The transfer of emission site in polymer LEDs with increasing voltages

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1. Introduction

Recently, polymer light-emitting diodes (PLEDs) have attracted much attention for the application of flexible display etc. They now have sufficient brightness and range of colors. However, the emission efficiency and lifetime is still not high and long in comparison with the low molecular based-organic electroluminescent devices, respectively. To improve the emission efficiency and lifetime, it is one of the most important information to elucidate the emission site (spatial distribution of emission center) within an active luminescent polymer layer. Several techniques, such as local-doping [1] and optical interference analysis [2], have been employed to study the emission site in organic LEDs. As PLEDs are generally fabricated using spin coating procedures, it may be difficult to use the local-doping method. While, in case of optical interference analysis, one can conveniently utilize not special but conventional PLED structure.

In this paper, we adopted classical optical intereference method and the applied-voltage dependence of emission site in two PLEDs, which have different thickness active luminescent polymer layers, was analyzed. Here, when comparing results between different voltages, one should note that this analysis cannot be applied in case that the change of emitting species, such as the formation of the new excited state by chemical reaction, excimer and exc iplex, is accompanied by the increase of applied-voltage.

2. Experimental and Analysis

We fabricated two PLEDs which have 275nm and 85nm thickness active luminescent polymer layers. Fig.1 shows device structure of these PLEDs. The poly(3,4-ethylenedioxythiophene): poly(styrene sulphonic acid) (PEDOT:PSS) was spin coated onto an indium-tin-oxide (ITO) glass substrate. Next, the active luminescent polymer film with the thickness of 275nm or 85nm was spin coated onto PEDOT:PSS layer and the metal was thereafter thermally evaporated under vacuum. Finally, PLEDs were encapsulated using an epoxy resin.

Electroluminescence (EL) was measured for two PLEDs as a function of angle. The sample was fixed in a vertical plane, centred on the axis of a rotation stage and was driven by pulse voltage (pulse width: 100µsec). EL spectra at different applied-voltages were measured at a series of outer angles θ_{out} ($\theta_{out}=0^{\circ}$ corresponds to normal incidence on the device surface) through a polarizing filter (to separately detect p- and s-polarization) and an optical fiber connected to a polychromator-ICCD multichannel spectrophotometer system (Princeton Instrument Inc.). All experiments were performed at room temperature and in air.

The EL emission from a PLEDs was simulated using classical theory of electromagnetic radiation (optical interference model) [3]. This simulation was done under the following assumptions, 1) Lorentzian function is used for the profile of the emission site, 2) all materials are isotropic and not required to be treated as birefringent materials, 3) the emission-transition dipoles orient isotropically. As the optical constants (*n*: refractive index, k: extinction coefficient) of each material were required on this simulation, we determined them by spectroscopic ellipsometry (Jobin Yvon Ltd.) and Kramers-Kronig analysis [4]. From the comparison between the experimental and simulated EL spectra, the parameters of emission site (d: distance from metal electrode, w: full width of half maximum of emission site) were determined, see fig.1.



Fig.1 Device structure of polymer LED and the spatial distribution function for emission site used in calculation.

3. Results and Discussion

Fig.2(A) shows the applied-voltage dependence (6V to 40V) of EL spectra measured at $\theta_{out}=0^{\circ}$ in a PLED with 275nm thickness polymer layer. Each spectrum was normalized at emission peak-wavelength. With increasing voltages, we found that the half width of EL spectra gradually decreased. Fig.2(B) shows the results of EL spectra simulated using parameters of w=10nm and d=150, 160, 170nm. A comparison between the experimental and simulated EL spectra was in good agreement. Now, we summarize these results to Fig.3, which indicates the schematic explanation for the transfer of emission site with increasing voltages. At low drive-voltage, the emission site is located at about 170nm from metal cathode. On the other hand, the



Fig.2 EL spectra ($\theta_{out}=0^\circ$: PLED with 275nm polymer layer) measured at different voltages (A) and simulated by optical interference model (B).



Fig.3 Transfer of emission site with increasing voltages in PLED with 275nm polymer layer.

emission site moves toward cathode with increasing voltages and arrives at about 150nm from cathode at 40V. We succeeded to visualize the transfer of emission site with the change of applied-voltage in PLEDs.

Next, the voltage dependence of EL spectra in a PLED with 85nm thickness polymer layer were observed. We found that these spectra didn't almost change with increasing voltages. Now, we shouldn't give an immediate answer that emission site doesn't move with increasing voltages. Fig.4 shows each polarization component (p and s) of the emission patterns (the spatial distribution of integrated intensity of EL spectra) in a PLED with 85nm polymer layer driven at 8V and 24V. These data were normalized by the intensity at $\theta_{out}=0^{\circ}$. We found that the emission patterns have a tendency to expand with increasing voltages, especially s-polarization component. We evaluated these results by optical interference model and reached a conclusion that the emission site moves toward ITO anode, which is clear different from the tendency in a PLED with 275nm polymer layer. This must be attributable to the change of charge-carrier valance, which may be caused by the difference of stack-structure of polymer chains, the surface po-



Fig.4 Emission patterns (p - & s-polarization) at 8V and 24V in PLED with 85nm polymer layer.

potential of polymer /metal interface and so on. In order to elucidate these mechanisms, it is important to investigate the polymer-metal surface structure, the electric properties of PLEDs, etc. in detail.

4. Conclusions

We demonstrated a novel analysis method to visualize the change of emission site. From analysis using a PLED with 275nm thickness active polymer layer, we found that the emission site transferred about 20nm toward metal cathode with increasing voltages. On the other hand, in a PLED with 85nm polymer layer, the emission site moved toward ITO anode. This difference must be attributable to the change of charge-carrier valance (carrier-injection process from cathode to polymer layer and carrier-transport process within polymer layer). By elucidating these mechanisms, it may be possible to improve emission efficiency and lifetime.

References

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