Controlled Charge Carrier Transport and Recombination for Efficient Electrophosphorescent OLED

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Abstract

In this paper, the light emitting efficiency, spectrum, and the lifetime of the phosphorescent devices, whose emission characteristics are strongly dominated not only by the energy transfer but also by the charge carrier trapping induced by the emissive dopant, are explained by differences in the energy levels of the host, dopant, and nearby transport layers. On the basis of our finding on device performance and photocurrent measurement data by time-of-flight (TOF), we investigated the effect of the difference of carrier trapping dopant and properties of the host materials on the efficiency roll-off of phosphorescent organic light emitting diode (OLED), along with a physical interpretation and practical design scheme, such as a multiple host system, for improving the efficiency and lifetime of devices.

1. Introduction

Phosphorescent OLEDs show much higher efficiency than fluorescent OLEDs since both singlet and triplet exciton can be utilized for the production of light [1]. However, relatively higher doping concentration is required for an efficient Dexter energy transfer from host material to phosphorescent dopant, which simultaneously induces rapid drop of efficiency at high current density and dopant concentration due to its long phosphorescent lifetime [2]. It should be also noted that the electrical properties of phosphorescent emission layer would be highly affected by the properties of dopant, which makes the optimum control of dopant concentration a decisive factor for the practical application of devices.

Due to the long diffusion length of the triplet exciton [2.3], an improvement of the efficiency and stability of the phosphorescent OLEDs often relies on the proper selection of blocking layers by effectively confining the triplet exciton in emissive layer and achieve the balance of charge during the device operation. The characteristics of an archetype phosphorescent green dopant *fac*-tris(2-phenylpyridine) iridium [$Ir(ppy)_3$] doped into CBP host have been widely studied and it is known that $Ir(ppy)_3$ work as a hole trap and most exciton is directly formed at dopant site, hence nearly no affect in electron conduction [3-5]. Previously, we have demonstrated that device efficiencies are significantly dependent upon the energy level difference between host/dopant and the position of the dopant-rich region [6]. Consequently, a biased (non-centered) recombination zone in the EML, induced by difference of charge trap, played a major role in the light emitting behavior and stability of both fluorescent [7] and phosphorescent devices [8]. Operational stability of OLED seems to be basically proportional to the efficiency if the same materials are used and the efficiency is highly affected by the charge balance. Therefore, the further information on the underlying phenomena such as the charge carrier injection and transport of the phosphorescent emissive layer will give more clear background to optimize the device structure.

In this work, based on the distinct charge carrier transport properties of red and green dopants placed in different host materials, estimation of the effective emission zone was provided. Not only changing the interfacial blocking layer adjacent to the light emission layer, engineering methods such as layered or blended heterojunction for host are introduced and analyzed in terms of the trapping behaviors and resultant device performance (efficiency, spectral characteristics, and lifetime).

2. Experimental

The patterned ITO (indium tin oxide) substrates

with a sheet resistance of 15Ω/□ were cleaned by ultra-sonication in deionized water and isopropanol. They were rinsed again in a chamber filled by hot isopropanol vapor. The cleaned substrates were subjected to UV-O_3 treatment for 15 minutes. Onto this cleaned substrate, Organic layer of N,N'-diphenyl-N,N'-bis (1-naphthyl)-(1,1'-biphenyl)-4,4'-diamine (NPB, 50nm) as HTL was vacuum-deposited. Small molecular phosphorescent OLEDs were fabricated using different host materials. The typical combination was 4,4'-N,N'-dicarbazole-biphenyl (CBP), 4,4',4"-tris(N-carbazolyl) triphenylamine (TCTA) or 1,3,5-tris(N-phenyl benzimidizol-2yl) benzene (TPBI), which were co-deposited with $Ir(ppy)_3$ or bis(2-(2´-benzo[4,5-a]thienyl)pyridinato-N,C3´)iridium(acetyl-acetonate) ($Btp_2Ir(acac)$) as green or red dopants, respectively (Fig. 1). When the hole blocking layer was introduced, Aluminum(III)bis(2-methyl-8-quinolinato)4-phenyl phenolate (BAlq) or other materials were employed. As an electron transport layer (ETL), 25nm-thick tris(8-hydroxyquinoline)-aluminum (Alq_3) was evaporated, followed by LiF/Al cathode. Fig. 1 illustrates the chemical structures and bandgap of materials used. The variation of the blocking layer thickness/type or configuration of host (single, layered heterojunction, and blended heterojunction, *etc., see Fig.2*) could explain the relationship between charge/exciton confinement, major emitting region, and device performance of phosphorescent light emitting system.

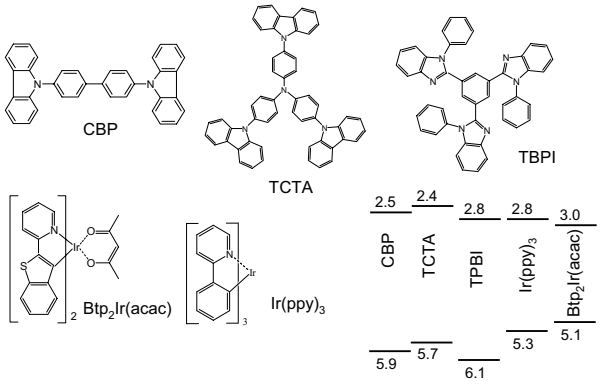

Fig. 1 Chemical structures and energy levels of organic materials for several light emitting hosts and dopants used in this study

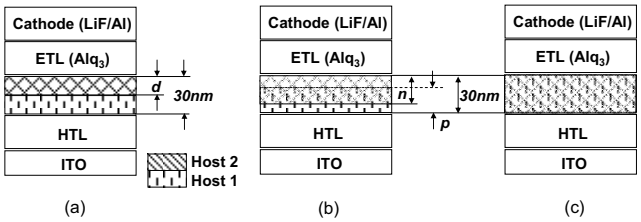

Fig. 2 Device structures of the phosphorescent devices with engineered heterojunction light emitting layer (a) layered (b) layered/blended (c) blended heterojunction

3. Results and discussion

The selective combination of host materials in the light emitting layer were conducted based on the relative values of energy level (highest occupied molecular orbital; HOMO and lowest unoccupied molecular orbital; LUMO) and charge carrier mobility. Room temperature TOF measurement showed that CBP showed rather high hole mobility $1.1\times10^{-3}cm^2/Vs$ ~$3.6\times10^{-3}cm^2/Vs$, whereas its electron mobility is almost one magnitude lower, which illustrates that CBP is a hole-transporting type host [9]. TCTA facilitates the hole injection and electron blocking, since HOMO and LUMO levels are shifted upward compared to those of CBP [10]. Electron transport and hole blocking properties of TPBI and other host materials are conspicuous. Fig. 3 illustrates brightness-voltage and efficiency-brightness curves of devices comprising CBP (host1), TPBI (host2) TCTA (host3), and their blended heterojunction structure (Fig. 2(c)).

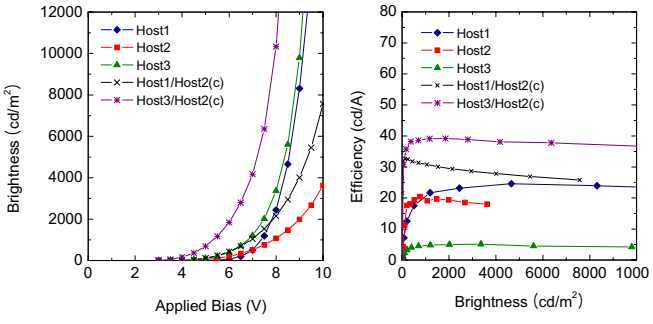

Fig. 2 Brightness-voltage-current efficiency curve of phosphorescent devices with single and layered heterojunction light emitting layers. Device structures were ITO/NPB (40nm)/ Host(s):$Ir(ppy)_3$ 7%, 30nm/ BAlq (5nm)/ Alq_3 (25nm)/ LiF/Al.

The highest brightness was obtained at host3/host2 layered structure due to the efficient hole injection through TCTA and electron injection from TPBI. The

HOMO level of TCTA (5.7eV) was beneficial, providing less hole-trap compared to CBP at the direct hole injection process from NPB to $Ir(ppy)_3$ HOMO level. When TPBI single host was employed, large carrier trap due to the offset with $Ir(ppy)_3$ HOMO (5.3eV) can explained the reduced brightness and efficiency. However, layered structure with TPBI on top of CBP or TCTA host yields effective electron injection (LUMO level of TPBI is 2.8 eV) and enhancement of brightness and current efficiency was shown.

Fig. 4 shows the charge injection and brightness-efficiency behavior of green phosphorescent devices with layered/blended heterojunction light emitting layer structures (see Fig 2 (a) and (c)). In case of charge injection at low bias, combination with TCTA(host2) both for heterojunction (a) and (c) represents lower bias voltage behavior due to the smaller HOMO level offset between hole transport layer and host. However, peak current efficiency and luminance at high-voltage are not dependent upon the type of hole-transport hosts. The efficiency value as high as 60cd/A (47lm/W) at 4V, 860cd/m^2 was obtained in case of host2/host4(electron-transporting) with layered junction as seen in Fig 2(a), while other configuration yields comparable high efficiency and low driving voltage characteristics. Further studies on the intermediate heterojunction (Fig 2(b)) for determining the recombination region as well as reduced charge trapping behavior is under progress.

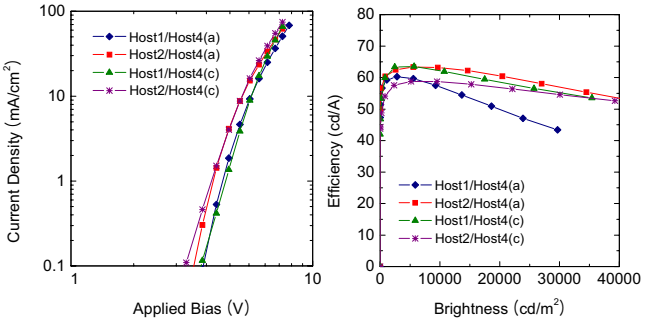

Fig. 3 Current density vs. bias voltage and current efficiency vs. luminance curves of phosphorescent devices with layered/blended heterojunction light emitting layers. Device structures were ITO/NPB (40nm)/ Hosts:$Ir(ppy)_3$ 7%, 30nm/ BAlq (5nm)/ Alq_3 (25nm)/ LiF/Al.

4. Summary

We have investigated the charge carrier trapping and conduction of phosphorescent light emitter composed of multiple host and dopant system. Characterization of the charge carrier mobility of the emitting layer(s) and interfacial layer material was performed. Such measurement technique of the carrier mobility is the useful characterization method for the understanding of carrier transport behavior and expectation of the recombination zone for specific hole- and electron-transporting type host materials in phosphorescent OLED. Moreover, long lifetime and reduced roll-off efficiency at high-current might be achieved by the concurrent device design (equal distribution of charge carrier trap for better balance) and the characterization of device properties clarifying their interplay.

Acknowledgement

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5. References

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