Optimal Structure of Color-Conversion Layer for White Organic Light-Emitting Diode on Silver-Nanowire Anode

Bong Sung Kim, Min Ki Kim, Deok Su Jo, Heeyeop Chae, and Sung Min Cho

SKKU Advanced Institute of Nanotechnology (SAINT), Sungkyunkwan University, Suwon 440-746, Korea
School of Chemical Engineering, Sungkyunkwan University, Suwon 440-746, Korea
School of Advanced Materials Science and Engineering, Sungkyunkwan University, Suwon 440-746, Korea

We fabricated fluorescent blue organic light-emitting diodes (OLEDs) using a silver-nanowire (AgNW) anode on a polyethylene terephthalate substrate. Flexible white OLEDs with high color rendering index (CRI) were successfully obtained through color conversion of the blue OLEDs. Using a single layer of color converting phosphor particles, we showed that the variation of CRI with viewing angle depended on the path length of light through the color conversion layer (CCL). We proposed a method to obtain an optimal number of CCL to maximize the CRI value of down-converted white OLEDs. The color conversion from blue to white using CCLs was compared for the OLEDs on AgNW and conventional indium-tin oxide (ITO) anodes. Due to the light scattering effect of AgNW, the OLED on AgNW electrode showed higher device efficiencies and lower CRI dependence on viewing angle than that on ITO anode.

© The Author(s) 2017. Published by ECS. This is an open access article distributed under the terms of the Creative Commons Attribution 4.0 License (CC BY, http://creativecommons.org/licenses/by/4.0/), which permits unrestricted reuse of the work in any medium, provided the original work is properly cited. [DOI: 10.1149/2.022180jss] All rights reserved.

White organic light-emitting diodes (OLEDs) have been commercialized for solid-state lighting applications and are now competing with another solid-state light source using inorganic light-emitting diodes (LEDs). The OLED lighting has a few advantages over the LED lighting such as a lower heat generation, lower glare, large-area and flexible light source, high production costs of the OLED make it difficult to enter the general lighting market. For this reason, a method of lowering the cost of OLED lighting is required. It is well known that there are two different methods to produce white OLEDs. The most commonly used method is to apply a tandem structure for the white OLEDs. The tandem OLED is a device that connects two or more OLED lighting units with one or more charge-generation layers, and allows the light-emitting units to emit simultaneously. Tandem white OLEDs can be realized by mixing different colors of the light-emitting units. Another method to produce white OLEDs is to use color changing materials with blue OLEDs for converting the blue to white emission. Currently, the second method is not commonly utilized because highly stable and efficient blue-emitting materials are not available, even though the method can potentially reduce the production cost of white OLEDs by utilizing simpler device structure and cheaper materials. To accelerate the market entry of the OLED lighting, considerable efforts are being made to develop blue emitting materials having high efficiency and long lifetime. In this regard, we focus on the second method to develop white OLEDs with a high color rendering index (CRI) in this study.

The most commonly used transparent electrode for OLEDs is indium tin oxide (ITO). Because the ITO is intrinsically rigid, it is difficult to use it as a transparent electrode of flexible OLEDs. Silver nanowire (AgNW) attracts much attention recently as a promising transparent electrode material to replace ITO. Apart from the effect of AgNW, the OLED on AgNW electrode showed higher device efficiencies and lower CRI dependence on viewing angle than that on ITO anode.

© The Author(s) 2017. Published by ECS. This is an open access article distributed under the terms of the Creative Commons Attribution 4.0 License (CC BY, http://creativecommons.org/licenses/by/4.0/), which permits unrestricted reuse of the work in any medium, provided the original work is properly cited. [DOI: 10.1149/2.022180jss] All rights reserved.

Experimental

Glass and PET films were cleaned thoroughly and dried with pure nitrogen. The glass substrate of 7 cm × 7 cm size was treated with octadecyl trichlorosilane in toluene for 24 h to form a self-assembled monolayer (SAM) on the surface. The hydrophobic SAM-covered surface facilitates the transfer of AgNW embedded in a hydrophilic UV resin onto a PET substrate. A solution of AgNPs (0.5 wt%, IPA based, purchased from Nanopyxis) was coated on the SAM-covered glass substrate by the Meyer rod method (rod No. 16), and then the layer was subsequently dried at 110°C. Separately, a poly-acrylic acid (PAA) layer was formed on the cleaned PET film surface using a bar coater and the layer was cured at 60°C for 3 min. The PAA layer was utilized as an optically clear adhesive. A green-yellow Sr2SiO4:Eu2+-bis-(1-naphyl)-N,N′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB) phosphor was evenly spread on the adherent PAA surface. Non-bonded phosphor particles to the PAA surface were removed by nitrogen blowing in order to obtain a single phosphor layer on the PET film. The repetition of the same process can produce multiple layers of the phosphor.

The separately prepared AgNPs on the SAM-coated glass and phosphor-coated PET were then laminated with an ultraviolet (UV) curable resin (NOA61, purchased from Norland) using a hand roller (Fig. 1b). Subsequently after the UV resin was cured for 150 sec under UV light, the PET film was peeled off the SAM-coated glass to have the AgNPs in the embedded UV resin on one side and the monolayer of phosphor on the other side of the PET film (Fig. 1c). The embedded AgNW electrode was found to have the optical transmittance, root-mean-square surface roughness, and sheet resistance of 84%, 3 nm, and 6.4 ohm/square, respectively. The AgNW electrode surface was patterned with an electrically insulating paste by the screen printing and finally a fluorescent blue OLED was deposited in vacuum on the patterned AgNW electrode (Figs. 1d and 1e). We fabricated blue-emitting fluorescent OLEDs, of which the emission spectrum has the peak wavelength at 460 nm. Organic materials and cathode metal were evaporated under 8 × 10−7 torr: 1,4,5,8,9,11-Hexazaatropine-hexa-carbonitrile (HAT-CN) and N,N′-bis-(1-naphyl)-N,N′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB) were used.
utilized as the hole injection layer and hole transport layer, respectively. 4,4′,4″-Tri(N-carbazolyl)triphenylamine (TCTA) was utilized as the electron blocking layer. For blue fluorescent emission, 2-methyl-9,10-di-(2-naphthyl)anthracene (MADV) and 1,6-bis(N-phenyl-p-CN-phenylamino)-pyrenes (Pyrene-CN) were utilized as the host and dopant materials, respectively. To complete blue OLED devices, electron transporting LG201 (LG Chem. Ltd) doped with lithium quinolate (Liq), electron injecting Liq, and aluminum (Al) cathode were successively deposited. The blue emitting OLED structure was HAT-CN (35 nm)/NPB (80 nm)/TCTA (20 nm)/MADV:Pyrene-CN (5 wt%, 20 nm)/LG201-Liq (50%, 30 nm)/LiQ (1.5 nm)/Al (100 nm). Figure 1f shows the flexible white OLED on AgNW electrode fabricated in this study.

The fabricated OLEDs were encapsulated with a glass for the device-performance measurement. For comparison, the same OLED device on a commercial ITO-coated glass was also prepared. The phosphor layer was coated after the glass encapsulation for the glass-based OLED to protect the device from ambient moisture and oxygen. The OLED performance was measured using a spectro-radiometer (CS-2000, Konika Minolta, Japan) and source meter (Keithley 2400, Tektronix, USA). The external quantum efficiency (EQE) was determined by a calibrated goniometer equipped with the spectro-radiometer. Light distribution was measured to obtain the angular dependence of the luminance (ENC technology, Korea). The color rendering index (CRI, R1-R14) was calculated using the emission spectra of the OLED devices.

**Results and Discussion**

Figure 2 shows the electroluminescence (EL) spectrum of a blue OLED along with the absorption and photoluminescence spectra of Sr₂SiO₄:Eu²⁺ phosphor. The blue EL spectrum has a main peak at 460 nm wavelength with a small shoulder peak at 480 nm wavelength. The full-width-half-maximum (FWHM) of the spectrum was 60 nm. As shown in the figure, the EL spectrum overlaps well the absorption spectrum of the Sr₂SiO₄:Eu²⁺ phosphor in the range from 425 to 530 nm. The green-yellow emission from the excited Sr₂SiO₄:Eu²⁺ phosphors was centered at 570 nm and showed the FWHM of 90 nm. The extent of color conversion can be controlled by the amount of the phosphor, and the CRI of white OLED is determined by an appropriate balance between the converted green-yellow emission and the blue EL emission which has not been converted. The amount of the phosphor should be precisely controlled in order to achieve a high CRI white emission. To achieve this goal easily, we prepared a single phosphor layer and the total amount of the phosphor was adjusted by controlling the number of layers.

Figure 3 shows the performance of OLEDs fabricated on ITO and AgNW electrodes with and without the CCL. The CCL used in this experiment was a single phosphor layer. Both OLEDs on ITO and AgNW electrodes were turned on at 2.8 V as shown in the figure. There was higher leakage current for the OLED on AgNW electrode than that on ITO electrode because of a higher surface roughness of AgNW electrode. The current injection was lower at low operating voltages for the OLED on AgNW electrode, but it increased as the voltage increased and became comparable to that on ITO electrode. As shown in Fig. 3a, the OLED on AgNW electrode showed higher luminance than that on ITO electrode at similar current densities. As expected from this observation, the power, current, and external

---

**Figure 1.** Schematic fabrication process of flexible white OLED on AgNW electrode and a photograph of white emitting OLED.

**Figure 2.** Normalized EL spectrum of a blue OLED and absorption/photoluminescence spectra of the Sr₂SiO₄:Eu²⁺ phosphor.
quantum efficiencies were found higher by around 25% for OLED on the AgNW electrode (Figs 2b and 2c). The blue emission spectra from both OLEDs on the AgNW and ITO electrodes were similar as shown in Fig. 2d, although there was a little difference due to the micro-cavity effect caused by the structure difference of the electrodes. The higher efficiencies of the OLED on AgNW electrode can be explained by the enhanced light extraction due to the light scattering by the AgNWs. With the color conversion layer (CCL), the luminance and efficiencies increased by 100% and 50%, respectively. The increases can be explained by the spectrum change and enhanced light extraction due to scattering when the CCL is utilized. After the color conversion, the efficiency increases occur because of the light intensity increases at 550 nm that has the highest sensitivity of the human eye. At the same time, the light generated in the OLED is scattered with the random phosphor particles as it exits out of the OLED, so that more light is extracted than in the case of no phosphor. As shown in the Table 1, the OLED efficiencies with AgNW electrode are higher by 25% than that with ITO electrode. The device efficiencies then increased by about 50% with applying a CCL, regardless of the electrodes. It means that the amount of the color conversion by a single phosphor layer is constant and the final efficiency of white emission after the conversion depends only on the blue-emission efficiency. In this regard, the blue OLED on the AgNW electrode is better for the color conversion into white emission than that on the ITO electrode.

Figure 4 shows the angular dependence of the emission from OLED on the AgNW and ITO electrodes before and after the color conversion by the CCL. The blue emission spectrum of the OLED on ITO electrode changes as the viewing angle changes while that on AgNW electrode is almost stationary. There exists a weak micro-cavity in the OLED with ITO electrode, which is responsible for the angular dependence of the emission. The measured reflectance of ITO electrode depends on the wavelength and was about 20% at 500 nm wavelength. On the other hand, the micro-cavity effect affects little for the OLED on the AgNW electrode because of the light scattering with AgNWs or low light reflectance (less than 10% in full wavelength region) of the embedded AgNW electrode. Measured haze of AgNW was about 7%. With a single CCL, the blue emission is converted into green-yellow color and the extent of the color conversion changes significantly at different viewing angles. As shown in Figs. 4c and 4d, the stronger color conversion occurs at higher viewing angles. However, the changes in the color conversion are relatively smaller for the OLED on AgNW electrode than that on ITO electrode.

We measured CRI at different viewing angles for the OLEDs on ITO and AgNW electrodes after the color conversion, as shown in Fig. 5. As the viewing angle increases, the CRI increases for the OLEDs on the two different electrodes until it reaches 60°. It is because the color conversion becomes stronger as the viewing angle increases. Past the CRI maximum at 60°, the CRI starts to decrease for both cases. The decrease in CRI is attributed to the fact that the white balance begins to
Table I. Performance of OLEDs on ITO and AgNW electrodes with and without CCL.

| Device          | Turn-on Voltage (V) | Maximum efficiency | Efficiency at 100 cd/m² | Efficiency at 1,000 cd/m² |
|-----------------|---------------------|-------------------|-------------------------|--------------------------|
|                 |                     | cd/A   | lm/W   | cd/A   | lm/W   | cd/A   | lm/W   |
| ITO             | 2.75                | 9.1    | 8.1    | 8.4    | 7.9    | 9      | 6.9    |
| ITO + CCL       | 14.2                | 12.1   | 12.5   | 12.1   | 13.6   | 11.4   |
| AgNW            | 12.5                | 9.4    | 11.2   | 9.4    | 11.4   | 8.2    |
| AgNW + CCL      | 18.1                | 15.4   | 17.3   | 14.2   | 17.8   | 14.7   |

deteriorate as the green-yellow portion becomes too strong. It should be noted here that a single CCL results in higher CRI at the viewing direction normal to the OLED surface and lower CRI at the maximum for the OLED on the AgNW electrode than that on ITO electrode. It is because the angular dependence becomes less sensitive due to the strong light scattering of AgNWs.

When the viewing angle increases, the light path is laid down with respect to the surface normal direction (0°), and the color conversion becomes large because the length passing through the CCL becomes long. At a viewing angle of 60°, at which the CRI becomes maximum, the path length through which the light travels through the CCL is twice compared with the normal direction. Past this angle, the color conversion becomes too strong and the CRI begins to decrease. We shows the dependence of the light path length on the viewing angle in Fig. 6. The light path length increases in proportion to the inverse of the cosine of the viewing angle. Figure 6 compares the extent of color conversion obtained experimentally with the calculated light path length. The extent of color conversion at a certain viewing angle is the ratio of the maximum peak intensity at 573 nm wavelength at the angle to that at 0°, as shown in Figs. 4c and 4d. The experimentally calculated extents of color conversion fit well to the estimated light path length when the viewing angle is less than 60°. The error at larger viewing angles is expected to be inevitable since the CCL is not a continuous layer and composed of single-layered particles.

In order to check the aforementioned discussion, we fabricated two layers of CCL and showed the color conversion at different viewing angles.

Figure 4. Emission spectra of the OLEDs at different viewing angles from 0° to 80°: (a) blue OLED on ITO electrode, (b) blue OLED on AgNW electrode, (c) blue OLED with CCL on ITO electrode, and (d) blue OLED with CCL on AgNW electrode.
angles for the OLED on AgNW electrode in Fig. 7a. At a viewing angle of $0^\circ$, the light path length for the double CCL case is twice that for the single CCL case. Therefore, the maximum CRI was obtained at a viewing angle of $0^\circ$ in the case of two layers of CCLs, just as the CRI was maximized at a viewing angle of $60^\circ$ at which the light path length doubled. And the maximum CRI value of 94.3 was the same for the single and double CCL cases. For the double CCL case, the CRI decreases continuously as the viewing angle increases because the color conversion becomes too strong past the optimum white balance.

Scanning electron microscopic images are shown in the insets of Fig. 7a. Figure 7b shows the changes in the CIE (International Commission on Illumination) coordinate as the viewing angle increases for the ITO and AgNW electrodes with CCLs. For the 1 layer of CCL, the highest CRI was obtained at around the viewing angle of $60^\circ$. In case of the 2 layers of CCL, however, the coordinate is moving away from the white region as the viewing angle increases from $0^\circ$ to $80^\circ$. From the above discussion, it can be concluded that the maximum CRI is obtained at the 2 layers of CCL, and the CRI is reduced when there are more CCL layers. The method of optimizing the extent of color conversion by controlling the number of monolayer CCL is useful not only for maximizing the CRI of down-converted white OLEDs but also for minimizing the consumption of the phosphor. Starting with an usual fluorescent blue OLED on ITO/glass substrate with a power efficiency of 6.9 lm/W at 1,000 cd/m² luminance, the application of the AgNW electrode and optimized CCL to the blue OLED enhances the efficiency by more than twice up to 14.7 lm/W at 1,000 cd/m² luminance with a high CRI of 94.3. It is expected that the down-converted white OLED with higher efficiency and CRI is possible if fluorescent, phosphorescent, or thermally activated delayed fluorescent blue emitting materials with the better performance is available.

**Conclusions**

The blue OLED on AgNW/PET electrode was found superior to that on ITO/glass electrode because of the effective light scattering of AgNW. The utilization of AgNW as the anode for OLEDs does not only enhance the efficiency of OLED but also reduce the variation of CRI according to the viewing angle. As the viewing angle increases, the path of the light through CCL becomes longer and thus the extent of color conversion becomes larger. Thus, the optimal structure of CCL can be obtained to maximize the CRI of OLEDs after color conversion. In this study, the maximum CRI of 95 and 94.3 was achieved for OLEDs on ITO and AgNW anodes, respectively. The maximum CRI values were at the viewing angle of $60^\circ$ for one layer of CCL and at the normal viewing angle for two layers of CCL. It is because the light path length at the viewing angle of $60^\circ$ is twice longer than that at the surface normal viewing angle for one layer of
CCL and is the same as that at the normal viewing angle for two layers of CCL. Addition of more layers of CCL reduces the CRI by changing the emission spectrum away from the optimum white balance.

Acknowledgments

This work was supported by the MSIP (Ministry of Science, ICT and Future Planning) under the ITRC (Information Technology Research Center) support program (IITP-2016-H8501-16-1009) supervised by the IITP (Institute for Information & communications Technology Promotion), Korea.

References

1. S. Reineke, M. Thomschke, B. Lüsem, and K. Leo, Rev. Mod. Phys., 85, 1245 (2013).
2. M. C. Gather, A. Köhnen, and K. Meerholz, Adv. Mater., 23, 233 (2011).
3. T. Komoda, N. Ide, K. Varutt, K. Yamae, H. Tsuji, and Y. Matsuhisa, J. Soc. Inf. Disp., 19, 838 (2011).
4. J. Y. Oh, J. H. Kim, Y. K. Seo, C. W. Joo, J. Lee, J.-I. Lee, S. Yu, C. Yun, M. H. Kang, B. H. Choi, and Y. H. Kim, Dyes Pigm., 136, 92 (2017).
5. S. Reineke, F. Lindner, G. Schwartz, N. Seidler, K. Walzer, B. Lüsem, and K. Leo, Nature, 459, 234 (2009).
6. Y. H. Kim, J. Lee, W. M. Kim, C. Fuchs, S. Hofmann, H.-W. Chang, M. C. Gather, L. Müller-Meskamp, and K. Leo, Adv. Funct. Mater., 24, 2553 (2014).
7. T.-W. Koh, H. Cho, C. Yun, and S. Yoo, Org. Electron., 13, 3145 (2012).
8. J. Lee, N. Chopra, D. Bera, S. Maslov, S. H. Eom, Y. Zheng, P. Holloway, J. Xue, and F. So, Adv. Energy Mater., 1, 174 (2011).
9. S.-H. Lee, D. S. Jo, B. S. Kim, D.-H. Yoon, H. Chae, H.-K. Chung, and S. M. Cho, Curr. Appl. Phys., 17, 1108 (2017).
10. X.-Y. Zeng, Q.-K. Zhag, R.-M. Yu, and C.-Z. Lu, Adv. Mater., 22, 4484 (2010).
11. X. Guo, X. Liu, J. Luo, Z. Gan, Z. Meng, and N. Zhang, RSC Adv., 5, 24953 (2015).
12. Y. Jiang, J. Ji, Z. Wu, H. Dong, Z. Zhao, B. Jiao, and X. Hou, Langmuir, 31, 4950 (2015).
13. K.-H. Ok, J. Kim, S.-R. Park, Y. Kim, C.-J. Lee, S.-J. Hong, M.-G. Kwak, N. Kim, C. J. Han, and J.-W. Kim, Sci. Rep., 5, 9464 (2015).
14. H.-F. Cui, Y.-F. Zhang, and C.-N. Li, Opt. Eng., 53, 077102 (2014).
15. L. Li, J. Liang, S.-Y. Chou, X. Zhu, X. Niu, Z. Yu, and Q. Pei, Sci. Rep., 4, 4307 (2014).
16. S. Nam, M. Song, D.-H. Kim, B. Cho, H. M. Lee, J.-D. Kwon, S.-G. Park, K.-S. Nam, Y. Jeong, S.-H. Kwon, Y. C. Park, S.-H. Jin, J.-W. Kang, S. Jo, and C. S. Kim, Sci. Rep., 4, 4788 (2014).
17. E. Jung, C. Kim, M. Kim, H. Chae, J. H. Cho, and S. M. Cho, Org. Electron., 41, 190 (2017).