Powder Electroluminescent Device with Flexible Invisible Silver-Grid Transparent Electrode

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ABSTRACT

Powder electroluminescent devices (PELDs) have high potential of commercial applications because of their flat light emission and printing processes. In this study, we develop PELDs on gravure offset printed invisible Ag-grid laminated with PEDOT:PSS transparent electrodes. The PELDs with the invisible Ag-grid transparent electrodes showed excellent electroluminescent properties.

1 INTRODUCTION

Inorganic electroluminescence is defined as the emission of light from inorganic materials such as ZnS-type phosphor induced by an accelerated electron under high electric field. In general, this phenomenon is utilized in back light devices such as watches, and mobile phones. Particularly, powder electroluminescent devices (PELDs) have high potential of commercial applications because of their flat light emissions and simple printing processes with low cost of production missing semiconductor processes using vacuum and plasma. Recently, PELD devices do not only emit light under an applied voltage, but also produce sound [1], generate electric power caused by triboelectric charging by changing its structure [2], recover the structure owing to the presence of self-recovering materials such as polyacrylic acid hydrogel, and polyurethane [3]. They are expected to facilitate production of versatile light-emitting devices for illumination and digital signage. In conventional PELDs, indium tin oxide (ITO) is used as a transparent electrode. However, ITO is unfavorable in flexible electronic devices owing to its intrinsic brittleness. It is reported that mesh patterned ITO has higher repeated bending durability [4]. Additionally, the efficiency of material utilization is low because the electrodes are developed by an etching process. As an alternative material for ITO, metal-grid electrodes are promising if printing technology enables grid electrodes to be constructed at lower costs because the printing technology helps to improve the efficiency of material utilization as well as simplify the process steps [5, 6]. To overcome this issue, we have developed a gravure offset printed invisible silver-grid laminated with PEDOT:PSS transparent electrodes by using Ag-nanoparticles ink [7, 8]. Nonaka et al. reported on the use of ITO-free PELDs comprising of Au comb-type interdigitated electrodes [9] or Al nano-stripe electrodes [10]. Considering PELDs with Au comb-type interdigitated electrodes, luminance increases with decreasing spacing between Au lines. PELDs with Ag-grid electrodes may improve luminescent property. The Ag-grid electrodes have a tradeoff between conductivity and transparency [7], suggesting that the behavior of luminance as a function of spacing between Ag-grid lines does not conform to that of the current.

In this study, an optimization of luminance in PELDs with Ag-grid laminated with and without PEDOT:PSS electrodes is evaluated by measurements of current and luminance versus voltage characteristics of PELDs with varying spacing between Ag-grid lines.

2 EXPERIMENTAL

2.1 Materials

Polyethylene naphthalate (PEN) film (Teonex-Q65HA) as a substrate was purchased from Teijin Film Solutions Ltd. We used the newly developed ink (L-Ag Nano Metal Ink, ULVAC) as Ag-grid electrode. Poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate)(PEDOT:PSS)(AS601D) used as a conductive polymer was purchased from ARAKAWA CHEMICAL INDUSTRIES, LTD. Zinc sulfide (ZnS)-type particles (GG64) regarded as a phosphor layer was purchased from OSRAM SYLVANIA. Barium titanium oxide ($BaTiO_3$) considered as a dielectric layer was purchased from KANTO CHEMICAL CO., INC. Ag-paste used as a back electrode was purchased from MINO GROUP Co., Ltd. Cyanoresin (CR-V) used as a high dielectric polymer was purchased from SHINETSU Chemical Co., Ltd and Cyclohexanone, the solvent was purchased from FUJIFILM Wako Pure Chemical Co.

2.2 Preparation of electrodes

The Ag-grid electrodes were patterned by gravure offset print using Ag-nanoparticles ink on PEN substrate, and sintered in an oven at a constant temperature of 180 °C for 60 min. The line width was 5 μm. The spacing between the Ag-grid lines range from 25 to 1000 μm. The

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thickness of the lines were 0.6 μm. Furthermore, the PEDOT:PSS dispersion liquid was coated on the Ag-grid electrodes, spaces between lines that range from 100 to 1000 μm, and cured at 100 °C for 1 min.

2.3 Preparation of devices

The high-dielectric polymer paste was prepared by mixing cyanoresin and cyclohexanone using a weight ratio of 3:7. The phosphor particle and $BaTiO_3$ were dispersed in the high-dielectric polymer paste using a weight ratio of 4:6. The developed phosphor and dielectric pastes were printed on the Ag-grid laminated with and without PEDOT:PSS electrodes using an automatic screen printing machine (TU2020-C: SERITECH Co., Ltd.) (Fig. 1). Each material was cured in a constant-temperature oven (FS-405: Yamato Scientific Co., Ltd.) at 150 °C for 6 min.

3 RESULTS and DISCUSSION

3.1 Electroluminescent Property

The characteristics of PELDs on the Ag-grid electrodes without PEDOT:PSS for varying spacing were evaluated using an alternating current voltage of ± 300 V and a frequency of 1 kHz. Fig. 2 shows current and luminance as a function of spacing between the Ag-gird lines of the PELD. The current increases with decreasing spacing between Ag-grid lines, and reaches approximately 0.8 mA at 25 μm spacing. This indicates that the current of the PELD is strongly dependent on the spacing between Ag-grid lines in the electrode. The current increased because electric charges increase with increasing density of the Ag-grid. The luminance increases with decreasing spacing between Ag-grid lines and reaches approximately 150 cd/m^2 at 50 μm spacing. However, the luminance decreases at 25 μm spacing. This is because of the tradeoff between conductivity and transparency of the Ag-grid electrodes. The conductivity of Ag-grid electrode increases with decreasing spacing between Ag-grid lines. Transparency of the Ag-grid electrodes decreases with decreasing spacing between Ag-grid lines as discussed [7]. Luminance of PELDs with Ag-grid electrodes has a maximal value and it decreases with increasing current of the PELD. The luminance decreased at 25 μm spacing, corresponding to the phosphor particle diameter. The phosphor particle diameter is 10 ~ 30 μm. The results reveal that adjusting the spacing between Ag-grid lines aids optimizing the electroluminescence property of the PELDs.

To explain the effects of laminating with PEDOT:PSS for the electroluminescence property, the current and luminance of a PELD with Ag-grid laminated with PEDOT:PSS electrodes was investigated. The characteristics of PELD with Ag-grid laminated with PEDOT:PSS electrodes for varying spacings were evaluated using an alternating current voltage of ± 300 V and a frequency of 1 kHz. Fig. 3 shows the current and

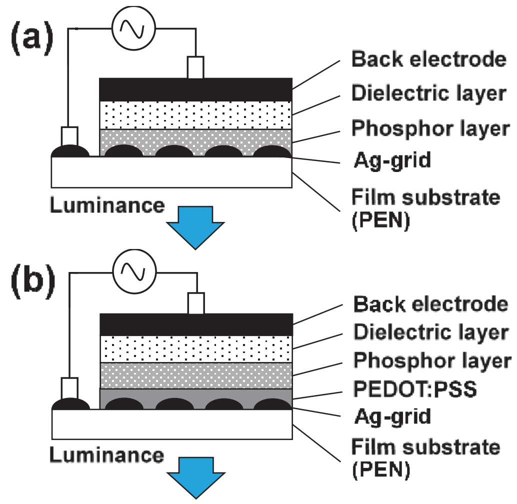

Fig. 1 Structure of the PELDs on the Ag-grid electrodes (a) without and (b) laminated with PEDOT:PSS.

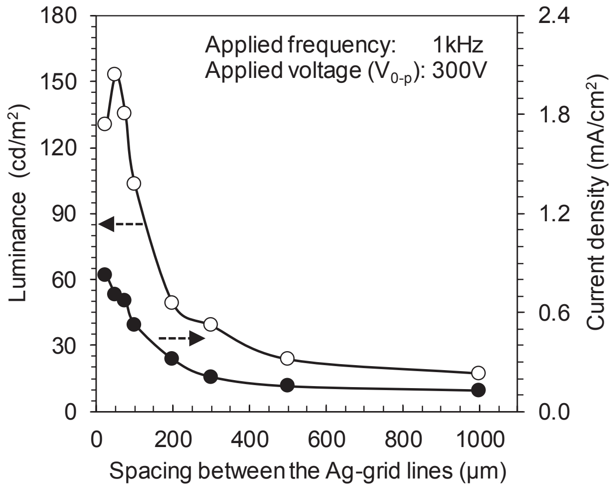

Fig. 2 Current and luminance versus spacing between the Ag-grid lines of the Ag-grid electrodes without PEDOT:PSS of the developed PELD.

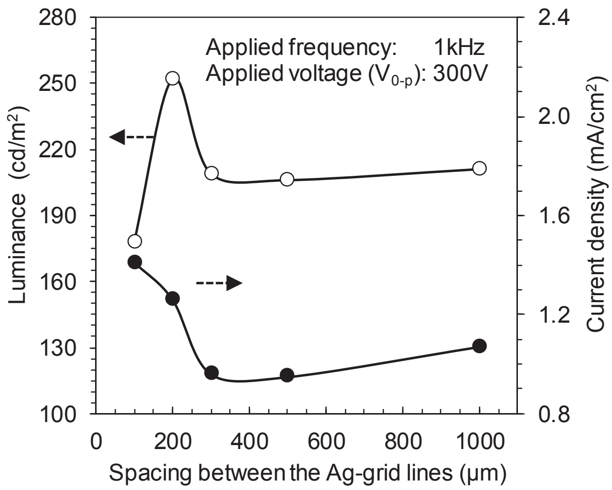

Fig. 3 Current and luminance versus spacing between the Ag-grid lines of the Ag-grid electrodes laminated with PEDOT:PSS of the developed PELDs.

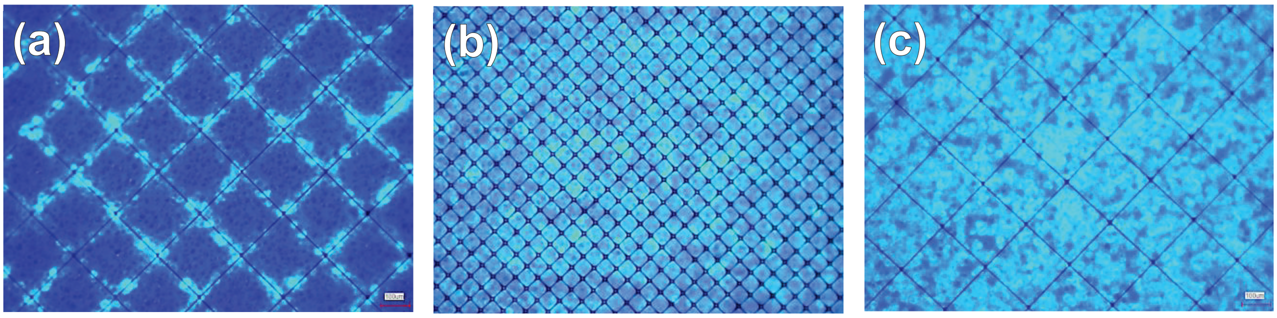

Fig. 4 Microscope images of light emitting from the PELDs on the Ag-grid electrodes (a), (b) without and (c) laminated with PEDOT:PSS. Spacing between the lines: (a) 200 μm, (b) 50 μm, and (c) 200 μm.

luminance as a function of spacing between the Ag-gird lines of the PELDs. The current increases with decreasing spacing between Ag-grid lines, and reaches approximately 1.4 mA at 100 μm spacing. The current showed similar behavior to that without PEDOT:PSS (Fig. 2). The luminance increases with decreasing spacing between Ag-grid lines and reaches a maximal value of approximately 250 cd/m^2 at 200 μm spacing. Generally, spacing between Ag-grid lines, current and luminance of the PELD with Ag-grid laminated with PEDOT:PSS electrodes recorded were higher values than that without PEDOT:PSS. The current and luminance of PELD with Ag-grid laminated with PEDOT:PSS electrodes are high because of the enhancement by laminating with PEDOT:PSS that brings conductivity to the spacing between Ag-grid lines, while keeping transparency of the spacing. The maximum luminance in the PELD with the Ag-grid laminated with PEDOT:PSS electrode occurs at a wider spacing between the Ag-grid lines in comparison with that without PEDOT:PSS.

3.2 Microscopic observation

The light emission behaviors of PELDs with the Ag-grid laminated with and without PEDOT:PSS electrodes were observed by optical microscopic images. Fig. 4(a)(b) shows a microscope image of light emission from the PELD on the Ag-grid without PEDOT:PSS electrode with the spacing between Ag-grid lines 200 μm and 50 μm, respectively. Notice that luminance regions are significantly restricted in the region where the Ag-grid lines meet the phosphor layer in the Ag-grid without PEDOT:PSS-based PELDs (Fig. 4(a)). Field-excited luminescence of the phosphor particles is limited only the surface of the Ag-grid electrodes. From Fig. 4(b), narrowing the spacing between Ag-grid lines brings more uniform luminescence in the entire region despite the Ag-grid lines blocking light. However, the luminance does not increase because of the decrease in transparency caused by narrowing the spacing compared to that of the Ag-grid laminated with PEDOT:PSS electrode-based PELDs. From Fig. 4(c), the lamination with PEDOT:PSS brings uniform luminescent in the spacing between the Ag-grid lines although the spacing is as wide as 200 μm. The luminance of the Ag-grid laminated with PEDOT:PSS electrode-based PELD (250 cd/m^2) shows 5 times higher than that of the Ag-grid without PEDOT:PSS-based PELD (50 cd/m^2) at the same spacing of 200 μm (Fig. 2 and 3). These results indicate that the Ag-grid laminated with PEDOT:PSS transparent electrodes introduces significant improvements of the luminance characteristics of PELDs.

4 CONCLUSIONS

The current and luminance characteristics of PELD on gravure offset printed invisible Ag-grid electrodes laminated with and without PEDOT:PSS transparent electrodes have been evaluated. The PELD on the Ag-grid without PEDOT:PSS electrode shows a maximum luminance of approximately 150 cd/m^2 at the spacing between Ag-grid lines of 50 μm, suggesting that the luminance is strongly dependent on the transparency and the light-extraction efficiency of the Ag-grid electrode. The PELD on the Ag-grid laminated with PEDOT:PSS electrode shows the maximum luminance of approximately 250 cd/m^2 at 200 μm spacing. This is because laminating with PEDOT:PSS brings conductivity to the spacing between Ag-grid lines, while keeping transparency of the spacing. These results indicate that the Ag-grid laminated with PEDOT:PSS transparent electrodes introduces significant improvements of the luminance characteristics of PELDs.

REFERENCES

[1] S. Lee, T. Kang, W. Lee, M. M. Afandi, J. Ryu, and J. Kim, "Multifunctional device based on phosphor-piezoelectric PZT: lighting, speaking, and mechanical energy harvesting", Scientific Reports, Vol. 8, p. 301, (2018).

[2] H. J. Park, S. Kim, J. H. Lee, H. T. Kim, W. Seung, Y. Son, T. Y. Kim, U. Khan, N. M. Park, and S. W. Kim, "Self-powered motion-driven triboelectric electroluminescence textile system", ACS Applied Materials & Interfaces, Vol. 11, pp. 5200-5207, (2019).

[3] G. Liang, Z. Liu, F. Mo, Z. Tang, H. Li, Z. Wang, V. Sarangi, A. Pramanick, J. Fan, and C. Zhi, “Self-healable electroluminescent devices”, Light, Science & Applications, Vol. 7, p. 102, (2018).
[4] K. Sakamoto, H. Kuwae, N. Kobayashi, A. Nobori, S. Shoji, and J. Mizuno, “Highly flexible transparent electrodes based on mesh-patterned rigid indium tin oxide”, Scientific Reports, Vol. 8, p. 2825, (2018).
[5] Y. Jang, J. Kim, and D. Byun, “Invisible metal-grid transparent electrode prepared by electrohydrodynamic (EHD) jet printing”, Journal of Physics D: Applied Physics, Vol. 46, p. 155103, (2013).
[6] S. M. Yang, Y. S. Lee, Y. Jang, D. Byun, and S. H. Choa, “Electromechanical reliability of a flexible metal-grid transparent electrode prepared by electrohydrodynamic (EHD) jet printing”, Microelectronics Reliability, Vol. 65, pp. 151-159, (2016).
[7] M. Ohsawa, and N. Hashimoto, “Flexible transparent electrode of gravure offset printed invisible silver-grid laminated with conductive polymer”, Materials Research Express, Vol. 5, p. 085030, (2018).
[8] M. Ohsawa, and N. Hashimoto, “Bending reliability of flexible transparent electrode of gravure offset printed invisible silver-grid laminated with conductive polymer”, Microelectronics Reliability, Vol. 98, pp. 124-130, (2019).
[9] T. Nonaka, Y. Uraoka, N. Taguchi, and S. Yamamoto, “Evaluation of distributed-type inorganic electroluminescence devices using comb-shaped electrodes”, Japanese Journal of Applied Physics, Vol. 54, p. 03CA03, (2015).
[10] T. Nonaka, and S. Yamamoto, “Optical properties of inorganic electroluminescent devices with nanostripe electrodes”, Japanese Journal of Applied Physics, Vol. 55, p. 03CD03, (2016).