PROCEEDINGS OF THE INTERNATIONAL DISPLAY WORKSHOPS, VOL.29, 2022

Development of Air Stable Inverted Organic Light-Emitting Diodes and Application to Thin Film Light Sources

Kenji Kuwada[1], **Naoki Kobayashi**[1], **Tsuyoshi Goya**[1] **and Katsuyuki Morii**[1,2]

kenji_kuwada@shokubai.co.jp
[1] NIPPON SHOKUBAI CO., LTD. 5-8 Nishi Otabi-cho, Suita, Osaka 564-0034, Japan
[2]Nippon Shokubai Research Alliance Laboratories, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565–0871, Japan
Keywords: Inverted OLED, Light Source, Air Stable, Flexible OLED

ABSTRACT

We have developed extremely air-stable inverted Organic light-emitting diodes. Our developed organic light-emitting diode, which can be with low barrier performance film, made highly flexible film light sources possible. Possibilities of new light created by highly flexible film light sources will be presented.

1 Introduction

Organic light-emitting diodes (OLEDs) can be applied to flexible devices, and various flexible OLEDs have been developed in recent years, including foldable phones, rollable TV, and so on. However, OLEDs require very robust encapsulation because the electron injection layer and cathode are inherently unstable to the air [1]. Since the encapsulation process requires the introduction of expensive equipment for multiple alternating layers of organic-inorganic barrier layers and a lot of accumulated know-how, there are currently very few applications other than conventional displays. Since expensive encapsulation equipment cannot be installed in lighting and light source applications, it is difficult to encapsulate except for using a barrier film. In general, OLEDs require a water vapor transmission rate (WVTR) of about 10^{-6} g/m^2/day [2], but this is not easy to achieve, especially when the film substrate is thin. As a result, only thick barrier films with low flexibility can be used, and the flexibility expected of OLEDs cannot be archived, so the flexible film light sources market has hardly been formed.

In such a background, inverted OLEDs, which do not use highly reactive materials such as alkali metals are inherently stable in the air, are attracting attention [3]. However, electron injection from electrodes such as ITO is difficult due to the large energy gap, resulting in very high operating voltage and short operating life time for inverted OLEDs. We found that a combination of a ZnO layer as an inorganic electron injection layer, and a combination of a boron-based organic semiconductor and an organic base compound as an organic electron injection layer, was effective in solving these problems [4] and we call our inverted OLEDs as iOLED™. In 2019, hydrogen-bonded organic compounds [5] and in 2020, coordination-bonded organic compounds [6] were found to produce even higher performance. And they were comparable operating voltage and operating life time compared to conventional OLEDs. Taking advantage of the high air stability of our latest iOLED™, it has made possible to fabricate OLEDs on thin and soft film substrate with low-barrier performance. This has resulted in a truly thin and flexible film light source that the market is demanding. Film light sources enable new and unprecedented expressions and uses of light, with unlimited possibilities. Here, we report on our developed highly flexible film light sources, its characteristics, and our efforts to develop the film light sources market.

2 Structure of iOLED™ film light source

2.1 The device structure of iOLED™

The iOLED™ were developed on a glass substrate coated with a 150-nm-thick ITO layer. Our original layers, inorganic electron injection layer and organic electron injection layer, were deposited on ITO. Figure 1 shows the structure of (a) a conventional OLED and (b) an iOLED™.

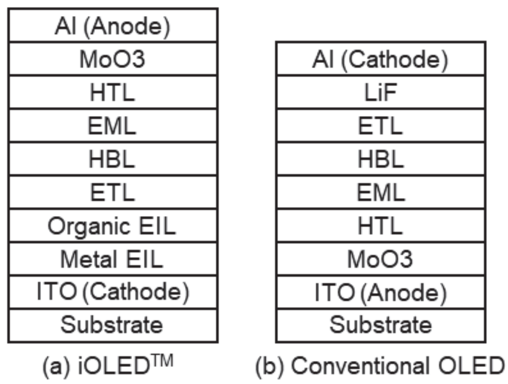

Fig.1 Device structures of iOLED™ and OLED

2.2 Fabrication of iOLED™ film light sources

Barrier film with a thickness of 25 μm and a WVTR of 3×10^{-3} was used as the substrate. Barrier films could not be physically cleaned with a brush, and ultrasonic cleaning is ineffective, making it very difficult to clean the surface. Therefore, we chose to apply a curable polymer on film as the planarization layer instead of cleaning. The planarization layer was coated on the barrier film attached to glass with a weak adhesive film. Several functional layers were deposited on the planarization layer. Finally, a barrier film with an adhesive film was

ISSN-L 1883-2490/29/0460 © 2022 ITE and SID

laminated on top of the functional layers. Then film light source was removed from the glass. The total thickness of the iOLED™ film light source was less than 100 µm.

3 Device performance of iOLED™ film light sources

3.1 Air stability of iOLED™

We fabricated the OLED and the iOLED™ on a glass substrate without encapsulation to check the air stability. They were stored in air under room temperature, The electroluminescence (EL) images were shown in Figure 2.

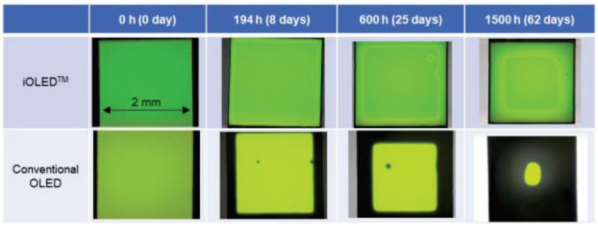

Fig.2 Comparison of EL images between iOLED™ and OLED exposed to the air

While the conventional OLED showed a significant decrease in emitting area, the iOLED™ showed virtually no change in emitting area after being exposed to the air for more than two months. Quite high air stable film light source has been realized by our developed iOLED™.

3.2 Characteristics of iOLED™ film light sources

EL characteristics of the iOLED™ film light sources are shown in Table 1. Even though there is no structure to increase the extraction efficiency, it shows high external quantum efficiency, and the operating life time of LT50 at initial luminance of 1,000 cd/m^2, is also at a level where there are no practical problems.

Color	External quantum efficiency	Operating life time
Red	~ 30 %	> 100,000 h
Green	~ 28 %	> 100,000 h
Blue	~ 6 %	> 5,000 h

Table.1 Characteristics of iOLED™ film light sources.

The luminance uniformity of the iOLED™ film light sources were shown in Figure 3. Although it depends on the current density, the uniformity (the difference between the brightest and darkest spots) is within 10 % even at high current density over 10mA/cm^2.

The iOLED™ film light sources were found to exhibit very high flexibility. The EL characteristics do not change before and after 200,000 bending tests, conducted at a bending radius of 2 mm in-fold and out-fold, respectively.

In order to evaluate a storage stability, the film devices were stored in an 85°C 85%Rh environment for 100 hours. It was confirmed that there were no dark spot or shrinkage of the emitting area and no change in the current-voltage characteristics.

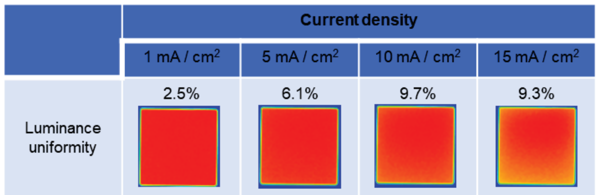

Fig.3 Current density dependence of the luminance uniformity

4 The Market development of the film light sources

4.1 Demonstration in exhibitions

OLED is not well known for applications other than displays, and although thin and flexible surface light sources have been commercialized by several companies, their existence is not well known. We believe that film light sources can form a new market because they are thin and flexible, allowing for uniform illumination in confined spaces and on curved surfaces where LEDs cannot be installed. Therefore, we exhibited at an exhibition to first make people aware of film light sources.

In 2019, we exhibited at an exhibition to demonstrate the thinness and flexibility of the product by using a fan to sway in the wind, as well as producing and showing an image video proposing various possibilities as a new light source that can be worn on clothing or skin. More than 400 companies from a variety of industries, including the car, the arts, the fashion, and the consumer electronics industry showed interest in iOLED™ film light sources, which convinced us of the need for film light sources. And they also provided us an opportunity to realize once again that the world didn’t know about film light sources.

After that, we collaborated between iOLED™ as a technology and traditional art in order to open new field. The following year, we collaborated with the traditional Japanese lacquerware, Wajima lacquerware. iOLED™ film light sources mounted on Wajima lacquerware was displayed at exhibition. The thinness of the iOLED™ film light source, combined with the technique of “Raden”, was able to create a new appeal for the coloring of “Raden”. And the following year, we collaborated with the traditional Japanese textile, Nishijin-Ori. We develop string-shaped iOLED™ film light sources and successfully incorporated it into Nishijin-Ori. Just as copper wire is more flexible than copper foil, thinness as well as fineness are important factor in flexibility. However, when fineness is pursued, storage stability cannot be ensured due to air infiltration from the side in case of conventional OLED, but the air stability of iOLED™ made this possible. Thin and elongated light sources have never been seen before, and the string-shaped iOLED™ film light sources will provide a new value. We believe we were able to propose new possibilities for film light sources.

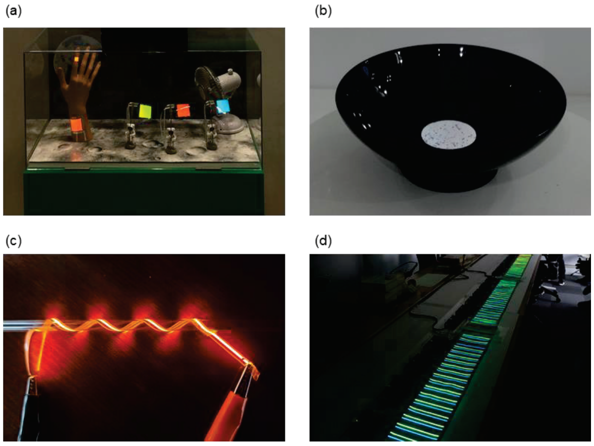

Fig.5 Exhibition samples.
(a) First demonstration (b) Embedded in Wajima lacquerware. (c) String-shaped iOLED^{TM} film light source wrapped around a 5mm diameter tube. (d) Embedded in Nishijin-Ori.

4.2 Medical applications

One of the reactions of the exhibition is an application to the medical field such as phototherapy. Since low level light irradiation (LLLI) to the cells has been shown to enhance cell proliferation [7], and light-based medicine have the advantages of being noninvasive, selective and safe, light-based medicine and cosmetics have been attracting attention in recent years. Especially wearable medical devices are attractive next-generation electronic devices because they can provide medical effects regardless of location or time and ultimately reduce the price of healthcare [8]. In addition to treatment, there are also attempts to use light to acquire biological information such as pulse wave [9] and blood oxygen levels [10], etc. Because LEDs are heavy, inflexible, non-uniform light, and poor adhesion to living bodies, OLEDs are being increasingly adopted, but encapsulation is extremely difficult and impractical. An iOLED^{TM} film light source with high air stability could solve these problems and boost the development of light-based wearable devices.

We have developed a technology to incorporate iOLED^{TM} and organic photo diode on a 1.5 μm ultra-flexible substrate in collaboration with Prof. Yokota, and succeeded in measuring blood pressure with high accuracy for long time [11]. Generally, integrating different optical devices on the same ultra-flexible substrate is difficult owing to the instability of the devices. In this study, we succeeded in improving the air stability of OLEDs and organic photodiodes by using an inverted structure, enabling the mounting of different optical elements on an ultra-flexible substrate. Using an ultra-flexible sensor, it will be possible to continuously measure blood pressure in daily life; therefore, these sensors can be applied in the medical field for monitoring health conditions and early detection of diseases.

5 Conclusion

We have developed an inverted OLED with a high air stability and a long operating life time and applied it to film light sources. We have shown the potential of film light sources by proposing their use as string light sources and sensor light sources. Currently we are receiving suggestions from various users on how to use our film light sources, and our mass production line is also getting ready. The film light sources market is still underdeveloped, but we are confident that we will be able to develop it soon. The performance of iOLED^{TM} is comparable to that of conventional OLEDs, we believe that it will contribute to making flexible displays at a lower cost in near future.

Acknowledgements

This work was supported by the results of joint research with NHK Science & Technology Research Laboratories. The authors thank Hirohiko Fukagawa (NHK Science & Technology Research Laboratories) for his help in designing EIL molecular and the device.

References

[1] J. McElvain, H. Antoniadis, M. R. Hueschen, J. N. Miller, D. M. Roitman, J. R. Sheats, R. L. Moon, H, “Formation and growth of black spots in organic light - emitting diodes,” J. Appl. Phys. Vol. 80, p. 6002 (1996).

[2] C.Charton, N.Schiller, M.Fahland, A.Holländer, A.Wedel, K.Noller, “Development of high barrier films on flexible polymer substrates,” Thin Solid Films. Vol. 502, p. 99 (2006).

[3] K. Morii, M. Ishida, T. Takashima, T. Shimoda, Q. Wang, M. K. Nazeeruddin, M. Grätzel, “Encapsulation-free hybrid organic-inorganic light-emitting diodes,” Appl. Phys. Lett. Vol. 89, p. 183510 (2006).

[4] H. Fukagawa, T. Sasaki, T. Tsuzuki, Y. Nakajima, T. Takei, G. Motomura, M. Hasegawa, K. Morii, T. Shimizu, “Long-Lived Flexible Displays Employing Efficient and Stable Inverted Organic Light-Emitting Diodes,” Adv. Mater. Vol. 30, p. 1706768 (2018).

[5] H. Fukagawa, M. Hasegawa, K. Morii, K. Suzuki, T. Sasaki, T. Shimizu, “Universal Strategy for Efficient Electron Injection into Organic Semiconductors Utilizing Hydrogen Bonds,” Adv. Mater. Vol. 31, p.1904201(2019).

[6] H. Fukagawa, K. Suzuki, H. Ito, K. Inagaki, T. Sasaki, T. Oono, M. Hasegawa, K. Morii, T. Shimizu, “Understanding coordination reaction for producing stable electrode with various low work functions,” Nat. Commun. Vol. 11, p. 3700 (2020).

[7] W.T.Li, Y.C.Leu, “Effects of Low Level Red-light Irradiation on the Proliferation of Mesenchymal Stem Cells Derived from Rat Bone Marrow,” Proc.

IEEE EMBS ’29, p. 5829 (2007).
[8] Y.Jeon, H.R.Choi, M.Lim, S.Choi, H.Kim, J.H.Kwon, K.C.Park, K.C.Choi, “A Wearable Photobiomodulation Patch Using a Flexible Red-Wavelength OLED and Its In Vitro Differential Cell Proliferation Effects,” Adv.Mater.Technol. Vol.3, p. 1700391 (2018).
[9] Y. Lee, J. W. Chung, G. H. Lee, H. Kang, J. Y. Kim, C. Bae, H. Yoo, S. Jeong, H. Cho, S. G. Kang, J. Y. Jung, D. W. Lee, S. Gam, S. G. Hahm, Y. Kuzumoto, S. J. Kim, Z. Bao, Y. Hong, Y. Yun, S. Kim, “Standalone real-time health monitoring patch based on a stretchable organic optoelectronic system,” Sci. Adv. Vol. 7, eabg9180 (2021).
[10] H. Lee, E. Kim, Y. Lee, H. Kim, J. Lee, M. Kim, H. J. Yoo, S. Yoo, “Toward all-day wearable health monitoring: An ultralow-power, reflective organic pulse oximetry sensing patch,” Sci.Adv. Vol. 4, eaas9530 (2018).
[11] T. Yokota, I. Shirayama, K. Kuwada, M. Koizumi, W. Yukita, K. Morii, H. Fukagawa, T. Shimizu, K. Fukuda, T. Someya, “Air-Stable Ultra-Flexible Organic Photonic System for Cardiovascular Monitoring,” Adv. Mater. Technol, p.2200454 (2022).