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Fast Transient Response of Multilayer Polymer Light-Emitting Devices Using Fluorene Derivatives for the Electro-Optical Conversion Devices

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This study focused on the performance of solution-processed multilayer polymer light-emitting diodes (PLEDs) with both the hole and electron transport layers using poly(9,9-dioctylfluorene-co-benzothiadiazole) (F8BT) as an active layer. By insertion of both the hole and electron transport layers, improved emission and transient characteristics were achieved for the electro-optical conversion devices. For the multilayer device, the maximum current efficiency of 2.2 cd/A and the short response times of electroluminescence (ca. 30 ns) were achieved.

Keywords Polymer light-emitting diode; transient characteristics; multilayer device; fluorene

1. Introduction

Organic devices utilizing conjugated polymers have attracted considerable interest because of their advantages in large-area device fabrication by solution-processes which bring the benefits of cost-effective mass production. Fluorene-type polymers have emerged as an important class of conducting polymers, due to their efficient emission, high stability, and relatively high mobility [1,2]. The polymer light-emitting diodes (PLEDs) can be expected to be applied to electro-optical conversion devices for generating high-speed optical pulses. PLEDs based on fluorine-type polymers, which exhibit short fluorescence lifetime, have the potential for the electro-optical conversion devices such as the optical link and sensor devices. However, solution-processed single-layer PLED is difficult to achieve good charge balance.

In this study, we demonstrated improved emission and transient characteristics of solution-processed multilayer PLEDs with hole and electron transport layers utilizing poly(9,9-dioctylfluorene-co-benzothiadiazole) (F8BT) as an emissive layer.

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2. Experimental

The glass substrate was degreased with solvents and cleaned in a UV ozone chamber. First, a 45-nm-thick poly(ethylenedioxythiophene):poly(styrene sulfonic acid) (PEDOT:PSS) hole-injection layer was spin-coated on an indium tin oxide (ITO)-coated glass substrate and baked. Next, poly(9,9-dioctylfluorene-co-N-(4-butylphenyl)-diphenyl amine) (TFB) as a hole transport layer (HTL) and F8BT as an emissive layer were fabricated by the spin-coating method [3]. Using anhydrous n-butyl acetate as a solvent, tris[3-(3-pyridyl)mesity]borane (3TPYMB) as an electron transport layer was formed on a F8BT layer without intermixing. The typical thicknesses of TFB, F8BT and 3TPYMB layers were 10, 50 and 15 nm, respectively. The cathode consisting of CsF(3nm)/Mg:Ag/Ag was deposited in vacuum at a chamber base pressure of about 10^{-4} Pa. The active areas were 4 or 0.3 mm^2.

The current density-voltage-luminance (J-V-L) characteristics were obtained using a digital multimeter (Keithley 2000), a regulated DC power supply (Kenwood PW36-1.5AD), and a luminance meter (Minolta LS-100). Transient electroluminescence (EL) was measured by applying square-wave voltage pulses generated by an HP8114A source (Agilent). The optical pulse was observed using a photomultiplier tube detector (Hamamatsu Photonics). The obtained EL response and voltage were simultaneously digitized by LeCroy 104MXi oscilloscope.

3. Results and Discussion

Figure 1 shows the current density dependence of EL spectra of the three types of PLEDs with and without hole and electron transport layers. The spectra of the device with the 3TPYMB layer were independent on the current density. By contrast, the peak wavelength in the spectra of the devices without the 3TPYMB layer was blue-shifted from 565 nm to 545 nm with increasing the current density. At high current density, the spectra of the devices without the 3TPYMB layer were almost the same as that with 3TPYMB layer. A yellow-green emitting multilayer PLED based on F8BT was used as the light source for plastic optical fiber, which has the advantages of low propagation loss.

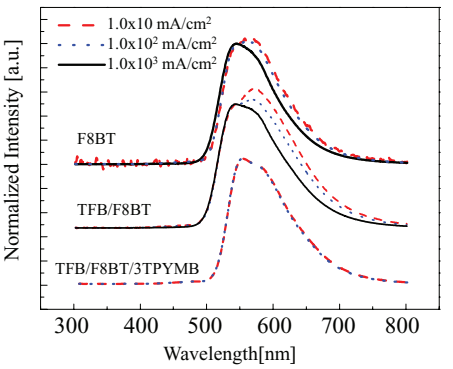

Figure 1. Current density dependence of EL spectra of the OLEDs of ITO/PEDOT:PSS (45 nm)/F8BT(50 nm)/CsF/Mg:Ag/Ag, ITO/PEDOT:PSS(45 nm)/TFB(10 nm)/F8BT (50 nm)/CsF/Mg:Ag/Ag and ITO/PEDOT:PSS(45 nm)/TFB(10 nm)/F8BT(50 nm)/ 3TPYMB(15 nm)/CsF/Mg:Ag/Ag.

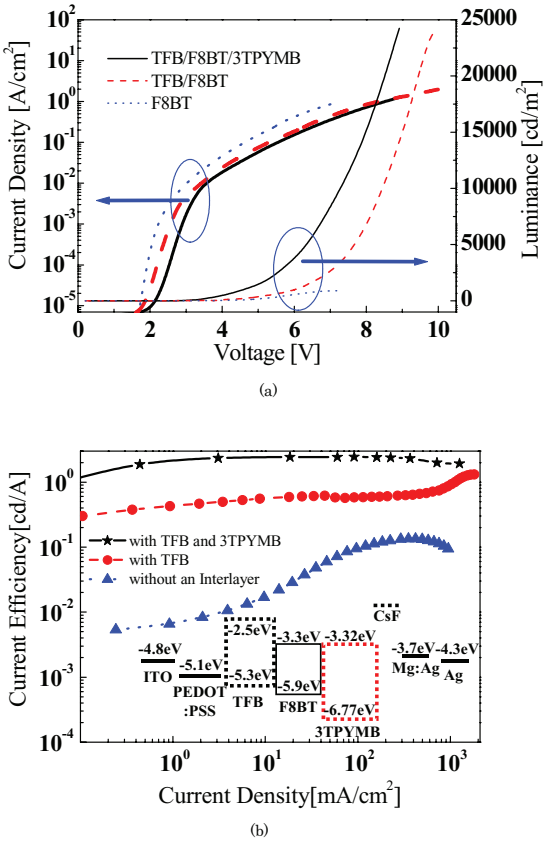

Figure 2. (a) J-V-L characteristics and (b) current efficiencies of the F8BT, TFB/F8BT and TFB/F8BT/3TPYMB devices. Inset of Fig. 2(b): the energy diagrams of materials used in this study.

Figure 2(a) and (b) indicate the J-V-L characteristics and current efficiencies of three types of multilayer PLEDs, respectively. The device without the TFB interlayer shows the maximum luminance of 900 cd/m^2 and the maximum current efficiency of 0.14 cd/A. By insertion of the TFB layer which acts as the hole transport and electron blocking layer as shown the inset of Fig. 2(a), the device performance was improved. Furthermore, for the TFB/F8BT/3TPYMB device, the emission intensity reached to 24,000 cd/m^2 at 9 V, and the maximum current efficiency of 2.2 cd/A was achieved. One of reasons of the improvement is that the 3TPYMB layer whose LUMO level is around 6.77eV acted as the hole block layer [4]. In addition, it is reported that an electron injection at F8BT/Cs (CsF) is not necessarily smooth because the performance of F8BT device is markedly dependent on the F8BT/Cs(CsF) interface due to the change of electron density state in F8BT at F8BT/Cs(CsF) [5]. It is considered that the current efficiencies of the devices without the 3TPYMB layer were lower than those with the 3TPYMB layer because of quenching of excitons by emitting at the side close to the F8BT/CsF interface and the 3TPYMB layer leads to the improvement of the electron injection at the CsF/F8BT interface.

It is important to focus on the transient properties and applications of PLEDs for transmitting high modulation signals in the field of optical link and optical sensor devices.

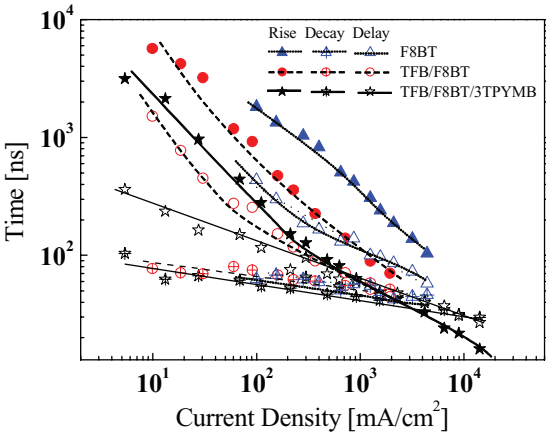

Figure 3. Current density dependence of the rise, decay and delay times of the F8BT, TFB/F8BT and TFB/F8BT/3TPYMB devices with the active areas of 0.3 mm^2.

So, to investigate the transient properties of this PLED, the devices were driven at a 1 ms period and duty ratio of 1/100 pulse voltage, i.e. pulse width was 10 μs. Figure 3 shows the current density dependence of the rise, decay and delay times of three types of PLEDs. The rise (decay) time is defined as the time required changing the optical response from 10% (90%) to 90% (10%) of its total intensity change. It was found that the minimum rise, decay, and delay times for the TFB/F8BT/3TPYMB device (15 ns, 30 ns and 27 ns) were shorter than those for other devices. The rise time of the TFB/F8BT/3TPYMB device was shorter than other devices owing to improvements of the carrier injection and the recombination rate of excitons at both the PEDOT:PSS/F8BT and F8BT/CsF interfaces. By contrast, the decay times of all the devices were almost the same because the time constant of resistance and capacitance and fluorescence lifetime of F8BT were mainly contributing factors [6]. Most importantly, the delay time of the device with the 3TPYMB layer at lower current density was improved markedly. The delay time that is the time delay between the onset of the applied voltage pulse and the initialization of the EL signal is related to the transient time of the charge carriers. The hole mobility is lower than the electron mobility in a F8BT film [7] and as mentioned above, for the devices without the 3TPYMB layer, the recombination zone of excitons is close to the F8BT/CsF interface. Therefore, at the low current density, the longer delay time of the devices without the 3TPYMB layer is attributed to the transient time mainly determined from slower hole carriers in the F8BT layer. At high current density, where the injection of the electron was smooth due to high applied voltage, the recombination zone was shifted from the F8BT/CsF interface to the TFB/F8BT interface, and then the delay time of the devices without the 3TPYMB layer became almost the same as that with the 3TPYMB layer. Thereby, as previously indicated, the EL spectra of the devices without the 3TPYMB layer were shifted with increasing current density and the current efficiency increased at high current density.

4. Conclusion

For the solution-processed multilayer PLEDs utilizing F8BT, the effects of the insertion of the hole and electron transport layers were investigated. We demonstrated that high-efficiency and high-speed in solution-processed PLEDs by constructing the multilayer

structure were achieved. For the TFB/F8BT/3TPYMB device, the maximum current efficiencies of 2.2 cd/A and the short response times of electroluminescence (ca. 30 ns) were achieved. It is considered that by the TFB layer, which acts mainly as an electron blocking layer, the recombination of excitons was enhanced and the 3TPYMB layer led to the improvement of the electron injection at the CsF/F8BT interface. We believe that this approach is applied as one of the methods to simply manufacture the optical link devices.

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