structure of the Ph-WOLEDs based on EBCz-ThX host material with a 4,4′-cyclohexylidenebis[N,N-bis(4-methylphenyl)benzenamine] (TAPC) hole transport layer (HTL) and a 2,2′,2″-(1,3,5-benzinetriyl)-tris(1-phenyl-1'H-benzimidazole) (TPBi) electron transporting layer (ETL). The bis(2-benzo[b]thiophen-2-ylpyridine)(acetylacetonate)-iridium(III) (Ir(bt)acac) and (bis(2-(4,6-difluorophenyl)pyridyl-N,C2′)iridium(III) picolinate (FIrpic) were used as phosphorescent orange and blue emissive dopants, respectively. The HOMO and the lowest unoccupied molecular orbital (LUMO) energy levels, quoted from the recent papers [19, 20], of the organic materials are presented in Fig. 1(b). Electrons are injected from the cathode and transported through LUMO of the ETL to LUMO of EBCz-ThX, while the holes are injected from the anode and transported through HOMO of the HTL to HOMO of EBCz-ThX. The HOMO and the LUMO energy levels of the organic materials are well-aligned to provide efficiently the electrons and the holes in the EML. The chemical structure of the EBCz-ThX is drawn in Fig. 1(c).

Fig. 1. (a) A schematic diagram, (b) the corresponding energy band diagrams for the white organic light-emitting devices based on carbazole/thioxanthene-S,S-dioxide (EBCz-ThX), and (c) the chemical structure of EBCz-ThX.

Patterned ITO-coated glass substrates were cleaned with acetone and methanol at 60 °C for 10 min, and then thoroughly rinsed in de-ionized water. After the chemically cleaned ITO substrates were dried with N2 gas with a purity of 99.999%, the dried ITO substrates were cured using UV-zone treatment for 20 min. Then the ITO substrates were introduced into the evaporation chamber, and the organic layers and the metal layer were deposited on the ITO substrates and a system pressure below 1.73 × 10−4 Pa. The deposition rates of the organic layers and the metal electrode, which were controlled by using a quartz crystal deposition rate/thickness monitor (Sigma, SQM-160), were approximately 1 and 2 Å/s, respectively.

The time resolved PL measurements were performed by using an X-ray fluorescence spectrometer (FL920, Edinburgh Instruments) equipped with a pulsed laser operating at 375 nm, and the temperature was controlled from room temperature (RT) to 80 K by using liquid nitrogen. The current-voltage characteristics were measured on a programmable electrometer with built-in current and voltage measurement units (M6100, McScience). The brightness and the electroluminescence (EL) spectra were measured by using a luminescence spectrometer (CS-1000, Minolta). All measurements of the electrical characteristics for the Ph-WOLEDs were carried out at RT under atmospheric conditions.
3. Results and discussion

Figure 2 shows (a) the normalized cw PL spectra and (b) the time resolved PL spectra of the blended solution of EBCz-ThX host material and the orange or blue dopants in chlorobenzene solvent. The excitation light with a wavelength of 465 nm is absorbed by the EBCz-ThX host molecules, and the energy of excitons in the EBCz-ThX generated by the absorbed light is transferred to the Ir(bt)$_2$acac or FIrpic molecules. The dominant and the second vibrational PL peaks of the blended solution of the Ir(bt)$_2$acac and the EBCz-ThX with a mixing ratio of 1:10 are at 569 and 600 nm, respectively, and those of the blended solution of the FIrpic and the EBCz-ThX with a mixing ratio of 1:10 are at 480 and 497 nm. Because the emission peak of the EBCz-ThX is well matched to the absorption peak of the FIrpic [18], the energy transfer process in the blended solution of the FIrpic and the EBCz-ThX is the Förster energy transfer. On the other hand, because the absorption peak of Ir(bt)$_2$acac is not well matched to the emission peak of the EBCz-ThX, Dexter energy transfer is the primary energy process between the EBCz-ThX to Ir(bt)$_2$acac [21–24].

The PL decay data in Fig. 2(b) were fitted by single-term exponential decay model:

\[ y(t) = A_0 \cdot \exp\left(-\frac{t}{\tau}\right), \]

where \( \tau \) is the PL decay time and \( A_0 \) is a constant. The decay time of the dominant peak of the blended solution of the FIrpic and the EBCz-ThX is 68 \( \mu \)s and that of the blended solution of the Ir(bt)$_2$acac and the EBCz-ThX is 109 \( \mu \)s. The PL decay time of the blended solution of the FIrpic and the EBCz-ThX was much faster than that of the blended solution of the Ir(bt)$_2$acac and the EBCz-ThX. This decay time difference in the solution of the FIrpic and Ir(bt)$_2$acac doped EBCz-ThX is associated with the difference of the energy transfer process, i.e., Förster and Dexter energy transfer.

The optical properties of the thin (30 nm) film of the Ir(bt)$_2$acac doped EBCz-ThX with various doping concentrations of 5%, 10%, and 20% were studied. A similar study with the film of the FIrpic doped EBCz-ThX has been done by us [17]. Figure 3 shows the cw PL intensity of 5%, 10%, and 20% Ir(bt)$_2$acac doped EBCz-ThX thin films. The dominant PL peaks for 5% and 10% Ir(bt)$_2$acac doped EBCz-ThX appear at almost same position, i.e., 565 and 564 nm, while the dominant peak is slightly red-shifted in the 20% Ir(bt)$_2$acac doped-EBCz-ThX due to the increase in the intermolecular interaction of the Ir(bt)$_2$acac dopant molecules [25, 26]. The intensity of PL for 10% Ir(bt)$_2$acac doped EBCz-ThX is the strongest due to low exciton quenching [27, 28].
The PL decay curves in Figs. 3(b) and (c) were also fitted by Eq. (1). The PL decay time of 5%, 10%, and 20% Ir(bt)2acac doped EBCz-ThX are 426, 421, and 401 μs for dominant peak and 444, 420, and 392 nm for the second vibrational peak, respectively. The longer decay time of 5% Ir(bt)2acac doped EBCz-ThX is ascribed to excitons trapped at defects [26, 29].

The low temperature (80 K) cw PL spectra and time resolved PL decay time of 5%, 10% and 20% Ir(bt)2acac doped EBCz-ThX films were measured to clarify the longer decay time of the 5% Ir(bt)2acac doped EBCz-ThX, as shown in Figs. 4(a), (b), and (c). When compared with RT data in Fig. 3(a), the intensity of the secondary peak in Fig. 4(a) is enhanced with respect to that of the main peak at low temperature PL, especially in the case of 5% Ir(bt)2acac doped EBCz-ThX. This remarkable enhancement in 5% Ir(bt)2acac doped EBCz-ThX is ascribed to the existence of the charge transfer (3CT) state, as will be explained later.

The low temperature PL decay curve of 10% and 20% Ir(bt)2acac doped EBCz-ThX were also fitted by Eq. (1). The decay times of 10% and 20% Ir(bt)2acac doped EBCz-ThX 497 and
443 μs for dominant peaks and 412 and 400 μs for secondary peaks, respectively. However, the low temperature PL decay curve of 5% Ir(bt)$_2$acac doped EBCz-ThX is not consistent with the single term exponential decay model and was fitted by a multi-term exponential decay model.

\[ y(t) = A_1 \cdot \exp \left( -\frac{t}{\tau_1} \right) + A_2 \cdot \exp \left( -\frac{t}{\tau_2} \right), \]

This multi-term exponential decay model is generally good in inhomogeneous molecular environments [30, 31]. The term with a very short lifetime in 5% Ir(bt)$_2$acac doped EBCz-ThX is associated with the $^3$CT state, an inter-molecular energy state between donor and acceptor with a very weak exciton binding energy [32–34]. The secondary peak at 619 nm in 5% Ir(bt)$_2$acac doped EBCz-ThX is close in energy to the vibrational PL emission states of EBCz-ThX [18] and greatly enhanced through the interaction with those states. The ligand of Ir(bt)$_2$acac or the heavy metal of dopant might develop the $^3$CT state in Ir(bt)$_2$acac doped EBCz-ThX at low doping concentration of 5% [32]. The shorter PL decay times $\tau_1$ of 5% Ir(bt)$_2$acac doped EBCz-ThX are 6.7 μs for the dominant peak and 64.5 μs for the secondary peak and the longer PL decay times $\tau_2$ of 5% Ir(bt)$_2$acac doped EBCz-ThX are 472 μs for the dominant peak and 416 μs for the secondary peak. The longer PL decay time $\tau_2$ of 5% Ir(bt)$_2$acac doped EBCz-ThX at low temperature is comparable to those of 10% and 20% Ir(bt)$_2$acac doped EBCz-ThX at the same temperature. Figure 4(d) shows a schematic diagram of the energy transfer process in 5% Ir(bt)$_2$acac doped EBCz-ThX. Because the thermal excitation from the $^3$CT state to the triplet metal-to-ligand charge transfer state ($^3$MLCT) in 5% Ir(bt)$_2$acac doped EBCz-ThX is difficult at low temperature [35, 36], the intensity of the secondary peak associated with the $^3$CT state is high. In general, the shorter exciton lifetime of 5% Ir(bt)$_2$acac doped EBCz-ThX is involved in longer lifetime of Ph-OLEDs because the shorter exciton lifetime reduces triplet-triplet annihilation [28].

Fig. 5. (a) Luminance-voltage, (b) current density-voltage, (c) quantum efficiency-luminance, and (d) electroluminescence spectra for white organic light emitting devices based on 5%, 10% and 20% (W-I, W-II, and W-III) Ir(bt)$_2$acac doped EBCz-ThX.

Ph-WOLEDs using EBCz-ThX host, FIrpic blue dopant and Ir(bt)$_2$acac orange dopant were fabricated and their electrical and optical properties were investigated. The doping
concentration of FIrpic was fixed at 10% and that of Ir(bt)\textsubscript{2}acac was varied at 5, 10, and 20%, which were labeled by W-I, W-II, and W-III, respectively. Figure 5 shows (a) the luminance-voltage, (b) the current density-voltage, (c) the quantum efficiency-luminance, and (d) the EL spectra for the WOLEDs. The maximum luminances of W-I, W-II and W-III at 16, 17.5 and 22 V are 12890, 9688, and 5370 cd/m\textsuperscript{2}, respectively. The quantum efficiencies of W-I, W-II, and W-III at 100 cd/m\textsuperscript{2} are 11.5%, 5.8%, and 7.3%. The current efficiency, power efficiency, and external quantum efficiency of the WOLEDs are summarized in Table 1. The quantum efficiencies of the WOLEDs are not higher than 78% of recently reported WOLEDs [37]. However, our results are still worthy of report for the WOLEDs based on EBCz-ThX host material. The efficiency of W-I is the highest among the WOLEDs because the emission in 5% Ir(bt)\textsubscript{2}acac doped EBCz-ThX involves the 3CT state, as explained in the above. Even though the brightness of W-III is the lowest among the WOLEDs, the quantum efficiency of W-III is 25% higher than that of W-II because the higher doping ratio increases the trap sites in the EML and the increased trap sites in W-III reduce the current flows in the device at the higher operation voltage and the lower current density of W-III. The commission internationale de l’Eclairage (CIE) coordinates for W-I at the maximum brightness were (0.45, 0.46), indicative of warm white emission, due to a good balance between orange and blue emissions.

| Table 1. Current efficiency ($\eta_l$), power efficiency ($\eta_p$), and external quantum efficiency ($\eta_{ext}$) at 100 and 1000 cd/m\textsuperscript{2} for white organic light emitting devices based on 5%, 10% and 20% (W-I, W-II, and W-III) Ir(bt)\textsubscript{2}acac doped EBCz-ThX. |
|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
|                | @ 100 cd/m\textsuperscript{2} | @ 1000 cd/m\textsuperscript{2} |                |                |                |                |
|                | $\eta_l$ (cd/A) | $\eta_p$ (lm/W) | $\eta_{ext}$ (%) | $\eta_l$ (cd/A) | $\eta_p$ (lm/W) | $\eta_{ext}$ (%) |
| W-I            | 29.4            | 14.2           | 11.5           | 26.2           | 8.6            | 10.2           |
| W-II           | 14.5            | 4.5            | 5.8            | 12.2           | 2.8            | 4.9            |
| W-III          | 17.8            | 4.1            | 7.3            | 16.3           | 2.7            | 6.7            |

4. Summary and conclusions

The energy transfer processes from the host to the dopants of the WOLEDs based on the bipolar EBCz-ThX host material were investigated to enhance their quantum efficiency. The 3CT state was revealed by analysis on PL decay of Ir(bt)\textsubscript{2}acac doped EBCz-ThX with 5% doping concentration. The WOLED based on EBCz-ThX exhibited the highest quantum efficiency with 5% Ir(bt)\textsubscript{2}acac and 10% FIrpic dopants. The maximum current efficiency and quantum efficiency of WOLEDs with the optimized doping conditions of EML were 29.4 cd/A and 11.5% at 100 cd/m\textsuperscript{2}, respectively. The WOLEDs emitted warm white emission due to a good balance between orange and blue emissions, which was achieved by optimizing the energy transfer between EBCz-ThX host and the Ir(bt)\textsubscript{2}acac dopant.

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