*Macromolecules* **1999,** *32,* 4849−4854

# Synthesis of Polymers Having a Hole Transporting Ability by Condensation Polymerization of *N*,*N*′-Diphenyl-*N*,*N*′-bis(4-methylphenyl)-(1,1′-biphenyl)-4,4′-diamine and Aldehydes

**J. M. Son,* T. Mori, K. Ogino, and H. Sato**

*Department of Material Systems Engineering, Faculty of Technology, Tokyo University of Agriculture and Technology, Koganei, Tokyo, 184-8588, Japan*

**Y. Ito**

*Materials Research Laboratory, Toppan Printing Co., Ltd., Sugitomachi, Kitakatsusikagun, Saitama, 345-0046, Japan*

*Received January 12, 1999; Revised Manuscript Received May 17, 1999*

ABSTRACT: Novel polymers having a hole transport ability were prepared by condensation polymerization of *N*,*N*′-diphenyl-*N*,*N*′-bis(4-methylphenyl)-(1,1′-biphenyl)-4,4′-diamine (TPD) and paraformaldehyde (FA) or benzaldehyde (BzA). From NMR spectra, it is revealed that addition condensation reactions occurred exclusively at the para positions of TPD. TPD−FA polymer was linked not only by a methylene linkage but also by a methylene ether linkage, while TPD−BzA polymer was only linked by a methine linkage. The glass transition temperatures of TPD−FA and TPD−BzA were 183 and 239 °C, respectively. The drift mobility of TPD−BzA measured by a standard time-of-flight (TOF) method was found to be on the order of $10^{-5}$ and $10^{-6}$ $cm^2/(V\ s)$. The multilayer EL devices were fabricated using TPD−FA and TPD−BzA polymers as a hole transport layer and Alq as an electron transport emitting layer. In both devices, the initial driving voltage is about 4 V, and the maximum luminance is above 10 000 $cd/m^2$ at 14 V. It is expected that these polymers can be used as a hole transport material in the EL device.

## Introduction

One of the most important contemporary problems in polymer science is the design, synthesis, characterization, and processing of new polymeric materials for applying to photonic and electronic technologies including an organic photoconductor (OPC) and electroluminescent (EL) devices. A related perennial problem is the fundamental understanding of the underlying structure−property relationships in the materials. Polymers containing triphenylamine units, such as *N*,*N*′-diphenyl-*N*,*N*′-bis(3-methylphenyl)-(1,1′-biphenyl)-4,4′-diamine (*m*-TPD)[1] invented by Xerox, in the main chain or side chain of polymers represent one of the areas of investigation relative to these problems.[2,3] Research in the past decade has led to important progress in the experimental and theoretical understanding of electronic and electrical properties in these polymers and achievement of drift mobility on the order of $10^{-5}$ $cm^2/(V\ s)$ in several polymers introducing *N*,*N*′-diphenyl-*N*,*N*′-bis(4-methylphenyl)-(1,1′-biphenyl)-4,4′-diamine (TPD).[4] To prepare charge transporting polymers, however, many investigators prepared a monomer by introducing one or two functional groups to a molecule having charge transporting ability.[5−7] This method needed a long sequence of reaction steps for preparation of monomers and its polymerization.

It is expected that aldehyde may attack protons at the ortho or para position of TPA to yield a polymer in the presence of an acid catalyst like a phenol−formaldehyde resin, since triphenylamine derivatives (TPAs) have an electron-donating amino nitrogen group. The condensation reactions of triphenylamine derivatives (TPAs) with carbonyl compounds have been studied in the presence of acid catalyst and found that paraformaldehyde (FA), benzaldehyde (BzA), and paraldehyde had a high reactivity toward TPAs.[8−10]

In our previous works, when triphenylamine (TPA) was reacted with equal molar FA, structural characterization indicated that the addition condensation reaction occurred exclusively at para positions of TPA. The reaction mixture became viscous due to the molecular weight increase, and finally it provided insoluble gel, because TPA has three reaction points. Therefore, we used TPAs having two functionalities by introducing methyl groups at the para positions of the phenyl group in order to obtain linear polymers having a tertiary aromatic amine as monomers and obtained soluble polymers by condensation polymerization in high yield. Some of the TPAs−FA polymers have not only a methylene linkage but also a methylene ether linkage. The methylene ether linkage increased with increasing the concentration of formaldehyde. These polymers showed good solubility and sufficient morphological stability after the film formation, suggesting that this condensation polymerization provides a simple method for preparing charge transport polymers.

In this study, new polymers having a hole transport ability were synthesized by condensation polymerization of TPD with FA or BzA. The polymer structures and characteristics were investigated in comparison with polymers synthesized previously.

## Experimental Section

All the reagents for the TPD monomer synthesis were commercially obtained and were used without further purification unless otherwise noted. The paraformaldehyde (purity over 92%, KANTO) and benzaldehyde (purity over 98%, KANTO) were purchased. The solvents (1,4-dioxane, 1,2-

10.1021/ma990040r CCC: $18.00

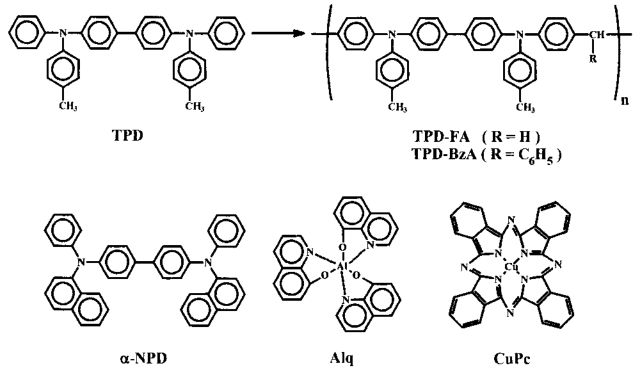

**Figure 1.** Synthetic route of polymers and chemical structures of materials used in this study.

dichloroethane, chlorobenzene) were freshly distilled and dried using common methods. The synthetic route of polymers and chemical structures of materials used in this study are shown in Figure 1.

**Synthesis of Monomer and Polymers.** *N*,*N*′-Diphenyl-*N*,*N*′-bis(4-methylphenyl)-(1,1′-biphenyl)-4,4′-diamine (TPD) was prepared from *N*,*N*′-diphenylbenzidine and 4-iodotoluene by the modified Ullmann reaction using 18-crown-6 as a phase transfer catalyst.[11] Into a 500 mL flask, 20 g (59 mmol) of *N*,*N*′-diphenylbenzidine, 311 g (1427 mmol) of *p*-iodotoluene, 36.5 g (264 mmol) of $K_2CO_3$, 30.34 g (16 mmol) of CuI, and 5.55 g (21 mmol) of 18-crown-6 were added and stirred at 200 °C until disappearance of the *N*,*N*′-diphenylbenzidine as verified by TLC. After the reaction, the precipitate was removed by filtration and 4-iodotoluene by distillation under reduced pressure. The product was purified by column chromatography. TPD was obtained with a yield of 80%. The purity was close to 99.0%, according to the $^1$H NMR spectrum. $^1$H NMR ($CDCl_3$) $\delta$ from TMS: 2.3 [s, 6H, $-CH_3$], 6.8−7.4 [m, 26H, aromatic protons].

Polymerization was carried out in a glass ampule by using 1,4-dioxane solvent which was purified conventionally and *p*-toluenesulfonic acid catalyst (purity over 98%) under nitrogen atmosphere. Polymer was recovered by pouring the reaction mixture into an excess amount of acetone and then purified by reprecipitating the toluene solution into acetone twice, followed by drying in a vacuum.

**Measurement.** $^1$H and $^{13}$C NMR spectra were measured on a JEOL α-500 spectrometer (500 and 125 MHz) in $CDCl_3$ at 50 °C with tetramethylsilane as an internal standard. The molecular weight of polymer was estimated by gel permeation chromatography using a JASCO 880-PU pump, a column packed with styrene−divinylbenzene gel beads, and a JASCO UV-970 detector. Chloroform was used as an eluent, and the molecular weight was calibrated using polystyrene standards (Shodex). Glass transition temperatures were measured by a Seiko Instruments DSC 220 at the temperature increase rate of 10 K/min. The redox potential was measured with cyclic voltammetry in a one-compartment cell with a polarization unit (TOHO PS-06). The measurement was conducted for the cast film on a platinum working electrode in dry acetonitrile containing tetrabutylammonium perchlorate (0.1 M) as an electrolyte under nitrogen atmosphere. Platinum spiral was used as a counter electrode and Ag/AgCl as a reference electrode.

Drift mobility was measured with a conventional time-of-flight method at room temperature using a xenon flash lamp (Hamamatsu Photonics, L2359) and a digitizing oscilloscope (GOULD, DSO 630). The cell for the measurement was prepared as follows: Titanyl phthalocyanine (TiOPc) was spin-coated from organic solvent onto the aluminum vapor deposited on the slide glass and then dried at 90 °C for 1 h. The chlorobenzene solution (typically 0.1 g/mL) of polymer was filtered through a 0.5 $\mu$m filter. The polymer solution was spread on a TiOPc layer using a wire bar or a spin coater. The films were allowed to dry for 3 h at room temperature and at 90 °C for 1 h. A semitransparent gold electrode was prepared on the film surface by vacuum deposition at a pressure of (3−7) × $10^{-6}$ Pa. Film thickness was determined by a profilometer (DEKTAK II, Solan).

EL devices were fabricated using TPD−FA and TPD−BzA polymer as a hole transport layer. On the ITO-coated glass was introduced copper phthalocyanine (CuPc) to improve the contact between the polymer and ITO, which suppressed voltage increase during operation of an EL device in a constant current mode. Thin films of these polymers were prepared by the spin-coating method on the CuPc layer. Aluminum tris-(8-hydroxyquinoline) (Alq) was introduced by vacuum deposition on the polymer film. The cathode was a MgAg alloy with an atomic ratio of 10:1, which was also formed by the coevaporation method. The luminance of the EL device was measured with a luminance meter (TOPCON, BM-3), and the $I-V$ characteristics were measured by an electrometer (ADVANTEST, TR-6143).

## Results and Discussion

**Molecular Weight and Solution Properties of Polymers.** TPD was reacted with FA and BzA in the presence of *p*-toluenesulfonic acid. The reaction mixture became blue when heated, which indicates that a portion of the catalyst was used to protonate TPD units. The blue color vanished when the reaction mixture was poured into acetone, precipitating solvent of the polymer.

Table 1 lists the conditions and the results of the polymerization. It is found that the weight-average molecular weight, polydispersity, and yield increased with increase of catalyst concentration and reaction temperature for the same reaction time. By using 1,4-dioxane as a solvent, the TPD−FA polymer in the presence of 1 mol % of catalyst at 60 °C had a molecular weight of about 0.47 × $10^4$. When the catalyst concentration was increased to 4 mol %, the molecular weight increased to 0.93 × $10^4$. As the reaction temperature increased to 80 °C, the molecular weight increased to 1.52 × $10^4$. The polymers, yielded in the presence of 2 mol % of catalyst at 80 °C, had a molecular weight about 1.02 × $10^4$ for 6 h and 3.30 × $10^4$ for 20 h, respectively.

The effect of reaction solvent was investigated on this condensation reaction system. With 2 mol % of catalyst, at 80 °C and for 6 h, the polymer yield increased on the order of 1,4-dioxane, chlorobenzene, and 1,2-dichloroethane. However, by using chlorobenzene or dichloroethane as a solvent, the polymer contained 10−20% of insoluble fraction. The difference of yield and gel formation among the three types of solvent can be explained by the basicity of the solvent; the ether oxygen of dioxane acted as an base, leading to the decrease of catalyst activity, and $\pi$-electrons of the benzene ring in chlorobenzene also acted as a weak base.

The condensation polymerization with TPD and BzA (no. 10) was carried out with 1, 2-dichloroethane, as a reaction solvent, in the presence of 4 mol % of catalyst at 80 °C. The polymer was obtained with a yield of 91% and had no insoluble parts. However, the molecular weight of the TPD−BzA polymer was lower than TPD−FA polymer, when the reaction conditions are the same. These results indicate that the hydroxybenzyl cation produced by the protonation of BzA is more stable and a weaker electrophlic species than the methylol cation due to the resonance with the benzene ring.

The effect of reaction temperature was investigated in the range 80−120 °C and used chlorobenzene, which had a higher boiling point than 1,2-dichloroethane (nos. 11−13). The products were obtained with a yield of about 80%, and the molecular weight increased with

**Table 1. Reaction Conditions and Molecular Weight of Polymers**

| no. | FA | BzA | solvent | catalyst (mol %) | reaction temp (°C) | reaction time (h) | yield (%) | $M_w{}^a$ ($\times 10^{-4}$) | $M_w/M_n$ |
|---|---|---|---|---|---|---|---|---|---|
| 1 | 1.0 | | dioxane | 1 | 60 | 6 | 64 | 0.47 | 1.53 |
| 2 | 1.0 | | dioxane | 4 | 60 | 6 | 85 | 0.93 | 1.56 |
| 3 | 1.0 | | dioxane | 2 | 80 | 6 | 83 | 1.02 | 1.91 |
| 4 | 1.0 | | dioxane | 2 | 80 | 20 | 90 | 3.30 | 3.77 |
| 5 | 1.0 | | dioxane | 4 | 80 | 6 | 91 | 1.52 | 2.36 |
| 6 | 1.0 | | dichloroethane | 2 | 80 | 6 | 98 (79) | 39.1 | 24.9 |
| 7 | 1.0 | | dichloroethane | 4 | 60 | 6 | 97 (86) | 1.67 | 2.43 |
| 8 | 1.0 | | chlorobenzene | 2 | 80 | 6 | 95 (80) | 5.65 | 5.52 |
| 9 | 1.0 | | chlorobenzene | 4 | 60 | 6 | 86 (77) | 1.27 | 2.21 |
| 10 | | 1.0 | dichloroethane | 4 | 80 | 6 | 91 | 1.10 | 1.46 |
| 11 | | 1.0 | chlorobenzene | 4 | 80 | 6 | 81 | 1.30 | 1.95 |
| 12 | | 1.0 | chlorobenzene | 4 | 100 | 6 | 80 | 1.09 | 1.67 |
| 13 | | 1.0 | chlorobenzene | 4 | 120 | 6 | 89 | 1.57 | 1.66 |
| 14 | | 1.2 | chlorobenzene | 2 | 120 | 6 | 92 | 1.90 | 1.61 |
| 15 | | 1.2 | chlorobenzene | 4 | 120 | 1.5 | 98 (80) | 32.0 | 11.7 |

[a] Estimated values by GPC (polystyrene standards, eluent as chloroform).

increasing reaction temperature. When the reaction temperature was 120 °C, the molecular weight increased to $2.69 \times 10^4$. When BzA concentration was increased to 1.2 times molar BzA (nos. 14, 15), the molecular weight of polymer increased to $2.60 \times 10^4$ and $32.0 \times 10^4$ in the presence of 2 and 4 mol % of catalyst, respectively. Thus, higher reaction temperature, higher catalyst concentration, or higher BzA concentration is needed to produce high molecular weight compounds by the condensation of TPD with BzA.

The TPD−FA and TPD−BzA polymers, except for gel fraction, are soluble in common organic solvents, such as toluene, chloroform, and chlorobenzene, and give solutions with light-yellow color. By casting from chlorobenzene solution, excellent transparent yellowish films on the order of 10 μm were obtained from TPD−BzA with a molecular weight of $32.0 \times 10^4$. On the other hand, the TPD−FA polymer was too brittle to provide high-quality films on the order of 1−10 μm; a few cracks appeared in the films during solvent evaporation.

**Polymer Structures.** $^1H$ NMR and $^{13}C$ NMR spectra of the polymers (TPD−FA (no. 3), TPD−BzA (no. 11)) are shown in Figures 2 and 3. Signals were assigned by considering the NMR spectra of the polymers reported previously[8,9] and of the phenol−formaldehyde resin.[12]

TPD−FA showed a methyl proton signal at 2.24 ppm and two types of methylene signals at 3.65 and 3.87 ppm with the intensity ratio of 15:85. The signal at 3.87 ppm is assigned to methylene proton linked with para positions and the signal at 3.65 ppm to methylene proton linked with meta positions of methylphenyl groups. The intensities of methylene at meta positions to para positions increased with increasing the yields and the polymer molecular weights. In the case of higher molecular weight polymers, such as nos. 4, 6, and 8, the intensity ratios of two methylene signals of TPD−FA were 22:78, 23:77, and 17:83. The presence of methylene group at the meta position indicates that some portion of the TPD monomer acted as a tri- or tetrafunctional monomer, which may be the reason for the production of cross-linked polymer when the reaction proceeded to a yield higher than 95%.

TPD−BzA showed only two types of aliphatic signals at 2.28 and 5.39 ppm. The signal at 2.28 ppm is assigned to methyl proton and the signal at 5.39 ppm to methine proton. In the case of BzA, the methylphenyl group was not reacted even by increasing the yield and by using an excess amount of BzA. This result can be explained by the larger steric hindrance of BzA unit in comparison to FA.

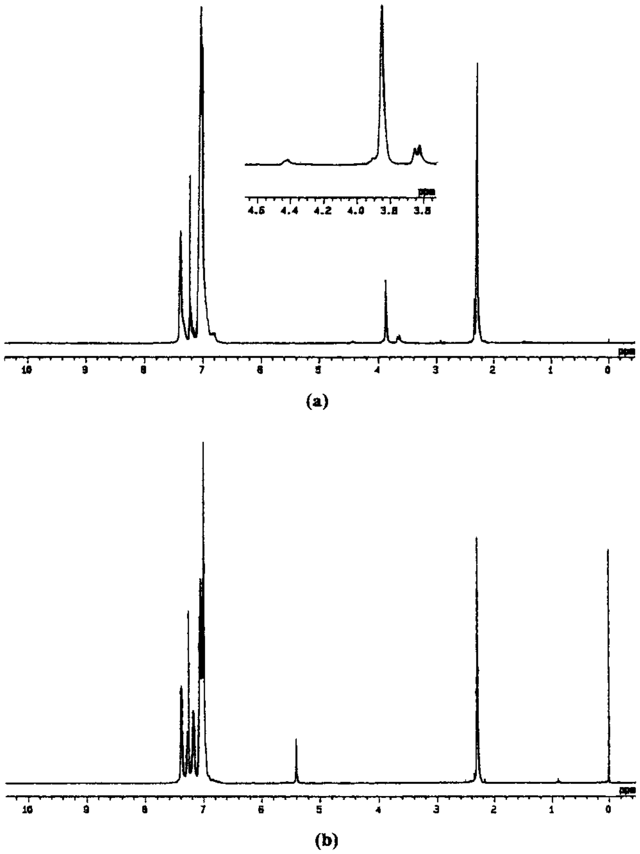

**Figure 2.** $^1H$ NMR spectra of TPD−FA (a) and TPD−BzA (b).

**Other Characterizations.** Charge transport in organic materials is believed to be governed by the hopping process involving redox reaction of charge transport molecules. Cyclic voltammetry (CV) is a preliminary characterization method to determine the redox properties of organic and polymeric materials. Table 2 shows the oxidation−reduction potentials of polymers. Chloroform solution of the polymer was cast on a platinum electrode, and the cyclic voltammogram was measured in acetonitrile containing 0.1 M $Bu_4NClO_4$ at a scanning rate of 100 mV/s. Two pairs of redox waves were observed in both TPD−FA and TPD−BzA polymers. TPD−FA showed two oxidation peaks

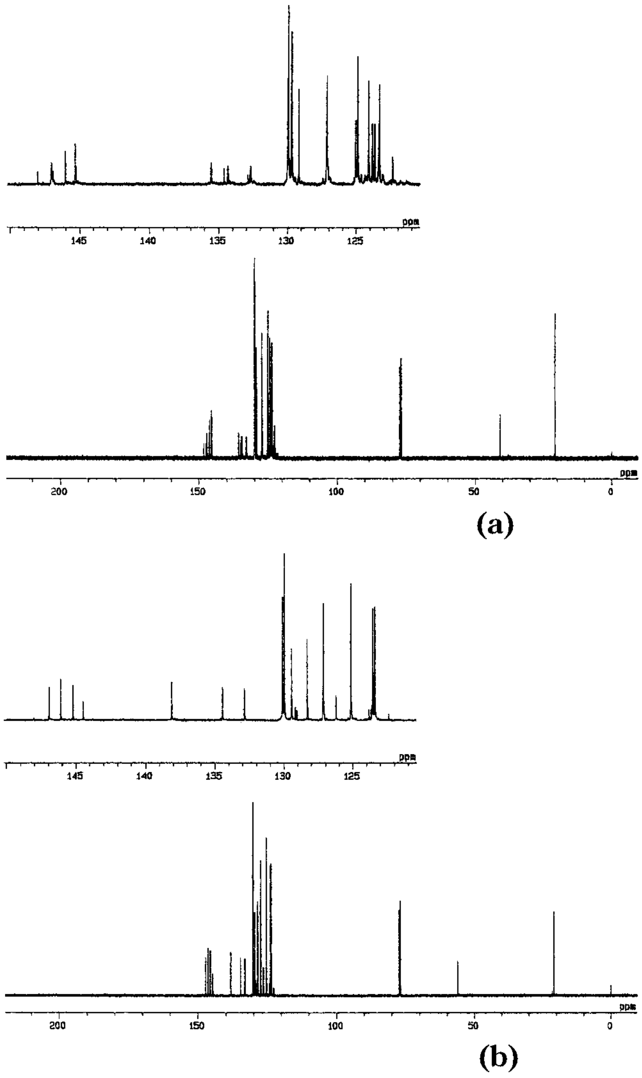

**Figure 3.** $^{13}C$ NMR spectra of TPD–FA (a) and TPD–BzA (b).

**Table 2. Redox Potentials of Polymers**[a]

| | Epa 1 (eV) | Epa 2 (eV) | Epc 1 (eV) | Epc 2 (eV) |
|---|---|---|---|---|
| TPD–FA | 0.92 | 1.01 | 0.68 | 0.82 |
| TPD–BzA | 0.87 | 0.98 | 0.73 | 0.83 |

[a] Epa and Epc are oxidation and reduction potential, respectively. Pt working and counter electrode, Ag/AgCl reference electrode, 0.1 M ($(n\text{-Bu})_4NClO_4$) in acetonitrile.

at 0.92 and 1.01 V and two reduction peaks at 0.68 and 0.82 V. The redox behavior of TPD–BzA was almost identical with that of TPD–FA. The intensity of redox peaks decreased as cycle time increased, which can be explained by the dissolution of ionized polymer into the solvent. These polymers were colorless in the neutral state and turned to blue in the ionized state.

TPD–FA has two UV absorption maxima at 313 and 353 nm. The absorption maxima of TPD–BzA were almost identical to those of TPD–FA. These values were almost identical to that of the TPD monomer. Thus, it is assumed that the electronic structures of the TPD units were not greatly changed after the reaction with aldehydes.

The transition temperature of the polymers was measured in the range −20 to 250 °C. TPD–FA and TPD–BzA polymers had glass transition temperatures of 183 and 239 °C, respectively. These values are much higher than those of the low molecular weight compound (TPD).

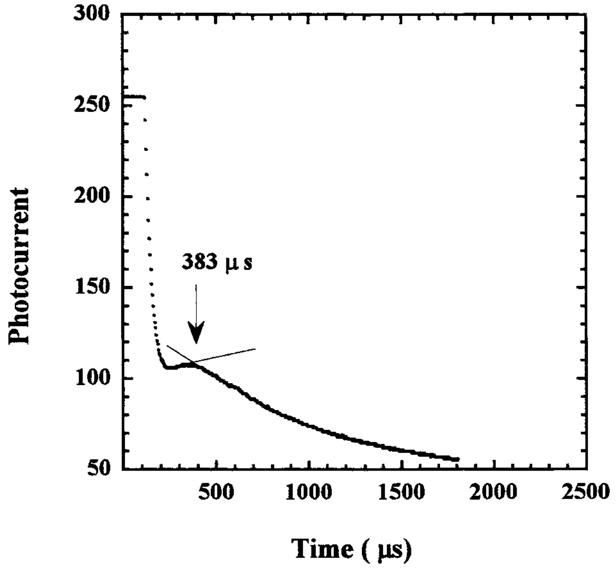

**Figure 4.** Photocurrent of TPD–BzA polymer at 2.67 × $10^5$ V/cm.

**Drift Mobility and Application to EL Device.** The hole drift mobility was measured by the standard time-of-flight. The mobility, $\mu$, is calculated according to the following equation:

$$\mu = L^2/t_T V$$

where $L$ is the sample thickness, $t_T$ the transit time, and $V$ the applied voltage. TPD–FA polymer was too brittle to provide high-quality thin films for TOF measurements. On the other hand, TPD–BzA polymer provided a good film with a thickness of 11.6 $\mu$m by the bar-coating method. Figure 4 shows the photocurrent change of TPD–BzA polymer at the applied voltage of 310 V or at the electric field of 267 kV/cm. From the figure the transit time was determined to be 383 $\mu$s, which leads to a mobility of 1.15 × $10^{-5}$ cm$^2$/(V s). The logarithm of the mobility of the polymer was plotted against the square root of the applied field $E$ (V/cm) as shown in Figure 5. The drift mobility of TPD–BzA polymer was on the order of $10^{-5}$ and $10^{-6}$ cm$^2$/(V s). The log $\mu$ vs $E^{1/2}$ plot for polymer showed a linear relationship with negative slopes. The negative slope was also observed in some poly(phenylenevinylene) derivatives, 1,1-bis(di-4-(tolylamino)phenyl)cyclohexane-doped polycarbonate, and conjugated photorefractive polymers.[13−15]

The multilayer EL devices were fabricated using TPD–FA ($C_{39}H_{32}N_2$ (repeating unit); Anal. Calcd: C, 88.60%; H, 6.10%; N, 5.30%. Found: C, 88.76%; H, 6.17%; N, 5.00%) and TPD–BzA ($C_{45}H_{36}N_2$ (repeating unit); Anal. Calcd: C, 89.37%; H, 6.00%; N, 4.63%. Found: C, 89.30%; H, 6.11%; N, 4.20%) as a hole transport layer and Alq as a electron transport emitting layer. Organic materials, except for these polymers, were evaporated onto an indium–tin oxide (ITO)-coated glass substrate. The voltage–luminance characteristics of each device are shown in Figure 6. The maximum luminance of device reached 18 290 cd/m$^2$ at 14 V when TPD–FA polymer was used and 10 060 cd/m$^2$ at 13 V for the device with TPD–BzA polymer. The voltage–current density characteristics are shown in Figure 7.

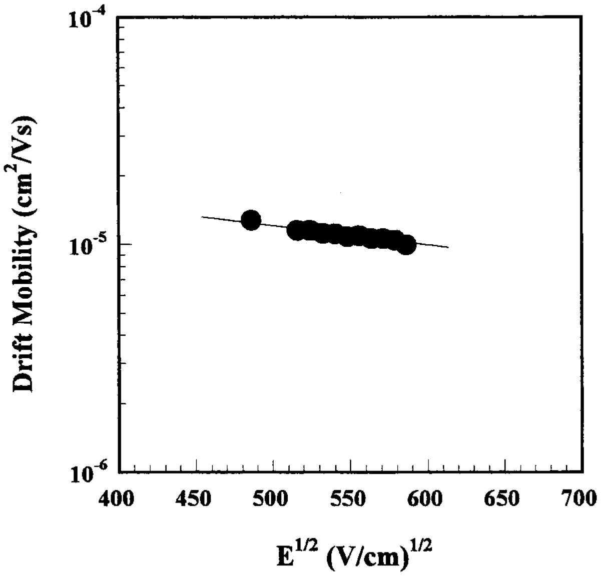

**Figure 5.** Drift mobility against the square root of applied field *E* (V/L) for TPD−BzA polymer.

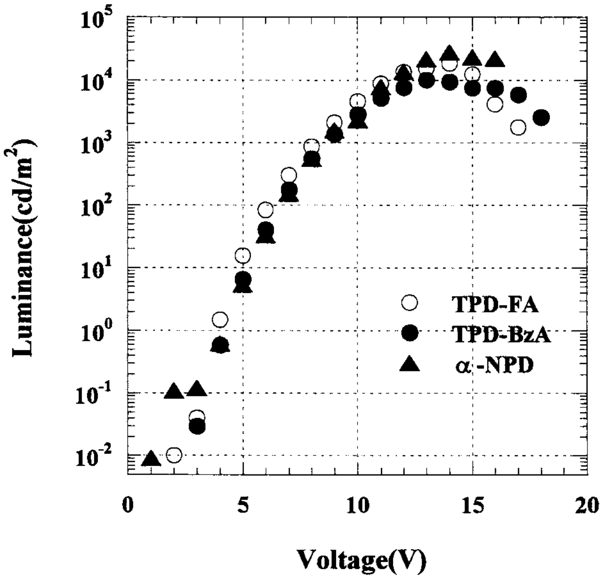

**Figure 6.** Voltage−luminance characteristics of the EL device using polymer (ITO/CuPc/polymer/Alq/MgAg).

The device using TPD−BzA polymer showed slightly higher current density at the same voltage than one using TPD−FA polymer. At the maximum luminance, the luminous efficiencies of these devices were 0.44 and 0.30 lm/W, respectively. For comparison, characteristics of the device consisting of *N*,*N*′-di(α-naphthyl)-*N*,*N*′-diphenyl-(1,1′-biphenyl)-4,4′-diamine (α-NPD) (device structure: ITO/CuPc/α-NPD/Alq/MgAg) are shown in Figures 6 and 7. The devices consisting of polymers had almost similar EL performance to that with the low molecular weight compound.

In addition, the devices containing α-NPD (10 nm) between polymers and Alq were fabricated. The voltage−luminance characteristics of each device are shown in Figure 8. The maximum luminance of a device reached 26 340 cd/m$^2$ at 14 V when TPD−FA polymer was used and 22 040 cd/m$^2$ at 14 V for the device with TPD−BzA polymer. At the maximum luminance, the luminous efficiencies of these devices were 0.73 and 0.66 lm/W, respectively. The EL performance of these devices improved by inserting α-NPD between polymer and Alq. It is considered that the polymer layer contains the impurities such as ionic compounds (catalyst), solvents (spin-coating), and low molecular weight compounds (monomer and dimer, etc.), and these impurities have an effect on the luminance processes occurring at the interface of polymer and Alq for devices without α-NPD. By inserting α-NPD, the effect of impurities became negligible. Thus, the higher EL performance was accomplished reflecting the inherent higher thermal stability of the polymer layer. Furthermore, the lifetime of the two types of devices using TPD−BzA was 5 h without α-NPD and increased to 70 h by inserting α-NPD with a constant-current dc drive at 20 mA/cm$^2$.

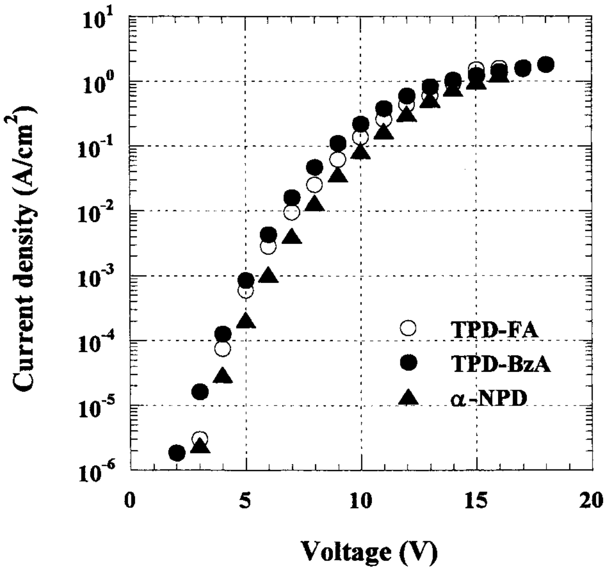

**Figure 7.** Voltage−current density characteristics of the EL device using polymer (ITO/CuPc/polymer/Alq/MgAg).

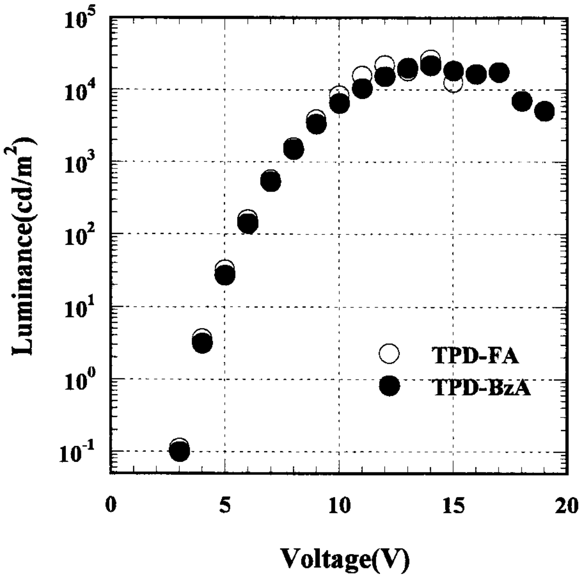

**Figure 8.** Voltage−luminance characteristics of the EL device by inserting α-NPD between polymer and Alq layer (ITO/CuPc/TPD−FA or TPD-BzA/α-NPD/Alq/MgAg).

## Conclusions

Novel polymers having a hole transport ability were prepared by condensation polymerization of *N*,*N*′-diphenyl-*N*,*N*′-bis(4-methylphenyl)-(1,1′-biphenyl)-4,4′-diamine (TPD) with paraformaldehyde or benzaldehyde. From NMR spectra, it is indicated that addition condensation reactions occurred almost at the para positions of TPD. And TPD−FA polymer was linked by not only the para position of the phenyl unit but also the meta position of the methylphenyl unit, while TPD−BzA polymer was only linked at the para position. The glass transition temperatures of TPD−FA and TPD−BzA were 183 and 239 °C, respectively. The drift mobility of TPD−BzA measured by the standard time-of-flight (TOF) was found to be on the order of $10^{-5}$ and $10^{-6}$ $cm^2/(V\ s)$.

EL devices were fabricated using TPD−FA and TPD−BzA polymer as a hole transport layer. In both devices, the initial driving voltage is about 4 V and the maximum luminance is above 10 000 $cd/m^2$ at 14 V. It is expected that these polymers can be used as a hole transport material in the EL device.

## References and Notes

(1) Stolka, M.; Yanus, J. F.; Pai, D. M. *J. Phys. Chem.* **1984**, *88*, 4707.

(2) Kido, J.; Komada, M.; Harada, G.; Nagai, K. *Polm. Adv. Technol.* **1995**, *6*, 703.

(3) Kido, J.; Harada, G.; Nagai, K. *Polym. Adv. Technol.* **1996**, *7*, 31.

(4) Ogino, K.; Nomura, T.; Shichi, T.; Park, S. H.; Sato, H.; Aoyama, T.; Wada, T. *Chem. Mater.* **1997**, *9*, 2768.

(5) Son, J. M.; Sakaki, Y.; Ogino, K.; Sato, H. *IEEE Trans. Electron Devices* **1997**, *44*, 1307.

(6) Jhon, S. F.; Martin, A.; William, L.; Fred, K.; Jhon, Y.; Dale, R. *J. Phys. Chem.* **1991**, *95*, 7908.

(7) Yamamori, A.; Adachi, C.; Koyama, T.; Taniguchi, Y. *Appl. Phys. Lett.* **1998**, *72*, 2147.

(8) Son, J. M.; Nakao, M.; Ogino, K.; Sato, H. *Macromol. Chem. Phys.* **1999**, *200* (1), 65.

(9) Son, J. M.; Ogino, K.; Yonezawa, N.; Sato, H. *Synth. Met.* **1998**, *98*, 71.

(10) Kraft, A.; Burn, P. L.; Holmes, A. B.; Bradley, D. D. C.; Friend, R. H.; Martens, J. H. F. *Synth. Met.* **1993**, *55−57*, 4163.

(11) Gauthier, S.; Frechet, J. M. J. *Synth. Commun.* **1987**, *383* (April).

(12) Yoshikawa, T.; Kimura, K. *Makromol. Chem.* **1974**, *175*, 1001.

(13) Takiguchi, T.; Park, D. H.; Ueno, H.; Yoshino, K. *Synth. Met.* **1987**, *17*, 657.

(14) Borsenberger, P. M.; Pautmeier, L.; Bassler, H. *J. Chem. Phys.* **1991**, *94*, 5447.

(15) Chan, W. K.; Chen, Y.; Peng, Z.; Yu, L. *J. Am. Chem. Soc.* **1993**, *115*, 11735.

MA990040R