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A Theoretical Approach to the Overshoot Effect in Transient Electroluminescence of Phosphorescent OLEDs with Mixed Host System

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Abstract: *We propose a theoretical approach that overshoot phenomena can be theoretically predicted for an electro phosphorescent OLEDs. The transient EL responses of phosphorescent OLEDs adopting the mixed-host emissive layer were studied in order to investigate the carrier recombination mechanism. It was observed that a prominent overshoot peak in the transient EL profiles, depending on the composition ratio of the HTM and the ETM, when the applied bias voltage decreased abruptly to zero. A good agreement is found between the theoretical and experimental results.*

1. Introduction

During the last decade, organic light emitting diodes (OLEDs) have attracted the attention of a large number of scientists all over the world, and consequently great advances have been made regarding the materials, device fabrication and device performance [1–3]. The transient electroluminescence (EL) measurement has been widely used to investigate the luminescence mechanism of OLEDs, since it enables us to analyze the carrier transport process and recombination process separately in the whole luminescence process [4], [5]. The transient EL has been used as a tool to study device properties [6–12], both under low current cw conditions [6–9] and under high excitation density [10, 11]. The use of transient EL has been made to obtain a dispersion parameter related to structural disorder [12] as well as for getting information on charge carrier dynamics [11] and trapping [12]. In this letter, we have studied transient EL responses of phosphorescent OLEDs adopting the mixed-host emissive layer in order to investigate the carrier recombination mechanism. The use of mixed host system composed of electron transporting materials (ETMs) and hole transporting materials (HTMs) is a powerful approach to get balanced carrier distribution in the emitting layer (EML) [13-14]. In the mixed-host-based OLEDs, we observed a prominent overshoot peak in the transient EL profiles, depending on the composition ratio of the HTM and the ETM, when the applied bias voltage decreased abruptly to zero. We propose a theoretical approach that overshoot phenomena can be theoretically predicted for an electro phosphorescent OLEDs

2. Theoretical Approach to the Overshoot Effect

Previously Chandra et al has reported that in single layer organic light emitting diodes the EL transient after the turn off of the external bias can be divided into the following three regions: (i) fast decay region, (ii) delayed peak region, and (iii) slow decay region[15]. The initial fast decay is attributed to the recombination of injected charge carriers; the rise and exponential decay just after the peak in the delayed EL peak are due to the recombination luminescence owing to the movement of detrapped holes with non-correlated electrons; and the slowly decaying EL following power law t^{-z} (z lying between 1.5 and 2) is due to geminate recombination of the correlated electron–hole pairs [15]. Same approach can be extended for predicting the transient EL responses of phosphorescent OLEDs adopting the mixed-host emissive layer.

The total EL intensity, that is, the area below EL intensity versus time curve for the EL overshoot may be expressed as

$$I_T = \frac{\beta_1 \eta_e r_{st} \eta_{PL} g_0}{\beta_0 \alpha} \tag{1}$$

and the peak of EL intensity can be expressed as

$$I_m = \frac{\beta_1 \eta_e r_{st} \eta_{PL} g_0 (\xi - \alpha)}{\beta_0 \alpha} \left(\frac{\alpha}{\xi}\right)^{1/(1-(\alpha/\xi))} \tag{2}$$

where β_1 is the rate constant for the recombination of released holes with the electrons in the emissive layer, ξ is the rate-constant for the decrease of voltage with time, $\alpha = 1/\tau_r$, τ_r is the release time of detrapped holes from the interface, $\beta_0 = (\beta_1 + \beta_2)$, β_2 is the rate constant for the formation of correlated electron–hole pairs in the emissive layer, η_e is the exciton formation efficiency of the charge carriers, r_{st} is the singlet-triplet ratio, and η_{PL} is the quantum efficiency of the photoluminescence of emissive layer, t_0 is the time taken by the holes to cross the quenching region of length, $N_0 = BV_b$, B is the proportionality constant , V_b is barrier potential and

$$g_0 = \frac{\alpha N_0 \xi exp\,(-\alpha t_0)}{\xi - \alpha} \tag{3}$$

3. EXPERIMENTAL SUPPORT TO THE PROPOSED THEORY

The devices structure of OLED is of indium tin oxide (ITO)/hole transporting layer (HTL, 140 nm)/emitting layer (EML, 106 nm)/electron transporting layer (ETL, 58 nm)/LiF (0.8 nm) /Al (150 nm). The HTL, poly (3, 4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT: PSS) (CH8000, Baytron Co.) was spin coated on the ITO surface. An emissive layer comprising a red phosphorescent emitter, tris-(1-phenylisoquinolinato) iridium (III) (Ir(piq)3) (5 wt. %) and a combined host system of 1,3,5-tris-(N –phenylbenzimidazol-2, yl) benzene (TPBI) and N, N' -bis(1-naphtalenyl)-N-N'-bis (phenyl benzidine) (NPB) dissolved in dichloromethane was spin coated on the HTL. The mixing ratio of TPBI and NPB was varied as 10:0, 8:2, 7:3, 5:5, 3:7 and 2:8. The TPBI (ETL), LiF and Al were thermally evaporated on the EML consecutively.

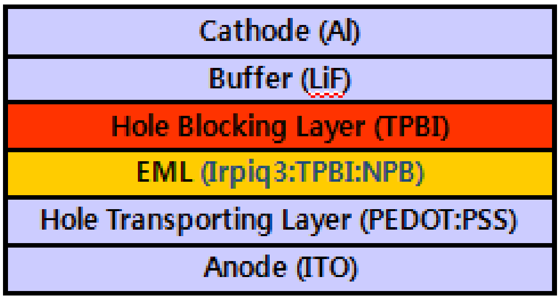

Figure1. *Device structure of OLEDs with mixed host*

Fig.2 shows luminous efficiency variation measured from the devices with different host compositions. The device with TPBI only host shows high turn-on voltage and low

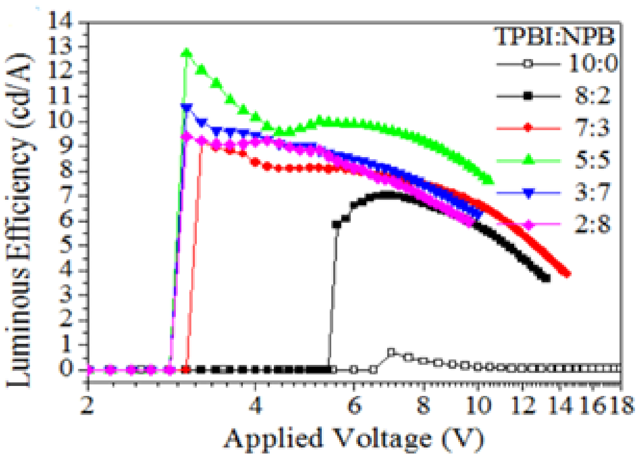

Figure2. *Luminous Efficiency variations depending on host compositions*

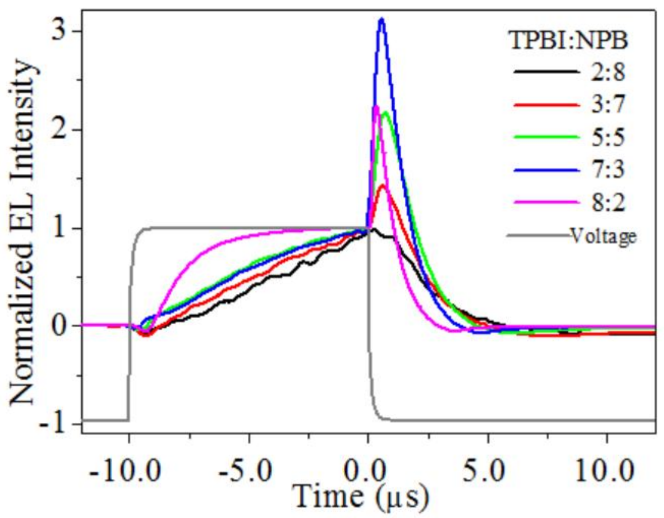

Figure3. *Overshoot Peaks in Transient EL profiles*

EL emission due to poor injection of holes and charge unbalance. As the concentration of NPB increases in the TPBI: NPB mixed host, the turn-on voltage decreases and the luminescence efficiency increases due to better hole injection and improved charge balance technique.

Fig. 3 shows the transient EL responses, normalized at the time when the applied voltage decreases abruptly from the forward bias (+10 V) to the reverse bias (-10 V) for the devices with various TPBI: NPB composition ratios. The transient EL responses exhibit a large overshoot peak followed by the residual EL intensity which lasts for about 10s. Fig. 3 shows that the EL overshoot increases as the NPB concentration increases up to 7:3 TPBI: NPB ratio.

Under this bipolar voltage pulses, the EL overshoot intensity increases rapidly with increasing NPB concentration and shows a maximum (~3.1 times higher than the steady-state EL intensity) at the 7:3 TPBi:

NPB ratio. Moreover, as the NPB concentration increases, the EL overshoot decay time decreases significantly. Even though our devices are designed to facilitate charge injection there exists energy difference between the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels of EML and HTL or ETL. The overshoot effects in our devices can be attributed to the enhanced recombination of electrons and holes in the EML and back drifted charges once accumulated at interfaces

4. CONCLUSIONS

A phenomenological theory is explored for the EL overshoot produced during the turn off of multilayer OLEDs. The EL transient after the turn off of the external bias can be divided into the following three regions: (i) fast decay region, (ii) delayed peak region, and (iii) slow decay region The initial fast decay following power law is attributed to the recombination of injected charge carriers; the rise and exponential decay just after the peak in the delayed EL are due to the recombination luminescence owing to the movement of detrapped holes with no correlated electrons; and the slowly decaying EL following power law t^{-z} (z lying between 1.5 and 2) is due to geminate recombination of the correlated electron–hole pairs. The release time r of holes trapped at TPBi: NPB/ polymer interface, the activation energy E_a for the detrapping of holes from the interface, and time-constant for the electro phosphorescent OLED circuit, can be determined from the measurement of the time dependence of the EL overshoot

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