Optica Applicata, Vol. XLIV, No. 4, 2014
DOI: 10.5277/oa140407

Effect of a weak ion beam treatment of emission layer on the top emission polymer light-emitting diode

TAEWON GONG, JIN SEOG GWAG*

Department of Physics, Yeungnam University, Gyeongsan 712-749, Republic of Korea

*Corresponding author: sweat3000@ynu.ac.kr

This study examined the influence of a weakly treated emission layer on ion beam processing for the performance of top emission polymer light-emitting diodes with an invert structure (top ITO as the anode). The emission layer used in this experiment was a polymer type SY, Livilux PDY-132 provided by Merck & Co. The surface of the emission layer was modified by a low energy ion beam treatment to obtain hydrophilic functional groups and improve the wettability. As a hole transfer layer, poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS, CLEVIOS AI 4083, Heraeus) was spin-coated on the ion beam treated emission layer and showed good adhesion properties. Consequently, through such an ion beam treatment that promotes the interface properties of these two layers, a uniform light emitting area was obtained and the light intensity in a top emission polymer light-emitting diode was improved.

Keywords: organic light emitting diode, light-emitting polymer, top emission, optical device, ion beam, surface treatment.

1. Introduction

Since TANG and VANSLYKE's reports of light emission from an organic material in 1987 [1], scientists and engineers have been showing increasing interest in organic light-emitting diodes (OLEDs) for the development of high quality displays and lighting. Furthermore, OLEDs can be applied to flexible electric devices through roll-to-roll production [2]. OLEDs have excellent electro-optical properties suitable for displays, such as rapid response, high color purity, wider viewing angle, low power consumption, vivid colors, light weight, small thickness, color tenability, and dimmability. Therefore, active-matrix drives with a thin film transistor (TFT) are required to expand these advantages in displays with high quality image information. Considerable progress has been made in the development of top-emitting organic light-emitting diodes (TEOLEDs) that use small-molecules [3–10], but there have not been any comparable developments in top-emission polymer light-emitting diodes (TEPLEDs), which are competitors of TEOLEDs that are based on the conjugated polymers. Polymer light-emitting diodes (PLEDs) are attractive for flat-panel display technology because they provide superior color reliability, wider viewing angles and faster response times compared to liquid crystal displays, as well as an easy fabrication process and low cost compared to OLEDs. In addition, top emission type polymer light-emitting diodes (TEPLEDs) have properties that can improve the display functions and specifications, which include increasing the aperture ratio, extending the freedom of a pixel and circuit design. A top-emitting configuration is more suitable for active-matrix displays than the bottom emitting configuration, at which emitting light must pass through the substrate. Therefore, a top-emitting device can fully utilize the substrate area for displays, so that the device can operate at a relatively lower power to produce the same luminescence that can increase lifetime.

On the other hand, TEPLEDs have a demerit, such as a poor binding property at the interface between the emitting layer (EML) and the hole transfer layer (HTL) because of the difference in the surface properties of two materials, which produces non-uniform light emitting characteristics.

To solve this issue, this study examined the influence of a weakly-treated EML by ion beam processing on improving the performance of the TEPLEDs. Figure 1 shows the device structure used in these experiments.

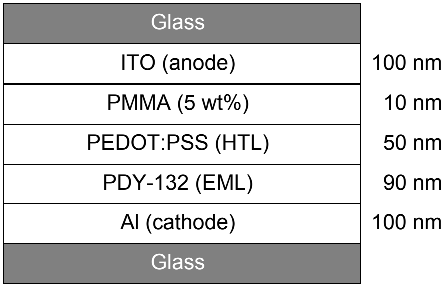

Fig. 1. TEPLED structure used in this experiment.

In this experiment, a polymer type SY, Livilux PDY-132 provided by Merck & Co. was used as the EML with the chemical structure shown in Fig. 2**a**. The surface of the EML was modified by a low energy ion beam treatment to obtain hydrophilic functional groups and improve the wettability. As HTL, poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS, CLEVIOS AI 4083, Heraeus), with the chemical structure shown in Fig. 2**b**, was spin-coated on the ion beam treated EML. The adhesion property between these two materials was then examined. In addition, indium tin oxide (ITO) was used as a top electrode (anode) to extract sufficient light

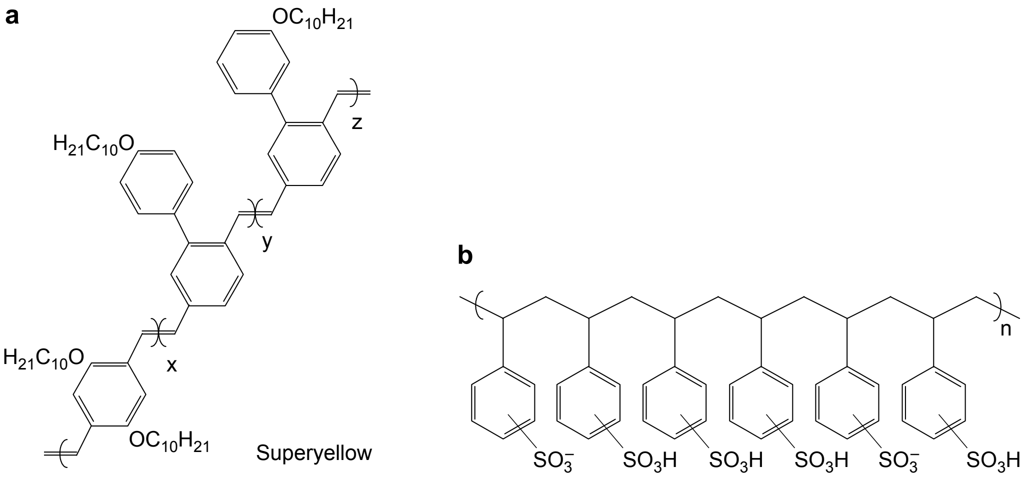

Fig. 2. Chemical structures of the EML (**a**) and HTL used in these experiments (**b**).

from the device and a very thin polymethylmethacrylate (PMMA) layer was adopted as a buffer layer to protect the HTL from damage during ITO deposition.

2. Measurements

To evaluate surface wettability, the water contact angles of the EML were measured at room temperature using a contact angle goniometer (Model G-I type, Erma, Inc., Tokyo, Japan). A droplet of water was placed on the air-side surface of a film. More than 5 measurements were taken and the values obtained were averaged (Figs. 3**a** and 3**b**). The samples were then measured under different conditions (100 to 300 eV and 30 to 90 s, respectively), as shown in Fig. 4. The luminance *vs.* voltage profiles were obtained using a luminance meter (Konica Minolta Sensing, Inc. T-10M, Japan) according to the ion beam treatment conditions. Intensity comparisons according to voltages

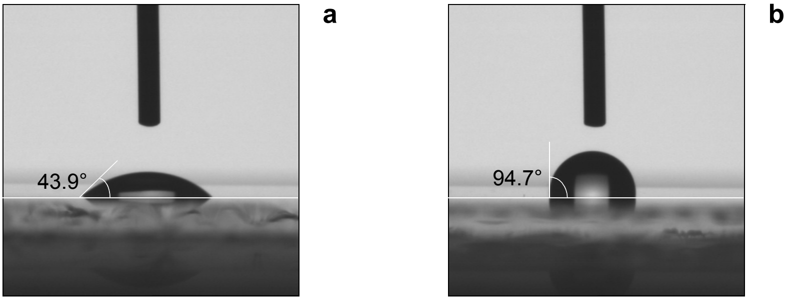

Fig. 3. Contact angles of EML treated by ion beam processing (100 eV, 10 mA for 60 s) (**a**) and of EML with no treatment (**b**).

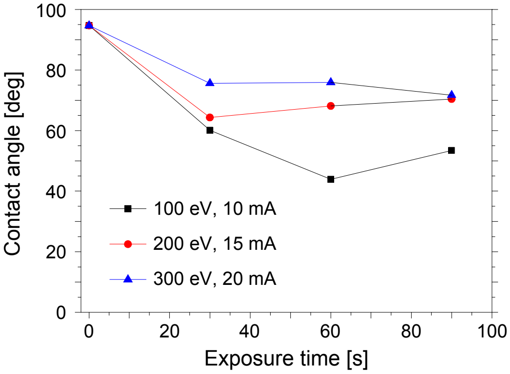

Fig. 4. Contact angles of the EML that was treated by ion beam processing with different exposure times.

Table 1. Specific resistivity and carrier mobility analyzed by Hall measurements as a substrate material of ITO.

Samples	Resistivity [×10^{-4} Ωcm]	Mobility [cm^2/Vs]
Glass	5.95	35
HTL	6.13	34
PMMA	6.18	30.8

between the used buffer layer with PMMA (5 wt%) and without the buffer layer are presented.

The specific resistivity and carrier mobility of ITO were analyzed by Hall effect measurements (Nanometrics HL5500 Hall System) according to different substrate materials under ITO, as shown in Table 1. This shows that the electrical properties of ITO are similar regardless of the substrate.

3. Results and discussion

When the TEPLEDs were fabricated, it was difficult to coat the HTL on the EML by spin-coating because of the wettability of the EML. Initially, a PLED with a HTL having an irregular wetting property on the ETL with no prior treatment was fabricated, as shown in Fig. 5**a**. As expected, irregular light-emission was observed, as shown in Fig. 5**b**. This problem was solved by an ion beam treatment. The surface of EML was modified by ion beam sputtering under different conditions.

Under 100 eV, 10 mA, and 60 s, the ETL surface had high wettability, as shown in Fig. 6**a**. PLED fabricated under these conditions showed regular light-emission, as shown in Fig. 6**b**.

In addition, the illumination of the PLEDs fabricated under various ion beam processing conditions was measured, as shown in Figs. 7**a** and 7**b**. In addition, the illumination of the PLED, which was treated under different conditions (100 eV, 10 mA and 60 s), was higher than the other PLEDs (Fig. 7**c**). A comparison of the contact angle

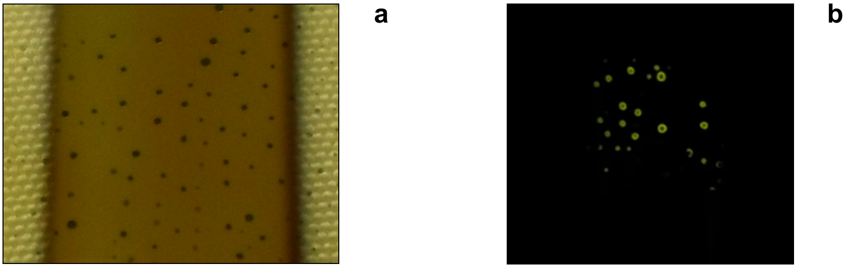

Fig. 5. HTL showing irregular wetting properties on the untreated ETL (**a**) and irregular light-emission at the TEPLED with the untreated ETL (**b**).

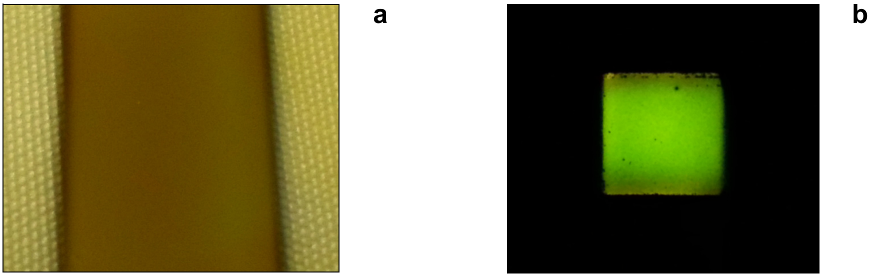

Fig. 6. HTL showing a uniform wetting property on the ion beam treated ETL (**a**) and uniform light-emitting at TEPLED with the ion beam treated ETL (**b**).

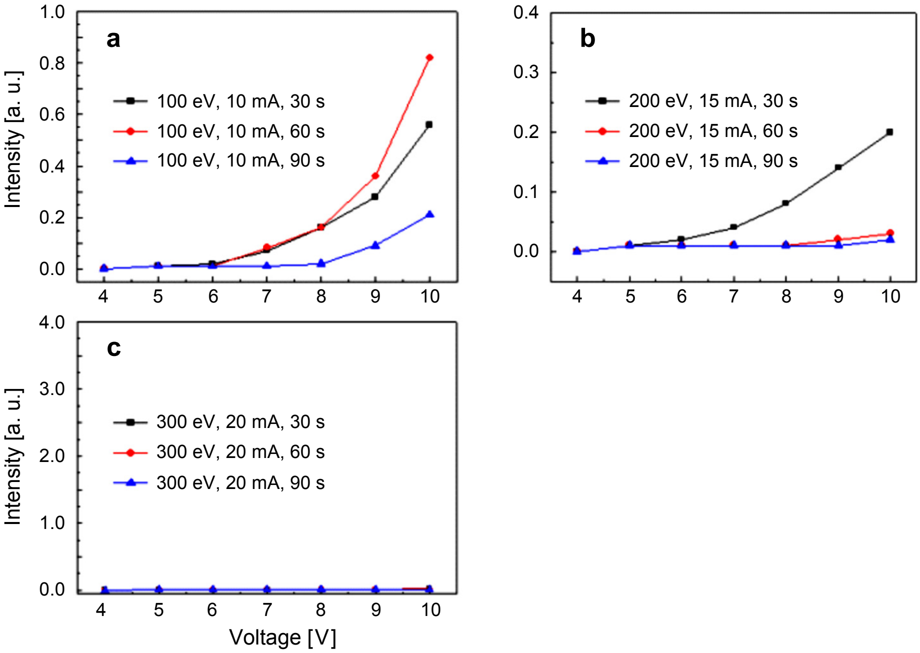

Fig. 7. Luminance according to the voltages with the conditions of the ion beam treatment.

and intensity showed that the intensity of illumination of PLEDs is related to an interfacial binding property. Hence, the contact angles of EML do not appear to depend completely on the voltage (eV) and exposure times, as presented in Fig. 4.

The intensity of PLED depending on the voltage was measured before and after using the PMMA on a HTL as a buffer layer. The intensity was also compared according to the voltage of the devices using PMMA (5 wt%) as a buffer layer and non-buffer layer, as shown in Fig. 8. Both samples were treated under the same ion beam conditions on EML. As a result, the TEPLED device with PMMA was more efficient, possibly because PMMA as a buffer layer protects the HTL from ITO deposition by sputtering.

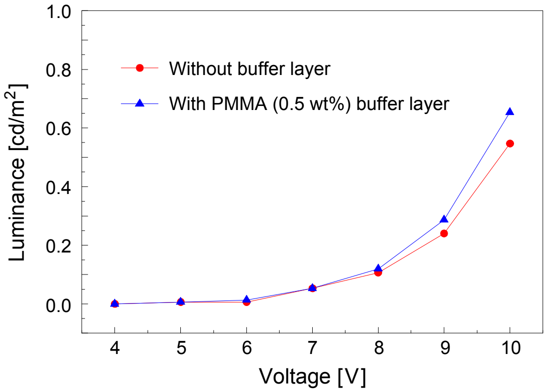

Fig. 8. Intensity comparison according to voltages of TEPLED samples with PMMA (5 wt%) as a buffer layer and without a buffer layer.

Figure 9 shows the spectral characteristics of the TEPLED sample with HTL+PMMA on weakly ion beam treated EML. As expected, its color shows clear superyellow.

Other polymer materials might be used as a protection layer of the HTL to achieve higher optical efficiency. In this experiment, PMMA was used as the protection layer

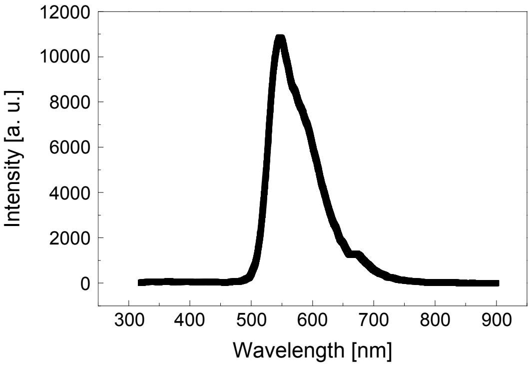

Fig. 9. Spectral characteristics of the TEPLED sample with HTL+PMMA on the weakly ion beam treated EML showing clear superyellow.

to reduce the glass transition temperature T_g. Polymers with a high T_g require a strong solvent to dissolve them. Such solvents can damage the underlayers, including the HTL, which may deteriorate the electro-optical properties of the device. Therefore, polymers with a lower T_g should be considered as the protection layer.

Under the exposure conditions of the low energy ion beam treatment, the EML could be etched about 5 nm from it. This had no effect on the physical properties of the EML because of the similar atomic force microscopy (AFM) images of the EML before and after the ion beam exposure. On the other hand, an ion beam treatment might affect the surface chemical properties of EML. This can be confirmed in other papers [11–14]. Consequently, the surface chemical modulation by an ion beam treatment changes only the wetting property of the EML.

4. Conclusions

The light emitting uniformity of TEPLED with an invert structure could be enhanced by an ion beam treatment, which could improve the surface adhesion between the EML and HTL. Moreover, the luminance was improved using PMMA as a buffer layer, which protects the HTL from ITO deposition by sputtering. In conclusion, a uniform TEPLED with enhanced luminance was demonstrated.

Acknowledgements – This study was supported by the Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Science, ICT and Future Planning (No. 2013R1A1A1A05006783) and the Human Resources Development Program (No. 20124030200100) of the Korea Institute of Energy Technology Evaluation and Planning (KETEP) grant funded by the Korea Government Ministry of Trade, Industry and Energy.

References

[1] TANG C.W., VANSLYKE S.A., *Organic electroluminescent diodes*, Applied Physics Letters **51**(12), 1987, pp. 913–915.

[2] HOFMANN S., THOMSCHKE M., LÜSSEM B., LEO K., *Top-emitting organic light-emitting diodes*, Optics Express **19**(S6), 2011, pp. A1250–A1264.

[3] GU G., BULOVIĆ V., BURROWS P.E., FORREST S.R., THOMPSON M.E., *Transparent organic light emitting devices*, Applied Physics Letters **68**(19), 1996, pp. 2606–2608.

[4] HUNG L.S., TANG C.W., *Interface engineering in preparation of organic surface-emitting diodes*, Applied Physics Letters **74**(21), 1999, pp. 3209–3211.

[5] HUNG L.S., TANG C.W., MASON M.G., RAYCHAUDHURI P., MADATHIL J., *Application of an ultrathin LiF/Al bilayer in organic surface-emitting diodes*, Applied Physics Letters **78**(4), 2001, pp. 544–546.

[6] LU M.-H., WEAVER M.S., ZHOU T.X., ROTHMAN M., KWONG R.C., HACK M., BROWN J.J., *High-efficiency top-emitting organic light-emitting devices*, Applied Physics Letters **81**(21), 2002, pp. 3921–3923.

[7] DENG X.Y., HO M.K., WONG K.Y., *Top-emitting polymer light-emitting diodes with environmentally stable cathodes*, Journal of Applied Physics **99**(1), 2006, article 016103.

[8] BRAUN D., HEEGER A.J., *Visible light emission from semiconducting polymer diodes*, Applied Physics Letters **58**(18), 1991, pp. 1982–1984.

[9] HUNG L.S., TANG C.W., MASON M.G., *Enhanced electron injection in organic electroluminescence devices using an Al/LiF electrode*, Applied Physics Letters **70**(2), 1997, pp. 152–154.

[10] Hyesung Park, Rowehl J.A., Ki Kang Kim, Bulovic V., Jing Kong, *Doped graphene electrodes for organic solar cells*, Nanotechnology **21**(50), 2010, article 505204.

[11] Jin Seog Gwag, Chul Gyu Jhun, Jae Chang Kim, Tae-Hoon Yoon, Gi-Dong Lee, Seong Jin Cho, *Alignment of liquid crystal on a polyimide surface exposed to an Ar ion beam*, Journal of Applied Physics **96**(1), 2004, pp. 257–260.

[12] Jin Seog Gwag, Kyoung-Ho Park, Dong Jin Kang, Chul Gyu Jhun, Hee Kim, Seong Jin Cho, Tae-Hoon Yoon, Jae Chang Kim, *Polyimide surface bombarded with Ar atomic beam*, Japanese Journal of Applied Physics **42**(Part 2, No. 5A), 2003, pp. L468–L471.

[13] Phil Kook Son, Seung Hun Yu, Jonghoon Yi, Jin Seog Gwag, *Electro-optical characteristics of two domain normally black-electrically controlled birefringence mode*, Journal of Applied Physics **114**(6), 2013, article 064506.

[14] Chaudhari P., Lacey J., Doyle J., Galligan E., Lien S.A., Callegari A., Hougham G., Lang N.D., Andry P.S., John R., Yang K., Lu M., Cai C., Speidell J., Purushothaman S., Ritsko J., Samant M., Stohr J., Nakagawa Y., Katoh Y., Saitoh Y., Sakai K., Satoh H., Odahara S., Nakano H., Nakagaki J., Shiota Y., *Atomic-beam alignment of inorganic materials for liquid-crystal displays*, Nature (London) **411**(6833), 2001, pp. 56–59.

Received April 4, 2014