Electroluminescence characteristics of organic light-emitting diodes with TPD doped PVK as the hole transport layer

Y. C. Shin[a]
Department of Electrical Engineering and computer science, Seoul National University, Seoul, Korea
J. H. Song
Department of physics, Inha university, Incheon, Korea
C. H. Lee
Department of Electrical Engineering and computer science, Seoul National University, Seoul, Korea

Abstract

We have fabricated organic light-emitting diodes using poly(N-vinylcarbazole)(PVK) doped with N,N'-diphenyl-N,N'-bis(3-methylphenyl)-[l,l'-biphenyl]-4,4/-diamine (TPD) as the hole transport layer. TPD molecules act as the trapping sites in PVK and reduce the hole mobility, which can enhance the electron-hole balance in the emitting layer, resulting in the enhanced device performance. We have found the optimum ratio of TPD to PVK for the EL efficiency.

1. Introduction

Organic light-emitting diodes (OLEDs) emerge as a new flat-panel display technology with superior display qualities since the first demonstration of efficient light emission by C. W. Tang[1]. In order to enhance the luminous efficiency of the OLEDs, it is necessary to optimize the device structure so that efficient injection and recombination of electrons and holes are possible.[1–7] One way to balance the electron and hole concentration in the emitting layer is to control the hole mobility of the hole transport layer (HTL) by doping with appropriate materials. Since the electron mobility of the electron transport layer (ETL) is much lower than the hole mobility of the HTL, the electron-hole balance can be improved by reducing the hole mobility of the HTL with doping. Therefore electron-hole recombination becomes more efficient, resulting in better electroluminescence (EL) efficiency.[7-12]

The carrier transport mechanism of poly(N-vinylcarbazole) (PVK) doped with N,N'-diphenyl-N,N'-bis(3-methylphenyl)-[l,l'-biphenyl]-4,4/-diamine (TPD) was well analyzed by D. M. Pai, J. F. Yanus, and M. Stolka.[13] They found that TPD acts as randomly distributed trapping sites and the hole transport proceeds primarily via PVK at low concentration ratio of TPD to PVK. Thus, the hole mobility is drastically reduced and it reaches its minimum at about 2 % weight ratio of TPD to PVK. As the ratio of TPD to PVK increases above this ratio, the hole transport proceeds mainly via hopping through TPD sites with little participation of PVK, and therefore the mobilities increase up to that of TPD.

Since the TPD doped PVK shows a broad range of hole mobility depending on the doping concentration, they offer a good opportunity to study the dependence of the EL performance on the hole mobility of the HTL. In this work, we have analyzed the current-voltage-luminescence (I-V-L) characteristics and the EL quantum efficiency for the devices with TPD doped PVK as the hole transport layer.

2. Experimental

We have fabricated organic light-emitting diodes using TPD doped PVK as the hole transport layer. The molecular structure of TPD and PVK and the device structure are shown in figure 1. Indium-tin oxide(ITO) glass was used as the substrate and anode, and routinely cleaned by ultrasonic treatment. The TPD doped PVK layer was deposited on top of the precleaned ITO glass by spin coating the solution of various ratio (0, 1, 5, 10, 20, 50, 70, and 100 %) of TPD to PVK. Tris-(hydroxyquinoline) aluminum (Alq3) is used as the emitting layer, LiF as the electron injecting layer and Al as the cathode. The thickness of TPD doped PVK layer was 60 nm and was verified by AFM. Alq3, LiF, and Al were

[a] Corresponding author Tel: +82-2-880-9132; e-mail address: tangsu6@snu.ac.kr

thermally evaporated onto the TPD doped PVK at a vacuum under 3×10^{-6} Torr with a thickness of 70, 0.5, and 100 nm respectively. The active area of the device was $1.96 mm^2$.

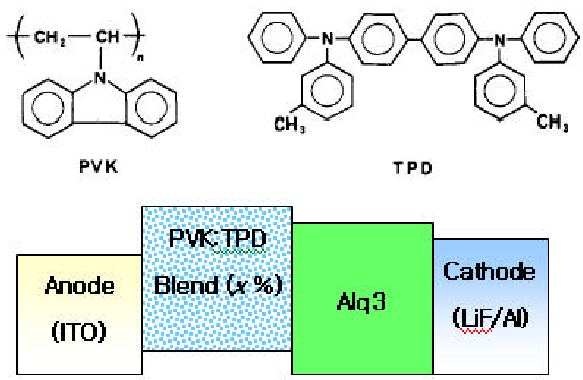

Figure 1. Molecular structure of TPD and PVK and the device structure

3. Results and discussion

Current-light-voltage (I-V-L) characteristics of the devices with different ratio of TPD to PVK are shown in figure 2. The current at a fixed voltage decreases, reaches a minimum at about 5 %, and then increases again as the ratio of TPD to PVK is raised from 5% to 70%. This result is consistent with the trend of the hole mobility of TPD doped PVK film, reported by Pai *et al.*[13] At low concentration of TPD to PVK, the hole current decreases with increasing the ratio of TPD to PVK since the mobility decreases due to the trapping at TPD. As the TPD concentration increases above a certain level where the hole transport starts to proceed via hopping through TPD sites rather than PVK, the hole mobility and the current increases. Figure 2-(b) shows that the light emission intensity is linearly proportional to the current density.

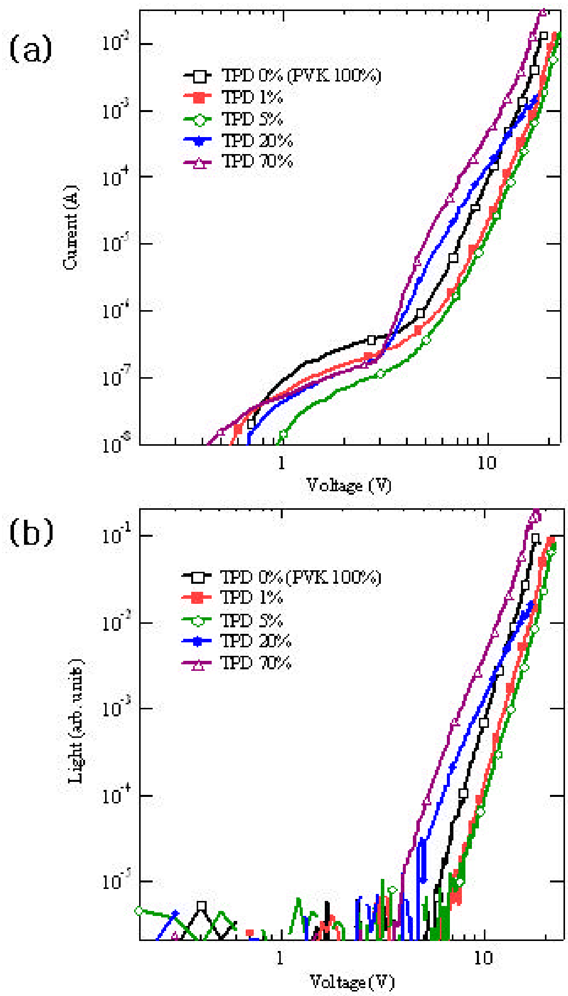

Figure 2. Current-voltage (a) and Light-voltage (b) characteristics of the devices with different ratio of TPD to PVK

Figure 3 shows the EL spectra of the devices with different ratio of TPD to PVK at a constant current density, $50 mA/cm^2$. The EL spectra are almost same for the devices with different ratio of TPD to PVK. Though the mobility of hole being adjusted by the addition of TPD to PVK, the recombination of electrons and holes still occur in emitting layer(Alq3) and the devices still show their peak in 530nm (peak of Alq3).

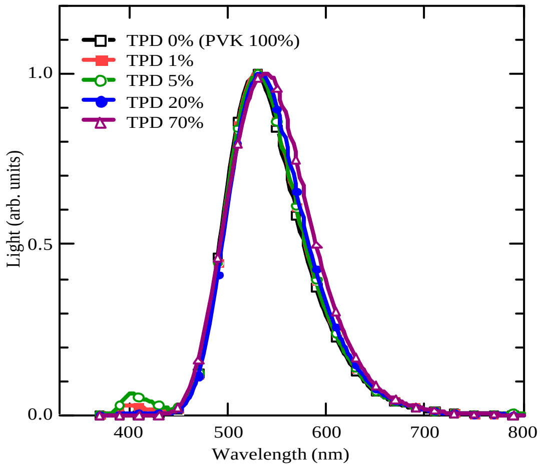

Figure 3. EL spectra of the devices with different ratio of TPD to PVK

The quantum efficiencies of various devices using different ratio of TPD to PVK are shown in figure 4. We have found that quantum efficiency reaches its maximum at 20 % weight ratio of TPD to PVK. We find that appropriate (in this case 20%) addition of trapping materials in hole transport layer can reduce hole mobility and enhance the performance of the device.

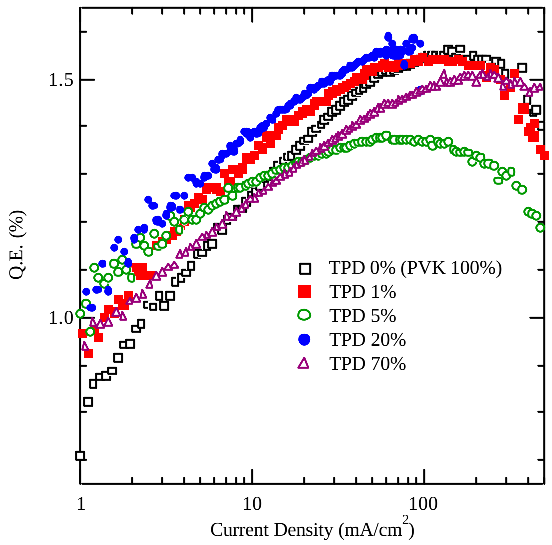

Figure 4. Quantum efficiencies of the devices with different ratio of TPD to PVK

To confirm our result, we have obtained mobilities at various ratio of TPD to PVK by transient EL method. Figure 5 shows the mobilities obtained from transient EL method at various concentration of TPD. We can verify that the hole mobilities decreases as the ratio of the trapping material TPD at low TPD concentration and then increases again, consistent with the result by Pai *et al.*[13]

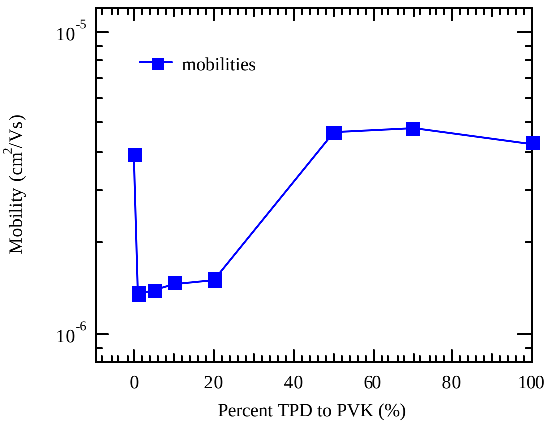

Figure 5. Mobilities obtained by transient EL method at a fixed bias voltage 11V

4. Conclusion

Carrier recombination as well as the balance of electrons and holes is important in determining the efficiency of OLEDs. In this work, we investigated the correlation of the EL quantum efficiency and the hole mobility of the HTP materials. We found that the QE can be optimized by controlling the hole mobility so that the electron and hole concentration is well balanced. To do this, we doped TPD in HTL as the trapping sites for holes and also verified that this really changed mobilities of the holes. This result can contribute to develop highly efficient OLEDs.

5. Acknowledgements

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6. References

[1] C. W. Tang and S. A. VanSlyke, Appl. Phys. Lett. **51**, 913 (1987)

[2] C. W. Tang, S. A. VanSlyke, and C. H. Chen, J. Appl. Phys. **65**, 3610 (1989).

[3] C. Adachi, S. Tokito, T. Tsutsui, and S. Saito, Jpn. J. Appl. Phys. **27**, L269 (1988).

[4] C. Adachi, S. Tokito, T. Tsutsui, and S. Saito, Jpn. J. Appl. Phys. **27**, L713 (1988).

[5] C. Adachi, T. Tsutsui, and S. Saito, Appl. Phys. Lett. **55**, 1489 (1989).

[6] H. Tokailin, M. Matsuura, H. Higashi, C. Hosokawa, and T. Kusumoto, SPIE **1910**, 38 (1993).

[7] Y. Shirota, Y. Kuwabara, H. Inada, T. Wakimoto, H. Nakada, Y. Yonemoto, S. Kawami, and K. Imai, Appl. Phys. Lett. **65**, 807 (1994).

[8] S. A. Van Slyke, C. H. Chen, and C. W. Tang, Appl. Phys. Lett. **69**, 2160 (1996).

[9] E. W. Forsythe, M. A. Abkowitz, and Yongli Gao, *J. Phys. Chem. B* **104**, 3948 (2000)

[10] H.H. Fong et al., Chem. Phys. Lett. **353**, 407 (2002).

[11] Y. Hamada, T. Sano, K. Shibata, and K. Kuroki, Jpn. J. Appl. Phys. Part 2 **34**, L824 (1995).

[12] M. S. Jang, S. Y. Song, H. K. Shim, T. Zyung, S. D. Jung, and L. M. Do, Synth. Met. **91**, 317 (1997).

[13] D. M. Pai, J. F. Yanus, and M. Stolka, J. Phys. Chem. **88**, 4714 (1984).