

Enhanced Luminescence of IEL Device With Carbon-Nanotube–Dielectric Composite Layer

Kuo-Feng Chen, Fang-Hsing Wang, Yu-Han Chien, Chin-Chia Chang, and Meng-Ying Chuang

Abstract—This letter proposes a new method for reducing power consumption and enhancing the luminous efficiency of powder inorganic electroluminescence devices by introducing a composite dielectric layer into their structure. The composite dielectric layer contains a composite BaTiO₃-carbon nanotube (CNT) film, which is formed by adding single-wall CNTs into a BaTiO₃ layer. With an appropriate CNT mixing ratio, the power consumption decreased by more than 30%, and the luminous efficiency increased by approximately 50% at a brightness of 400 nits and an operation frequency of 1 kHz.

Index Terms—Efficiency, inorganic electroluminescence (IEL), power consumption, single-wall carbon nanotubes (CNTs) (SWCNTs).

I. INTRODUCTION

THE INORGANIC electroluminescence (IEL) device was developed in 1936 [1]. IEL devices possess numerous advantages, such as low cost, high brightness, and low environmental usage restriction, and are well suited for serving as flat panel displays. The primary principle of an IEL component is to utilize a high electric field to inject electrons, which may collide with luminescent centers of phosphor layer to emit light. Two types of IEL devices exist: the thin-film IEL (TFEL) [2]–[4] and powder IEL (PDEL) [5]–[7]. TFEL devices usually employ a vacuum process at high temperature and therefore have a shortcoming of high manufacturing cost, while PDEL devices use a low-cost screen printing process, which is quite suitable for manufacturing large-size and flexible displays.

In traditional PDEL devices, electrons are injected into the interface between the dielectric and phosphor layers. These accumulated electrons in the interface will inject and accelerate in a high electric field to attain a high energy state and then collide with the luminescent centers of the phosphor layer to

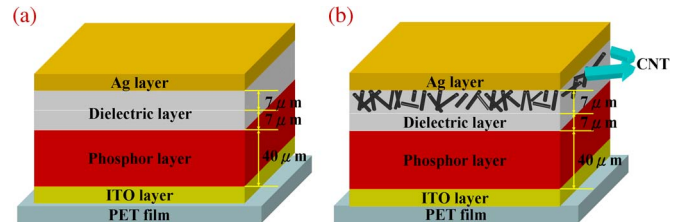


Fig. 1. Schematic structures of the PDEL devices. (a) Without and (b) with a CNT-dielectric composite layer.

emit light. However, PDEL devices have a serious shortcoming of low luminous efficiency due to its low electron injection effect. To enhance luminous efficiency of PDEL devices, this letter introduces single-wall carbon nanotubes (CNTs) (SWCNTs) into the dielectric film of PDEL devices. CNTs possess a high aspect ratio and an excellent electrical conductivity. The role of CNT can be understood as enhancing the local electrical field, allowing electron injection to the active center of phosphor at relatively low operating voltages [8]–[11]. Bae *et al.* [8] reported that EL was enhanced from CNT-incorporated cathodoluminescent phosphor. Utilizing the property that CNTs can carry charges and enhance electric fields, the effects of the CNT–dielectric composite layer on the current consumption, brightness, and efficiency of PDEL devices were investigated.

II. EXPERIMENTS

Fig. 1 shows the structure of the traditional and proposed PDEL devices. A transparent conducting electrode (ITO film) with a thickness of 200 nm was sputtered on a transparent polyethylene terephthalate (PET) substrate with an area of 4.5 cm × 4.5 cm. A phosphor layer (ZnS, produced by Dupont Inc., paste 8150L) was subsequently laminated on the ITO layer by screen printing and then baked at a temperature of 130 °C for 10 min. The thickness of the dry phosphor layer was 40 ± 2 μm obtained by surface microprofile analysis (Kosaka Laboratory Ltd., ET4000LK). Afterward, the first dielectric layer was coated on the phosphor layer by screen printing using BaTiO₃ paste (produced by Dupont Inc., paste 8153) and then baked at 130 °C for 5 min. The dry-film thickness of the first dielectric layer was approximately 7 μm. The second dielectric layer was screen printed on the first dielectric layer using BaTiO₃ paste (Dupont Inc., paste 8153) mixed with SWCNTs (produced by CarbonLex Inc., AP-grade, arc discharge) and then baked at 130 °C for 5 min. The SWCNT had a diameter of 1 nm, a length of 2 μm, and

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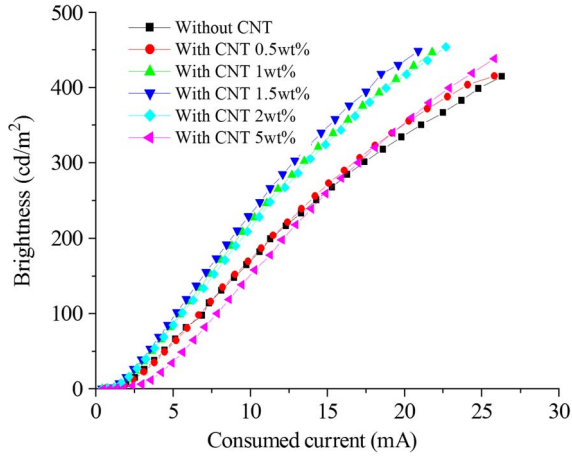


Fig. 2. Brightness of PDEL devices as a function of consumed current at 1 kHz with various CNT mixing ratios.

an electric field enhancement factor β of 2000. The content of SWCNTs in the composite paste varied from 0 to 5 wt%. The CNT-incorporated dielectric paste was initially mixed using three-roller equipment. Subsequently, the CNT-dielectric paste performed dispersion and was debubbled using a planetary centrifugal mixer. It is worth noting that the CNTs are not fully dispersed within the dielectric paste while most CNTs are bundled after these processes. The dry-film thickness of the second dielectric layer was approximately $7 \mu\text{m}$. The total thickness of the dielectric layer was $14 \pm 2 \mu\text{m}$, as shown in Fig. 1.

The electrical and optical properties of the PDEL devices were measured by a hybrid electrical-optical measurement system, containing a multifunctional amplifier system and a colorimeter. The multifunctional machine included a functional generator (Tektronix, AFG-3021) and a piezo driver/amplifier (TREK, PZD-350). For EL measurement, sinusoidal pulses with bias voltage were applied using a pulse generator amplifier at a frequency of 1 kHz. The applied voltage and ac current were monitored using an oscilloscope (Tektronix, TDS-1012). The brightness of EL devices was measured with a luminance colorimeter (KLEIN, K8) from the PET side in a dark room. The measured samples had an area of $4.5 \times 4.5 \text{ cm}^2$.

III. RESULTS AND DISCUSSION

Fig. 2 shows the brightness versus the consumed current of PDEL devices at 1 kHz with various CNT mixing ratios. It was found that, at the same brightness, a lower current consumption was achieved with CNT mixing ratios ranging from 1 to 2 wt%. With a lower or a higher CNT mixing ratio (i.e., 0.5 or 5 wt%), the current consumption is larger than the optimal case and comparable to that without CNTs. Under the condition of a brightness of 400 nits, the current consumption of the sample with the CNT mixing ratios of 1–2 wt% was only 69%–76% as compared to that without CNT incorporation.

Fig. 3 shows the brightness and efficiency of the PDEL devices as functions of power consumption with various CNT mixing ratios. The experiments and measurements were repeated at least three times, and the deviation of the brightness

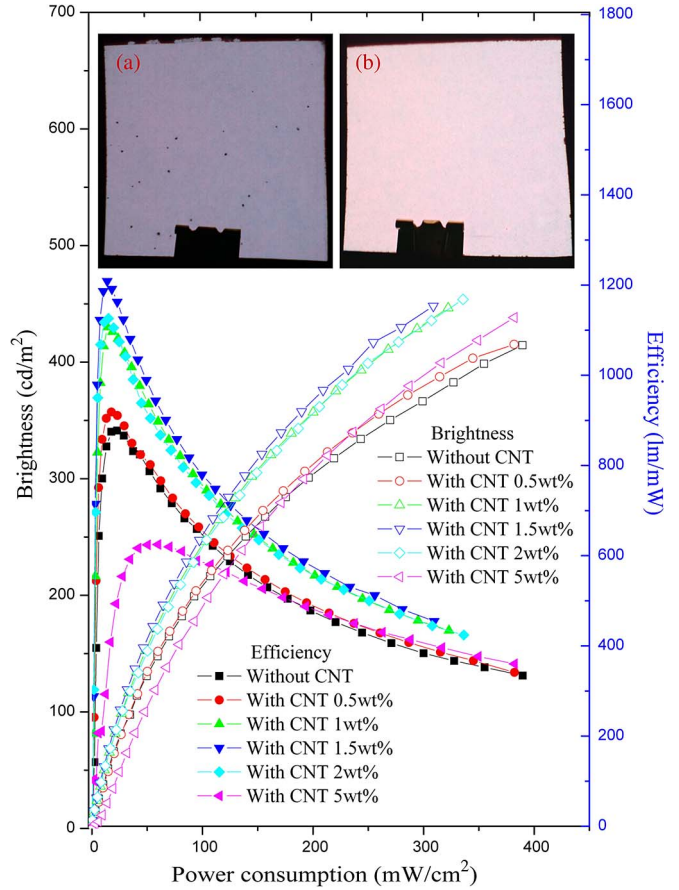


Fig. 3. Brightness and efficiency of the PDEL devices as functions of power consumption at 1 kHz. The insets display the light emission photographs of (a) the traditional PDEL device and (b) the CNT-PDEL device incorporated with 1.5 wt% CNTs into the dielectric layer.

TABLE I
POWER CONSUMPTION AND LUMINOUS EFFICIENCY OF THE PDEL DEVICES UNDER THE CONDITION OF 400 nits AND 1 kHz

SWCNT mixing ratio (%)	0	0.5	1.0	1.5	2.0	5.0
power consumption (mW/cm^2)	358	333	250	239	252	317
luminous efficiency (lm/mW)	351	376	502	525	498	396

and efficiency was within 3%. It was determined that all the brightness increased as the CNTs were incorporated into devices, regardless of the mixing ratio. The power consumption and the luminous efficiency are listed in Table I under the condition of 400 nits and 1 kHz. The power consumption decreased by 30%–33% and the efficiency increased by 42%–50% with the CNT mixing ratios of 1–2 wt% as compared to those without the addition of CNTs. The insets in Fig. 3 show the light emission photographs of the traditional PDEL device and the CNT-PDEL device incorporated with 1.5 wt% CNTs into the dielectric layer at a power consumption of $309.6 \text{ mW}/\text{cm}^2$. The brightness of the PDEL device without CNT addition was 374.5 nits. It increased by 73.5 nits (or 20%) to 448 nits when the device was with a 1.5 wt%-CNT mixing ratio.

The aforementioned results demonstrate that the addition of CNTs into the dielectric layer of the PDEL devices could

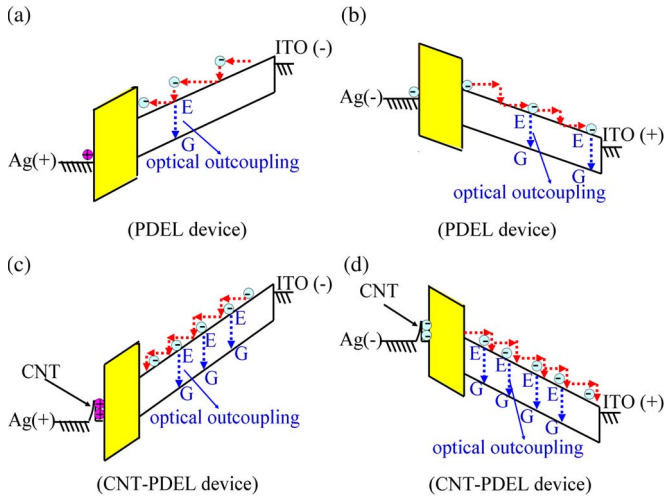


Fig. 4. Energy band diagrams of (a and b) the traditional PDEL device and (c and d) the CNT-PDEL device under forward and reverse bias conditions.

effectively decrease power consumption and enhance luminance efficiency. Previous literature indicated that CNTs on the surface of phosphor can act as local electrical field enhancement sites, inducing the thinning of tunneling barrier thickness, leading to electron emission and acceleration [8]. In this letter, the CNTs incorporated in the dielectric film may enhance the local electric field and thin the thickness of the dielectric layer, leading to more impact excitation with luminescent centers. The field enhancement factor β for CNTs used in this study is estimated to be about 2000 with the length and diameter of 2 μm and 1 nm, respectively. However, excess CNTs (i.e., 5 wt%) cause a possible short circuit, resulting in current leakage in the dielectric layer. Moreover, excess CNTs may enlarge the average CNT bundle size, leading to a decreased field enhancement factor β . Hence, the power consumption and luminance efficiency of the PDEL devices deteriorate.

To understand the mechanism of the enhanced efficiency for the CNT-PDEL devices, Fig. 4(a)–(d) shows the energy band diagrams of the traditional PDEL and CNT-PDEL devices biased at a positive and a negative voltage, respectively. The electric field in the traditional PDEL device totally results from the external voltage source applied between two electrodes. In the CNT-PDEL device, the incorporated CNTs in the composite dielectric layer acted as a conductor and might carry charges supplied from the Ag electrode. The incorporated CNTs might thin the equivalent thickness of the dielectric film while not increasing the leakage current. The charges existing in the CNT-incorporated dielectric layer might build a temporary electric field in the device. Hence, the total electric field increased in the CNT-PDEL device, and thus a larger slope of the energy band in the phosphor layer was obtained. This effect further accelerates electrons in the conduction band of the phosphor layer and increases the electron energy to cause more impact excitation with luminescent centers at a lower bias voltage. Moreover, CNTs with a high aspect ratio can enhance the local electrical field on their surface ($\beta \sim 2000$). Therefore, electrons

can easily overcome the tunneling barrier even at lower bias voltages to arrive around the phosphor layer. Then, the accelerated electrons with sufficient energy cause impact excitation of luminescent center, generating light emission. When the bias polarity is reversed, the incorporated CNTs can carry positive carriers to enhance the electric field, and the electrons from the ITO electrode are injected and accelerated in the opposite direction. The increased electric field also enhances the impact excitation. These processes are repeated, and thus radiative light generation is produced by alternating bias voltage applications.

IV. CONCLUSION

This letter has investigated the effects of a CNT–dielectric composite layer on PDEL devices. After introducing SWCNTs into the dielectric film of the PDEL device with CNT mixing ratios of 1–2 wt%, the current and power consumption decreased by 24%–31% and 30%–33%, respectively, at a brightness of 400 nits and an operation frequency of 1 kHz in comparison with those without CNT incorporation. Thus, the luminous efficiency increases by 42%–50%. The developed composite CNT–dielectric film uses simple and low-cost processes and has potential for the manufacturing of large-area and flexible PDEL displays.

REFERENCES

- [1] G. Destriau, "Recherches sur les seintiuafion de zinc aux rayons," *J. Chem. Phys.*, vol. 33, p. 587, 1936.
- [2] J. Heikenfeld and A. J. Steckl, "Electroluminescent devices using a high-temperature stable GaN-based phosphor and thick-film dielectric layer," *IEEE Trans. Electron Devices*, vol. 49, no. 4, pp. 557–563, Apr. 2002.
- [3] J. F. Wager, J. C. Hitt, B. A. Baukol, J. P. Bender, and D. A. Keszler, "Luminescent impurity doping trends in alternating-current thin-film electroluminescent phosphors," *J. Lumin.*, vol. 97, no. 1, pp. 68–81, Apr. 2001.
- [4] N. Shepherd, D. C. Morton, E. W. Forsythe, and D. Chiu, "The influence of insulator properties on the electro-optical performance of flexible ZnS:ErF3 alternating current thin film electroluminescent devices," *Thin Solid Films*, vol. 515, no. 4, pp. 2342–2346, Dec. 2006.
- [5] J. S. Kim, S. G. Lee, H. L. Park, J. Y. Park, and S. D. Han, "Optical and electrical properties of ZnGa₂O₄/Mn²⁺ powder electroluminescent device," *Mater. Lett.*, vol. 58, no. 7/8, pp. 1354–1357, Mar. 2004.
- [6] J. H. Park, S. H. Lee, J. S. Kim, A. K. Kwon, H. L. Park, and S. D. Han, "White-electroluminescent device with ZnS:Mn, Cu, Cl phosphor," *J. Lumin.*, vol. 126, no. 2, pp. 566–570, Oct. 2007.
- [7] S. D. Han, I. Singh, D. Singh, Y. H. Lee, G. Sharma, and C. H. Han, "Crystal growth of electroluminescent ZnS:Cu,Cl phosphor and its TiO₂ coating by sol-gel method for thick-film EL device," *J. Lumin.*, vol. 115, no. 3/4, pp. 97–103, Nov. 2005.
- [8] M. J. Bae, S. H. Park, T. W. Jeong, J. H. Lee, I. T. Han, Y. W. Jin, J. M. Kim, J. Y. Kim, J. B. Yoo, and S. G. Yu, "Carbon nanotube induced enhancement of electroluminescence of phosphor," *Appl. Phys. Lett.*, vol. 95, no. 7, p. 071 901, Aug. 2009.
- [9] S. H. Jo, Y. Tu, Z. P. Huang, D. L. Carnahan, D. Z. Wang, and Z. F. Ren, "Effect of length and spacing of vertically aligned carbon nanotubes on field emission properties," *Appl. Phys. Lett.*, vol. 82, no. 20, pp. 3520–3522, May 2003.
- [10] K. W. Lee, S. P. Lee, H. Choi, K. H. Mo, J. W. Jang, H. Kweon, and C. E. Lee, "Enhanced electroluminescence in polymer-nanotube composites," *Appl. Phys. Lett.*, vol. 91, no. 2, p. 023 110, Jul. 2007.
- [11] J. H. Choi, S. H. Choi, J. H. Han, J. B. Yoo, H. Y. Park, T. Jung, S. G. Yu, I. T. Han, and J. M. Kim, "Enhanced electron emission from carbon nanotubes through density control using in situ plasma treatment of catalyst metal," *Appl. Phys. Lett.*, vol. 94, no. 1, pp. 487–490, Jul. 2003.