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Improvement of carrier transport and luminous efficiency of organic light emitting diodes by introducing a co-deposited active layer

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Abstract. We evaluated carrier transport and luminous efficiency of organic light-emitting diodes (OLEDs) whose active regions consist of a single co-deposited layer. One organic material is a hole transport material N,N'-Bis(3-methylphenyl)-N,N'-diphenylbenzidine (TPD), while the other is an electron transport/emissive material Tris(8-hydroxyquinolinato)-aluminum (Alq3). It was found that the luminous efficiency strongly depends on the thickness and the ratio of the TPD:Alq3 co-deposited layer. This indicates that the carrier balance in the active region can be improved by changing the co-deposited layers. In addition, we performed the dye-doping method to clarify the recombination region. As a result, we found that the radiative recombination is caused in the whole TPD:Alq3 co-deposited layer.

1. Introduction
Recently, organic light-emitting diodes (OLEDs) have attracted much interest due to their application in flat-panel displays and illumination light sources [1,2]. OLEDs generally consist of hetero-structures involving three or more organic layers, which work as electron/hole transport and emissive layers [3], and the characteristics of OLEDs are controllable by changing the layer construction [4]. We have investigated the influence of introducing co-deposited layers in the active region on the carrier transport and luminous efficiency of OLEDs. We found that luminous efficiency of OLEDs can be improved by introducing the co-deposited layer in the active region [5].

In this paper, we investigated the current-voltage (I-V) characteristics and electroluminescence (EL) properties of various samples in which the thicknesses and compound ratios of the co-deposited layers are different. In addition, we performed δ-function-like dye-doping method to clarify the radiative recombination region [6]. This research leads to the answer the relation between the luminous efficiency and the recombination region in OLEDs.

2. Experimental and sample structures
We fabricated OLED samples consisting of a single co-deposited layer consisting of two organic materials; one is a hole transport material N,N'-Bis(3-methylphenyl)-N,N'-diphenylbenzidine (TPD) and the other is an electron transport/emissive material Tris(8-hydroxyquinolinato)-aluminum (Alq3). The indium-tin-oxide (ITO) coated glass substrates were ultrasonically cleaned in acetone, ethanol and isopropyl alcohol for 10 minutes, respectively. The organic materials were vacuum deposited under a
pressure of $1 \times 10^{-4}$ Pa on ITO-coated glass substrates. The organic layers consist of two organic materials; one is TPD as a hole transport material, and the other is as an electron transport/emissive material Alq3. In the co-deposition process, two organic sources were independently controlled. The compound ratio of TPD:Alq3 co-deposited layers were precisely controlled by the deposition rates using a thickness monitor. Rubrene was additionally doped in some samples for the dye-doping method. Finally, the aluminum (Al) layer was vacuum deposited as cathode metal. The active area of sample was $2 \times 2 \text{mm}^2$. We fabricated six types of samples as follows; A) ITO / TPD (50 nm) / Alq3 (50 nm) / Al, B) ITO / TPD:Alq3=x:y (100 nm) / Al, C) ITO / TPD (x nm) / TPD:Alq3=1:1 (100-2x nm) / Alq3 (x nm) / Al, D) ITO / TPD (50 nm) / Alq3+Rubrene [2 %] (50 nm) / Al, E) ITO / TPD (30 nm) / TPD:Alq3=1:1+Rubrene [2 %] (40 nm) / Alq3 (30 nm) / Al, F) ITO / TPD (30 nm) / TPD:Alq3=1:1 (x nm) / TPD:Alq3+Rubrene [2 %] (5 nm) / TPD:Alq3=1:1 (35-x nm) / Alq3 (30 nm) / Al. Note that the thicknesses of the total organic layers are the same as to be 100 nm. Figure 1 shows schematic illustrations of Rubrene-doped three sample structures; samples D, E, and F. Please note that we fabricated three different type of sample F, in which the position of the δ-function-like Rubrene doping is different to clarify the radiative recombination position in TPD:Alq3 co-deposited layer. Each δ-doping layer thickness is 5 nm.

I-V characteristics were measured by using a semiconductor parameter analyzer (4155C, Agilent Technologies). EL spectra were measured by using a CCD spectrometer (USB2000, Ocean Optics). Luminous efficiencies were measured by using an integrating sphere. All measurements were performed in air and at room temperature.

![Figure 1. Schematic illustrations of Rubrene-doped three samples.](image)

3. Results and discussion

3.1 External luminous efficiencies in samples A, B, and C
In the comparison of samples A, B and C, we have found that the brightest EL signal was observed in sample C. Thus, we concluded that the carrier balance was improved in sample C whose active region consists of TPD:Alq3 co-deposited layer sandwiched between TPD and Alq3 layers [5]. In this paper, we evaluated the external luminous efficiencies $\eta_{ex}$ of samples A and C by using an integral sphere. The external luminous efficiency of sample A is about 0.89 %. On the other hand, $\eta_{ex}$ of sample C strongly depends on the layer thickness of TPD:Alq3 co-deposited layer. We found that $\eta_{ex}$ of sample C becomes greater by narrowing the TPD:Alq3 co-deposited layer. The maximum $\eta_{ex}$ of sample C is
2.02% when the thickness of TPD:Alq3 co-deposited layer is 5 nm. However, $\eta_{\text{ex}}$ of sample C becomes smaller than that of sample A, when the thickness of TPD:Alq3 co-deposited layer is wider than 50 nm. These results indicate that the mobility of carriers in the TPD:Alq3 co-deposited layer is much slower than that in TPD layer and Alq3 layer, resulting in the smaller luminous efficiency in wider TPD:Alq3 co-deposited layer.

3.2 Identification of the radiative recombination position in the co-deposited layer

The starting voltage of light emission of sample C is about 12 V, which is independent of the thickness of TPD:Alq3 co-deposited layer. This means that the radiative recombination is most likely caused at the same position in the co-deposited layer. Thus, we have evaluated the position of the radiative recombination by using the dye-doping method [6].

Figure 2 shows normalized EL spectra of samples D, E, and F. Note that EL spectra of samples D and E are almost equal. In addition, EL spectra of samples F [x=17.5] and [x=35] are also equal. The peaking EL wavelengths of Alq3 and Rubrene are 520 and 560 nm, respectively. Thus, the EL spectra of samples D and E originate from only Rubrene. This indicates that the Rubrene was uniformly doped in the Alq3 layers of samples D and E, resulting in the carrier relaxation into the lower bandgap material Rubrene. On the other hand, the three EL spectra of sample F originate from the combination of Alq3 and Rubrene. This means that the contribution of Rubrene to EL emission is not independent of the position of 8-function-like Rubrene doping in the TPD:Alq3 co-deposited layer. This result clearly indicates that the radiative recombination occurs in the whole TPD:Alq3 co-deposited layer.

3.3 Radiative recombination and carrier transport process in sample C

Figure 3 shows schematic energy band diagrams and carrier transport processes at the interface of TPD and Alq3: one has TPD:Alq3 co-deposited layer corresponding to sample C, the other has TPD:Alq3 interface corresponding to sample A. In the case of sample A, the interface of TPD and Alq3 plays a role of the obstruction of hole transport, resulting in the accumulation of holes at the interface. In the TPD and Alq3 heterostructure system, hole transport is more important because the mobility of hole in TPD is much faster than that of electron in Alq3.
In the case of sample C, on the other hand, the TPD:Alq3 co-deposited layer makes the interface unclear, resulting in the improvement of hole transport. Holes can transfer into the whole TPD:Alq3 co-deposited layer then radiatively recombine with electrons. This leads to the lower starting voltage of light emission as well as the higher external luminous efficiency of sample C [5]. This carrier transport model can easily explain the TPD:Alq3 co-deposited layer thickness dependence on the external luminous efficiency of sample C. The narrower thickness of TPD:Alq3 co-deposited layer can increase the carrier concentration, thus the external luminous efficiency is improved.

**4. Conclusion**
We have investigated the radiative mechanism in organic light-emitting diodes consisting a co-deposited layer in the active region by using the dye-doping method. We found that the radiative recombination occurs in the whole co-deposited layer. The external luminous efficiency becomes larger in the sample consisting the narrower co-deposited layer. This is most likely caused by the increased carrier concentration due to the narrower co-deposited layer.

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