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Organic Electroluminescence of Styryl Type Dyes on Vacuum Deposited Thin Films

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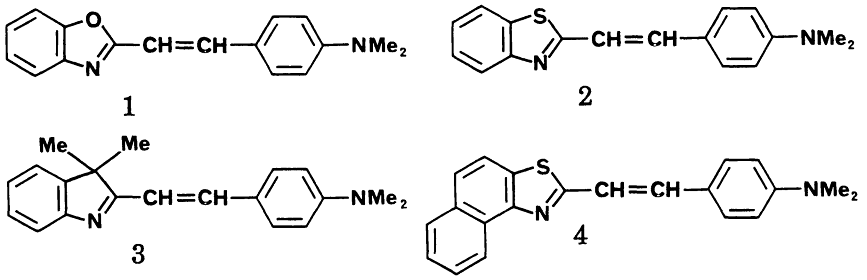
Some styryl type dyes, such as 2-(4-Dimethylaminostyryl) benzoxazole (**1**), were found to be useful as novel emitting materials for the electroluminescent (EL) device. The EL devices with hole transporting layer have more efficiency than that with only emitting layer, but the electron transporting layer do not work effectively. The EL intensity of the H-type device with **1** was 3.8 cd/m^2 at the dc bias voltage of 60 volt.

Recently demands of flat type thin displays with large-area increase with the development of computer systems and visual apparatus. Electroluminescence (EL) in organic crystals has been known,1) but it was not practically useful for EL devices. In 1987 Tang and VanSlyke 2,3) succeeded in the fabrication of the excellent organic EL device with a two-layer structure having hole transporting layer and emitting layer. Their device is composed of MgAg-electrode / emitting layer of aluminum chelate complex / hole transporting layer of diamine, which is constructed on a indium-tin-oxide (ITO) coated glass substrate. Bright emission is achieved with the low drive voltage of several volt. Organic EL devices have been recently investigated to realize the systems with full color, low cost and low environmental pollution.

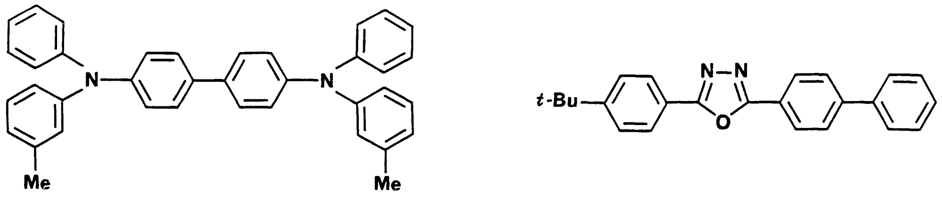
We intend to search novel emitting materials in the fluorescent dyes, and to make efficient EL devices with the charge transporting layer. Four styryl type dyes **1** - **4** shown in Scheme 1 were tested as emitting materials which have intense photoluminescence. Three types of device configurations shown in Fig.1 were evaluated to correlate the EL characteristics with device structures. The N-type device has only emitting layer, but the H-type or E-type devices have hole or electron transporting layer with the emitting layer, respectively. 4,4'-bis[N-phenyl-N-(3-methylphenyl)amino] biphenyl (TPD) was used for hole transporting layer and 2-(4-tert-

Butylphenyl)-5-(p-biphenylyl)-1,3,4-oxadiazole (Butyl-PBD) for electron transporting layer (Scheme 2). These two compounds are well known as the charge transporting materials reported by Adachi and coworkers.[4-6]

The EL devices were fabricated by conventional vacuum deposition method. The organic materials were deposited from tungsten-boat onto ITO-coated glass substrate at 10^{-5} - 10^{-6} Torr under room temperature. And then magnesium was deposited from tungsten-basket onto the dye deposited layer. The deposition rate of organic materials is usually 50 - 200 Å/s. The thicknesses of the emitting layer, charge transporting layer and cathode are 10000, 300, and 1000 Å, respectively. The emitting area of the device is a rectangle of 0.7 X 0.7 cm^2 , and the luminescence is observed through the ITO-coated glass substrate. The intensity of EL was measured with a MINOLTA luminance meter LS-100 at about 10^{-5} Torr.



Scheme 1.



TPD(Hole Transporting Material) Butyl-PBD(Electron Transporting Material)

Scheme 2.

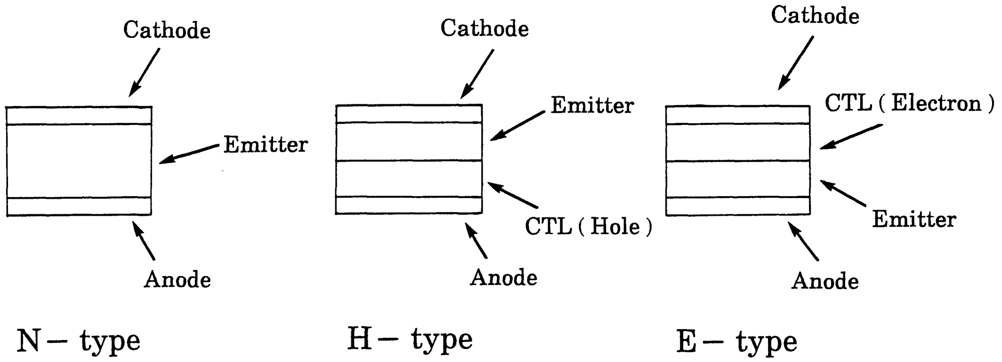


Fig.1. Typical EL device structure.

Since dye **1** gave the strongest luminescence among the four **1** - **4** tested, it was examined in detail. The N-type device with **1** was firstly made in compositions of ITO / **1** / Mg. The curves (A) in the Figs.2 and 3 showed the bias voltage dependence of the current and EL intensity of the N-type device with **1**, respectively. The current and EL intensity are approximately proportional to the bias voltage, and the EL intensity of 0.1 cd/m^2 is achieved at dc 60 V. The H-type and E-type devices with **1**, having charge transporting layers, were also made in compositions of ITO / TPD / **1** / Mg, and ITO / **1** / Butyl-PBD / Mg, respectively. The EL properties of the H-type device, presented by the curves (B) in the Figs.2 and 3, showed the intenser EL at lower current as compared with those of the N-type device. The maximum EL intensity of 3.8 cd/m^2 was achieved at dc 60 volt by the H-type device and was clearly observed under room light. The H-type device attained intenser luminescence with the lower electric power by addition of the hole transporting layer. The EL properties of the E-type device are shown by the curves (C) in Figs.2 and 3. Comparing the curve (A) (N-type) with the curve (C) (E-type) in Fig.3, intenser EL in (C) is attained in the range from dc 45 to 60 V, while the curves in Fig.2 show that the E-type device (C) needs slightly higher electric power than the N-type device (A). Therefore it is hardly said that the E-type device is more efficient than the N-type device.

The performance of the thin film formation is depended on the deposition rate of dye **1**. If the deposition rate is too fast to fabricate, the films become a little nebulous. Lowering the deposition rate to 2 - 4 Å/s, the performance of the thin films is further improved. The EL properties can be improved with reducing the thickness of films.

The EL properties of dyes **2**, **3**, and **4** were also examined with the N-type devices. The EL device with the dye **2** gleamed very weakly but the luminescence could not be measured, and the device with the dye **4** showed only a luminous dot on the surface of the device. The dye **3** gave no luminescence in the EL device. Consequently, these three dyes are not suitable for the emitting material of the EL device.

In conclusion, a novel emitting material of dye **1** for the H-type EL device was found. The work with the other styryl type dyes is currently under investigation.

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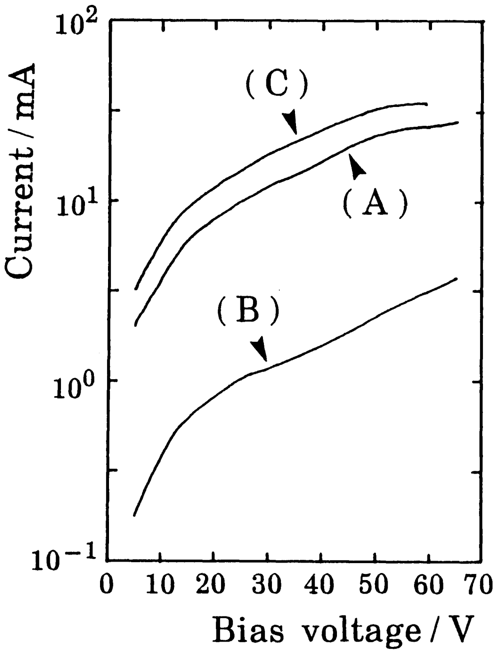

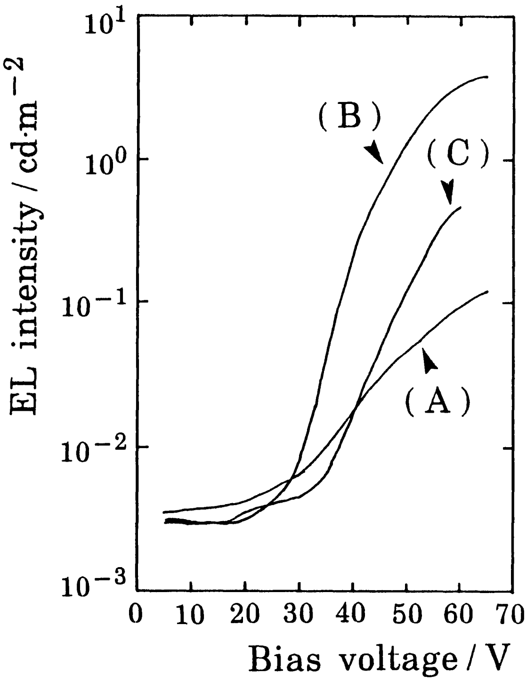

(A): ITO/1/Mg,(N-type)
(B): ITO/TPD/1/Mg,(H-type)
(C): ITO/1/Butyl-PBD/Mg,(E-type)

Fig.2. Current-voltage characteristics for the EL devices.

(A): ITO/1/Mg,(N-type)
(B): ITO/TPD/1/Mg,(H-type)
(C): ITO/1/Butyl-PBD/Mg,(E-type)

Fig.3. EL intensity-voltage characteristics for the EL devices.

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