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Molecular Organic Light-Emitting Diodes Based on a Guest-Host Active Layer: Approaches for Enhancing Device Performance

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Molecular Organic Light-Emitting Diodes Based on a Guest-Host Active Layer: Approaches for Enhancing Device Performance

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We present a comprehensive study of enhancement mechanisms in electroluminescence devices where the active layer is doped with fluorescent dye. We demonstrate two typical systems where the emission mechanism is based on either energy transfer or direct carrier recombination. For the energy transfer mechanism, we find that the maximum quantum efficiency of multilayered devices depends on the nature of the host in addition to the luminescent properties of the guest, by comparing electroluminescence and absolute photoluminescence quantum yields in various guest-host systems. For the direct carrier recombination mechanism using rubrene as the dopant and TPD as the host, we find that the dual electronic nature of rubrene acting as a hole trap and an electron hopping site is responsible for maximizing the carrier recombination on the dopant molecule.

Keywords: light-emitting diode; emission mechanism; energy transfer; direct carrier recombination

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INTRODUCTION

Doping a fluorescent dye into the emitting layer provides several advantages for organic light emitting diodes. With a suitable combination of host and guest molecules, substantial gain in the electroluminescence (EL) efficiency can be achieved.[1] Color tuning of the emitted light can be easily realized throughout the entire visible region along with an improvement of color purity.[2] Device lifetimes are also influenced by the choice of host-guest pairs,[3,4] and the nature of the guest molecule.[5] Recently, we have found that doping can prevent exciplex formation and leads to an increase in the EL efficiency.[6] Exciplex formation is occasionally observed at the organic/organic interface where the difference of ionization potential (I_p) between the organic layers is large, and results in poor EL performance[7-9] due to a decrease of photoluminescence (PL) quantum yield. We have also demonstrated that doping improves temperature dependence of EL quantum efficiency.[10]

The enhancement of EL efficiency by dye doping can be attributed to Förster[11] and/or Dexter[12] energy transfer from host to guest, and also direct carrier recombination at the dopant molecules.[13] In this paper, we discuss the EL enhancement mechanism due to doping in the framework of energy transfer and direct carrier recombination.

EXPERIMENTAL SECTION

The chemical structure of the materials used in this study are shown in Fig.1. We used N,N'-diphenyl-N,N'-bis(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD) as the hole transport layer (HTL), tris-(8-hydroxyquinolinato) aluminum (III) (Alq_3) or tris-(4-methyl-8-hydroxyquinolinato) aluminum (III) ($Almq_3$)[14] as the electron transport layer (ETL). Rubrene (Aldrich Chem. Co.) and N,N'-diethyl quinacridone (DEQ) were used as the guest materials. DEQ was synthesized from quinacridone (TCI America Ltd.) and iodoethane by use of a base and a phase transfer catalyst.[6]

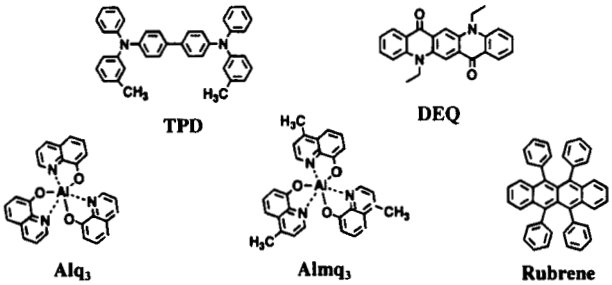

FIGURE 1 Chemical structures of materials used in this study.

The device structure consists of ITO/HTL/EML/ETL/Mg:Ag (12:1 by weight) on a glass substrate. For the devices using DEQ as the guest, the thicknesses of HTL, EML and ETL are 50 nm, 35 nm and 35 nm, respectively. EML consists of the films where Alq_3 or $Almq_3$ is used as the host for DEQ. For the devices using rubrene as the guest, TPD is employed as the host. For spectroscopic characterization and absolute PL quantum yield (ϕ_{PL}) measurements, pure and composite films similar to those used as the EML in the devices were prepared on silica substrates. Absolute ϕ_{PL} was measured using an integrating sphere inside a glove box purged with dry nitrogen. Details of the PL measurements are described elsewhere.[15] The measurement of EL spectra was also carried out inside the glove box with the luminescence collected through an optical fiber and directed into the spectrograph outside the glove box. Current density-voltage-luminance measurements were taken using a Keithley 238 high current source measurement unit and a Minolta LS-110 luminance meter. Device performance was evaluated based on the external quantum efficiency (η_{EL}) defined as the ratio of the number of photons emitted to the number of injected electrons.[6]

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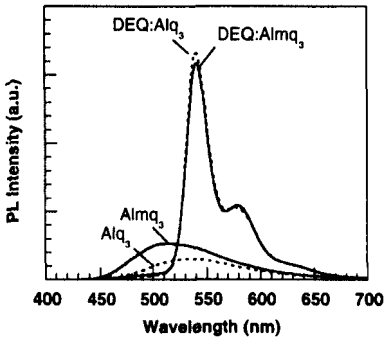

FIGURE 2 Photoluminescence spectra of films of Alq_3, $Almq_3$, DEQ-doped Alq_3 and DEQ-doped $Almq_3$. DEQ concentrations in Alq_3 and $Almq_3$ are 0.85 mol% and 0.7 mol%, respectively.

ENERGY TRANSFER MECHANISM

The external EL quantum efficiency (η_{EL}) can be expressed by the following equation:[16]

$$\eta_{EL} = \alpha \gamma \eta_r \phi_{PL} \tag{1}$$

where, α is the fraction of photons that are coupled out from the device surface and is approximately given by $\alpha = 1/(2n^2)$ (n is the refractive index of the emissive medium),[17] η_r is the production efficiency of singlet exciton and ϕ_{PL} is the absolute PL quantum yield. The probability of carrier recombination, γ, depends on the carrier injection efficiency from both electrodes and the mobilities in the organic films. A comparison between η_{EL} of the device and ϕ_{PL} of the solid film used as the emitting layer allows one to study the contribution of doping to ϕ_{PL} and to the other determining factors of the EL efficiency.

Figure 2 shows the PL spectra of films of $Almq_3$, Alq_3, DEQ-doped $Almq_3$, and DEQ-doped Alq_3. The PL yields of these films are summarized in Table 1. The ϕ_{PL} of Alq_3 film (25 ± 4%) is in good agreement with the published value.[18] The PL efficiency of $Almq_3$ solid film (42 ± 4%) is 1.7

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TABLE 1. Absolute photoluminescence quantum yield of films of Alq_3, $Almq_3$, DEQ-doped Alq_3 and DEQ-doped $Almq_3$.

Material	PL Quantum Yield (%)[a]
Alq_3	25 ± 4
$Almq_3$	42 ± 4
DEQ: Alq_3[b]	74 ± 4
DEQ: $Almq_3$[c]	72 ± 4
DEQ in solution	75 ± 4 [d]

a) measured with an integrating sphere using a 400nm-thick film.
b) DEQ concentration is 0.85mol%.
c) DEQ concentration is 0.7 mol%.
d) measured in a chloroform solution (~10^{-5}M). The PL yield was determined relative to a MeOH solution of Rhodamine 590 as a standard.

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times larger than that of Alq_3. Similar PL ratio is observed for dilute solutions of Alq_3 and $Almq_3$.

Once Alq_3 and $Almq_3$ films are doped with DEQ, the PL emission primarily arises from the dopant molecule. No contribution from the hosts is observed in the PL spectra at the optimal dopant concentration. These results suggest that energy transfer from the host to the guest molecule is extremely efficient in these systems.

Figure 3 compares the ϕ_{PL} of Alq_3 and $Almq_3$ films doped with DEQ as a function of dopant concentration. The maximum ϕ_{PL} for both DEQ: Alq_3 and DEQ: $Almq_3$ films is obtained at 0.5~1.0 mol%. It should be noted that for both composite films, the same ϕ_{PL} value (~75 %) is achieved at this optimal concentration even though the ϕ_{PL} of $Almq_3$ is 1.7 times larger than that of Alq_3. These results suggest that the maximum ϕ_{PL} for these composite films is primarily determined by the guest molecule, and independent from the host molecule as long as efficient energy transfer has been achieved from host to guest.

After photoexcitation of the host molecule in a solid composite film, the emission from the guest molecule takes place via two consecutive

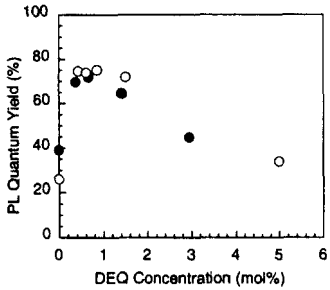

FIGURE 3 Absolute PL yield of the films of Alq_3 (open circle) and $Almq_3$ (filled circle) as a function of DEQ concentration

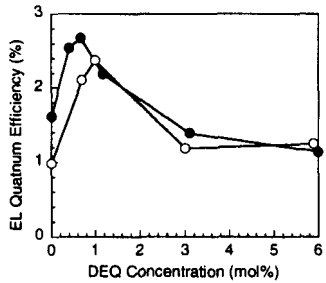

FIGURE 4 External EL quantum efficiency of multilayered devices as a function of DEQ concentration. EML is composed of Alq_3 (open circle) or $Almq_3$ (filled circle) doped with DEQ.

processes: (i) energy transfer from the host to the guest through dipole-dipole interaction and/or electron exchange, and (ii) fluorescence from the excited guest. As shown in Table 1, a chloroform solution of DEQ (~10^{-5} M) has a ϕ_{PL} of 75 ± 5%, a value which is identical to the absolute PL quantum yields of the solid composite films. These results imply that the efficiency of the first process, energy transfer from the host to the guest molecules, is close to unity. A more detailed analysis and discussion of the ϕ_{PL} of solid composite films is reported elsewhere.[15]

Figure 4 depicts the η_{EL} of the multilayered devices as a function of dopant concentration, where the emitting layer consists of Alq_3 or $Almq_3$ doped with DEQ. At low DEQ concentration, a more significant increase in η_{EL} is measured for $Almq_3$-based devices in comparison with Alq_3-based devices. This concentration dependence in η_{EL} seems to deviate from that of ϕ_{PL}. This result is also inconsistent with the results of ϕ_{PL} where doping with DEQ led to the same maximum value for ϕ_{PL} regardless of the host materials used.

Figure 5 shows a simplified schematic diagram of EL and PL processes in host-guest systems. The critical difference between EL and PL is the

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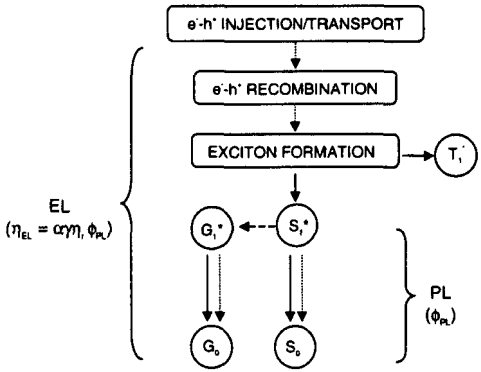

FIGURE 5 Schematic diagram of emission process in electro-luminescence vs. photoluminescence.

process that leads to the creation of the singlet exciton of the host molecule (S_1^*). [19] Decay processes of the singlet exciton should be the same in PL and EL, assuming there is no interaction between the singlet exciton and the charge carriers. As shown in Fig.3, the PL quantum yield of the guest-host system is governed by the ϕ_{PL} of guest molecule. Hence, if the same number of the host molecules in singlet excited state (S_1^*) are created by carrier recombination, one would expect the same maximum value for η_{EL} when using the same guest molecule in the emitting layer. However, this is not the case for these devices where films of DEQ:Alq_3 and DEQ:$Almq_3$ are used as the emissive layers. Differences in the maximum value for the EL efficiencies obtained, due to the change of host molecule, may be explained by the difference of the number of singlet excitons created in the host, Alq3 or $Almq_3$.

Figure 6 compares the EL spectra of the devices using films of Alq_3 and $Almq_3$ doped with DEQ as a function of dopant concentration. The device using DEQ-doped Alq_3 shows very small contribution from Alq_3, which disappears upon increasing the concentration of DEQ, consistent with the PL spectra of the films of DEQ-doped Alq_3. In contrast, there is still some

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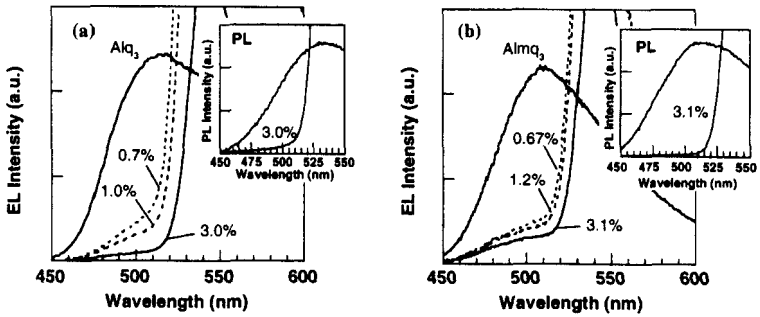

FIGURE 6 Electroluminescence spectra of multilayered devices as a function of DEQ concentration.
(a) Device structure: ITO/TPD(50nm)/EML(35nm)/ Alq_3 (35nm)/Mg:Ag, where EML is composed of Alq_3 doped with DEQ.
(b) Device structure: ITO/TPD(50nm)/EML(35nm)/ $Almq_3$ (35nm)/Mg:Ag, where EML is composed of Almq3 doped with DEQ.
Insets show photoluminescence spectra of EML.

contribution from the host in the EL spectra of the device using DEQ-doped $Almq_3$. (Fig.6 (b)) This is inconsistent with the PL spectra of films of DEQ-doped $Almq_3$, where the host emission is totally quenched by the efficient energy transfer to DEQ. The residual EL emission from $Almq_3$ can be ascribed to mismatching of the width between the doping zone and the carrier recombination zone in $Almq_3$. If the carrier recombination zone is wider than the width of DEQ-doped $Almq_3$ layer, some of the $Almq_3$ excitons, which are created outside the energy transfer radius, can radiatively decay and contribute to the EL spectra. This hypothesis was confirmed by the measurements of the EL spectra of the device as a function of the thickness of DEQ-doped $Almq_3$. A minimum thickness of 53 nm for the doped layer is necessary in order to totally eliminate emission from the host $Almq_3$. This width is much wider than that of the device using DEQ-doped Alq_3 (35nm).

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DIRECT CARRIER RECOMBINATION MECHANISM

In the previous section, we used Alq_3 (or $Almq_3$) as the host for the guest molecule. This is a rational approach based on the accepted EL emission mechanism of a double layered device composed of an aromatic diamine as a HTL and Alq_3 as the active layer.[11] In this device, electron-hole recombination and the creation of Alq_3 exciton occur inside the Alq_3 layer near the HTL/Alq_3 interface since the HTL layer effectively blocks electron injection. Consequently, the selection of Alq_3 (or $Almq_3$) as the host is crucial for both carrier recombination on the guest and energy transfer from host to guest.

Recently, the use of rubrene as a dopant in either Alq_3 or in the hole transporter TPD has been shown to improve device efficiency.[4, 20, 21] Interestingly, the higher efficiency was observed when TPD rather than Alq_3 was selected as the host. In addition, the half-decay time of the initial luminance in these devices was prolonged by more than one order of magnitude when TPD was used as the host.[4] These experimental results cannot be explained by the emission mechanism based on the exciton creation inside Alq_3 layer.

In order to understand the emission mechanism in the device where the active layer consists of rubrene-doped TPD, we have performed extensive studies on devices based on rubrene-doped TPD films, including absolute PL quantum yield, [15] I-V curve measurements,[13] drift mobility[22] and ionization potentials.[23] We measured very high PL yield, close to unity, in rubrene-doped TPD films indicating efficient energy transfer from TPD exciton (TPD^*) to rubrene. We confirmed that rubrene acts as a shallow hole trap in TPD from the results of the shift of I-V curves towards higher voltage and a decrease in drift mobility, with increasing rubrene concentration. These results are consistent with the ionization potential (I_p) of rubrene (~5.4 eV) and TPD (~5.6eV), namely rubrene has slightly lower I_p than that of TPD. Unexpectedly, we have observed an EL emission in single-layered devices

composed of either undoped or doped TPD. EL emission of TPD is an evidence that electrons can be directly injected into TPD, previously known as an electron blocker at the TPD/ Alq_3 interface.

Since electrons can be injected into TPD and energy transfer from TPD^* to rubrene is quite efficient, two emission mechanisms appear to be possible in addition to an energy transfer from Alq_3^* to rubrene. One is direct electron-hole recombination on rubrene molecules. The other is energy transfer from TPD^* to rubrene, which may follow after initial electron-hole recombination on the host TPD near the TPD/Alq_3 interface. One can distinguish between these mechanisms by comparing the spatial relationship of the emission zone and the recombination zone in the emitting layer. If the direct carrier recombination on the guest is the dominant mechanism, the recombination and emission zones should be identical. On the other hand, if energy transfer from Alq_3^* is the dominant mechanism, this should only occur near the TPD/ Alq_3 interface since the emission zone would be determined by the range of Förster energy transfer of Alq_3^* to rubrene. In the case of energy transfer from TPD^*, the emission zone would be governed by the diffusion

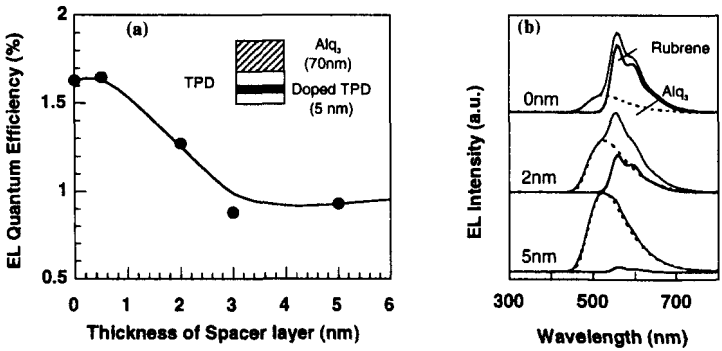

FIGURE 7 (a) External EL quantum efficiency as a function of the thickness of the spacer layer. Inset shows the structure of devices. (b) EL spectra of multilayered devices with different spacer thickness. The EL spectra has been deconvoluted to those of rubrene and Alq_3.

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length of TPD^* and would be greater than that of the recombination zone determined by the penetration depth of electrons.

The maximal range for TPD^*, which is defined as the sum of the diffusion length of TPD^* and the penetration depth of electrons, can be determined by using a doped layer as the sensing layer. Figure 7 shows the EL quantum efficiency of these multilayered devices and their EL spectra as a function of spacer thickness. Within the undoped TPD layer, a doped layer with 5 nm thickness was inserted. (inset of Figure 7(a)) Changing the spacer thickness significantly affects the EL quantum efficiency as well as the EL spectra. By increasing the spacer thickness to 5 nm, the quantum efficiency is quickly reduced and approaches the value of the undoped device. It is obvious that increasing the spacer thickness above 5 nm nullifies the contribution from rubrene in the EL spectra and leads to that characteristic of Alq_3. These results suggest that, in undoped TPD, the maximal range of TPD^* cannot be greater than 5 nm from the Alq_3 interface. On the other hand, energy transfer from Alq_3^* to rubrene can only take place by Förster mechanism whose interaction distance is on the order of 3-4 nm.[24] Hence, the range of energy transfer from either TPD^* and Alq_3^* to rubrene would be also limited to this range.

Figure 8 shows the external EL quantum efficiency of multilayered devices (see inset) as a function of the thickness of the rubrene-doped TPD layer. The primary enhancement in the efficiency occurs within a 5 nm thick region from the Alq_3 interface. The EL efficiency continues to increase up to 20 nm and remains essentially constant going from 20 to 50 nm. These results indicate that the width of the emission zone in rubrene-doped TPD is about 20 nm from the Alq_3 interface. This wide emission zone cannot be explained by energy transfer range of TPD^* or Alq_3^* alone. A plausible mechanism to consider is the direct electron-hole recombination on rubrene.

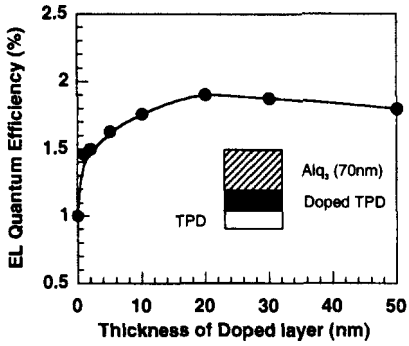

FIGURE 8 External EL quantum efficiency as a function of thickness of rubrene-doped TPD layer. Inset shows the structure of devices. The EL quantum efficiency were determined at 10 mA/cm^2.

This emission mechanism would require that rubrene doping extends the electron penetration depth beyond 5 nm.

In order to study the nature of electron transport in rubrene-doped TPD, I-V curves of "electron-only" devices were measured as a function of rubrene concentration.[13] Initially, we observed a decrease of current density with an increase in rubrene concentration at low concentration around 2 mol%, indicating that rubrene acts as an electron trap. At higher concentration more than 3 mol%, we observed an increase of current density with increasing rubrene concentration suggesting that electron mobility is enhanced by rubrene doping.

The increase of electron mobility by rubrene doping was directly confirmed by drift mobility measurements in rubrene-doped TPD films. Table 2 summarizes the drift mobilities of undoped and rubrene-doped TPD films measured by time-of-flight techniques. For undoped TPD, its hole transport property has been extensively investigated and the hole mobility reported is on the order of 10^{-3} cm^2/Vs.[25] The electron mobility of TPD, however, has not been detected by conventional time-of-flight techniques[26]

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TABLE 2 Drift mobilities of undoped and rubrene-doped TPD films.

	Hole mobility (cm^2/Vs)	Electron mobility (cm^2/Vs)
TPD	1.3×10^{-3}	------
Rubrene:TPD*	2.3×10^{-6}	1.8×10^{-7}

*) E= 5.8×10^5 V/cm. Rubrene concentration is 29 mol%.

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due to the too small photocurrent signal. Here, we succeeded to measure the current signal from electron transport in TPD films by doping of rubrene and determined its electron mobility to be on the order of 10^{-7} cm^2/Vs. This is a direct evidence that rubrene enhances the electron mobility in TPD. The increase of electron mobility by rubrene doping is probably due to a new electron hopping path created by rubrene. The increase in electron mobility would extend the penetration depth of electron into the doped TPD layer and result in the wide emission zone observed in electroluminescence.

Electron-hole recombination on the highly fluorescent rubrene, maximized by the dual nature of rubrene acting as an electron hopping site and a hole trap in the host TPD, is proposed to be the dominant emission mechanism, and is largely responsible for the observed enhancement in electroluminescence.

CONCLUSION

In this paper, we have presented two approaches for enhancing EL efficiency based on either energy transfer or direct carrier recombination. For a system based on energy transfer mechanism, we found that the maximum EL efficiency depends more on the nature of the host. A comparison between the absolute PL yield and the external EL efficiency suggests that the creation efficiency of the host exciton by carrier recombination is playing an important rule for determining the maximum EL efficiency. For direct carrier recombination mechanism in rubrene-doped

TPD, we proposed that the dual electronic nature of rubrene is responsible for maximizing the carrier recombination on the guest molecule and leads to enhancement in electroluminescence.

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