Improvement of stability for organic light emitting devices by thermal and electrical treatment

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Abstract

Highly stable organic electroluminescent devices have been achieved by treatment of thermal and electrical annealing. We investigate here the performance of theses devices at temperatures and pulse aging. We also demonstrate improved device stability due to thermal and electrical treatment, and the brightness decays at no treatment, thermal only, electrical only and thermal/electrical treatment were 86.6%, 89.5%, 93.0%, and 96.7%, respectively, in the after 150 h of operation driven with an initial luminance of 1,000 cd/m^2.

1. Objective and Background

Organic electroluminescence (EL) devices are of considerable interest in various display applications because of their high efficiency and colors. Using multilayer structures[1], doped emitting layers[2], novel transport and luminescent materials[3,4], including polymers[5–7], and efficient injection contacts[8,9], these organic-based devices can be operated with a dc voltage as low as a few volts and provide luminous efficiencies greater than 1 lm/W over a wide spectral range[10–12]. To be practical, it is also necessary to demonstrate that these devices have sufficient reliability. However, most organic EL devices reported so far have a short operational lifetime. The improvements in device stability due to thermal treatment which was shown to be an inherent property of some phosphorescent dopants[13]. However it is needed to get longer lifetime of EL device and to be more deeply understood the main mechanism determining the degradation of EL device.

In this study, we will show that excellent stability can also be produced with a phosphorescent emitter using a combination of thermal and suitable pulse treatment.

2. Results

The organic and metal layers were thermally deposited at 0.1 nm/s in a vacuum of $<10^7$ Torr to yield 9 mm^2 devices. The structure of our EL device is ITO/HIL/HTL/EML(phosphorescent emitter) /ETL/EIL/Al. The EL devices were encapsulated in a dry nitrogen atmosphere (<1 ppm H_2O/O_2) using a glass lid and UV cured epoxy edge seal. A desiccant was added inside the package to react with any byproducts of the epoxy cure and any residual water or oxygen present within the encapsulated volume.

The EL luminance-Current-Voltage (L-I-V) characteristics and stability were measured using a DMS 401 spectrophotometer and OLED lifetime measurement system, respectively. For the electrical treatment test, the constant voltage source was modified to include a constant reverse voltage component in its temporal wave form. This excitation was equivalent to a symmetric ac power source. In the forward half-cycle, a constant voltage source was impressed on the EL device, whereas in the reverse cycle a constant voltage was also applied. The light output and the forward bias voltage were monitored continuously throughout the stability test.

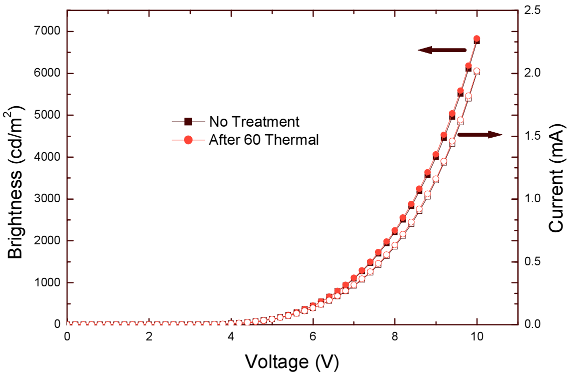

Figure 1. The variation of L-I-V characteristics of an EL device between no treatment and thermal annealing of 60℃ for 2 hours.

The variation of L-I-V characteristics of an EL device between no treatment and thermal annealing of 60℃ for 2 hours plotted in Fig. 1. After exposure to 60℃ the change in the brightness and current for devices was less than 1%. And Figure 2 shows the variation of L-I-V characteristics of an EL device between no treatment and electrical ±10V square pulse aging for 2 hours at room temperature. At high voltage region, the degradation of EL device is occurred due to high electrical stress. It was generally found that the luminance decay rate is directly proportional to the injection current density. This behavior means that the luminance degradation is coulombic, and that the degradation event resulting from charge injection is cumulative and irreversible [14]. Therefore, excessive high electrical aging of EL device is not suitable for increasing the performance of EL characteristics. However, it was found that this instability can be further reduced using an ac excitation scheme, which provides a reverse bias voltage component in the temporal wave form [14].

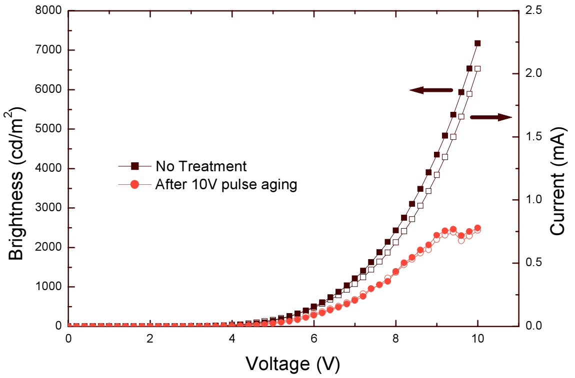

Figure 2. The variation of L-I-V characteristics of an EL device between no treatment and electrical aging of ±10V square pulse for 2 hours.

This reverse bias presumably is effective in preventing a buildup of trapped space charges in the organic layers accumulated during the current flow in the forward cycle by ‘‘detrapping’’ them during the reverse cycle. This ac stabilization effect is relatively independent of the frequency of the drive wave form, but the amplitude needed for the reverse bias should be as large as the forward bias. Another effect that the reverse bias may have is to cause the ‘‘burn out’’ of any localized conducting filaments that might be connecting the two parallel electrodes in the thin-film EL device structure. The reverse bias voltage is considered more effective in removing these defects because the reverse bias current is not shunted by the EL diode as in the case of forward bias.

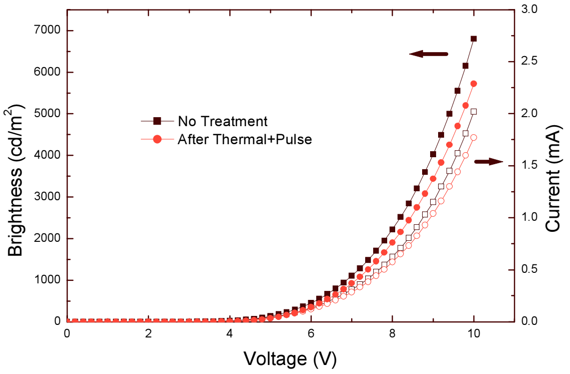

Figure 3. The variation of L-I-V characteristics of an EL device between no treatment and 60℃ thermal and ±10V square pulse aging for 2 hours.

Figure 3 shows the variation of L-I-V characteristics of an EL device between no treatment and 60℃ thermal and ±10V square pulse aging for 2 hours. After exposure to 60℃ and ±10V square pulse aging, the change in the brightness and current for devices was less than 4%. However the degradation of EL characteristics is not occurred in spite of applying high electrical stress. One possible explanation for this observation is diffusion of the interface layer molecules, at temperatures above the Tg of one or layers [15,16]. Diffusion can lead to fused mixed layers formation, which is known to improve OLED lifetimes [17]. Thermal treatment of devices has been reported [18] and shown to cause an improvement in efficiency and a reduction in operating voltage

Lifetime of the EL device prior to and after exposure to thermal and electrical treatment was measured at room temperature at a dc constant current density of about 45 mA/cm^2.

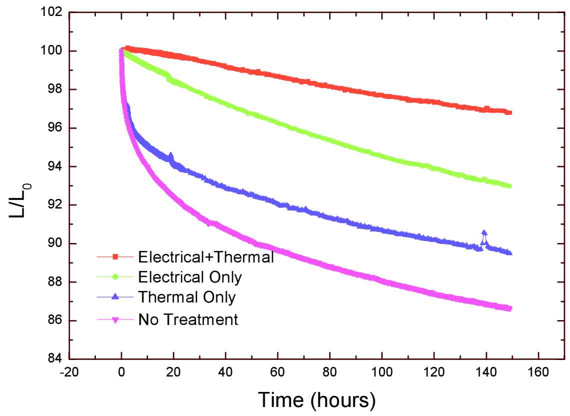

Figure 4. Brightness decays at no treatment, thermal only, electrical only and thermal/electrical treatment.

Figure 4 shows the lifetime of the EL device exposed to various treatments for 2 hours. The brightness decays at no treatment, thermal only, electrical only and thermal/electrical treatment were 86.6%, 89.5%, 93.0%, and 96.7%, respectively, in the after 150 hours of operation driven with an initial luminance of 1,000 cd/m^2. The relative percentage improvement at different operating treatment indicates that the device has undergone a change in its activation energy. With a pulse drive wave form, the problem of space–charge accumulation in the EL layers has been alleviated. As a result, the EL characteristics are stabilized during operation.

It is also interesting to note that the tested device suffered a few instant drops in luminance within the first 150 h of operation. The introduction of a burn-in procedure prior to lifetime would reduce the initial efficiency slightly but would dramatically lessen the observed change in luminance as a function of time. This is an important consideration when producing commercial OLED displays where differential aging issues can affect the useable life of a display long before the 50% luminance decay point.

3. Impact

In conclusion we have demonstrated improvements in device stability due to thermal and electrical treatment. The brightness decays at no treatment, thermal only, electrical only and thermal/electrical treatment were 86.6%, 89.5%, 93.0%, and 96.7%, respectively, in the after 150 h of operation driven with an initial luminance of 1,000 cd/m^2. The resultant lifetime performance would help enable long lived OLED displays by reducing the impact of differential thermal/electrical aging effects.

4. Acknowledgements

The authors acknowledge the assistance of Sang Pil Lee, Jung Mi Choi, Un Cheol Sung and Dr. Hoon Kim in the fabrication of EL devices.

5. References

[1] C. W. Tang and S. A. Van Slyke, Appl. Phys. Lett. **51**, 913 (1987).

[2] C. W. Tang, S. A. Van Slyke, and C. H. Chen, J. Appl. Phys. **65**, 3610 (1989).

[3] S. Saito, T. Tsutsui, M. Era, N. Takada, E. Aminaka, and T. Wakimoto, Mol. Cryst. Liq. Cryst. Sci. Technol., Sect. A **253**, 417 (1994).

[4] C. Hosokawa, H. Higashi, H. Nakarmura, and T. Kusumoto, Appl. Phys. Lett. **67**, 3853 (1995).

[5] J. H. Borroughes, D. D. C. Bradley, A. R. Brown, R. N. Marks, R. H. Friend, P. L. Burn, and A. H. Holmes, Nature **347**, 539 (1990).

[6] D. Braun, E. G. J. Staring, R. C. J. E. Demandt, G. L. J. Rikken, Y. A. R. R. Kessener, and A. H. J. Venhuizen, Synth. Met. **66**, 75 (1994).

[7] D. R. Baigent, N. C. Greenham, J. Gruner, R. N. Marks, R. H. Friend, S. C. Moratti, and A. B. Holmes, Synth. Met. **67**, 3 (1994).

[8] Y. Yang, E. Westerweele, C. Zhang, P. Smith, and A. J. Heeger, J. Appl. Phys. **77**, 694 (1995).

[9] T. Wakimoto *et al.*, Technical Digest, Int. Symposium on Inorganic and Organic Electroluminescence, p. 77 (1994).

[10] Y. Shirota *et al.*, Appl. Phys. Lett. **65**, 807 (1994).

[11] H. Tokailin, H. Higashi, C. Hosokawa, and T. Kusumoto, Proc. SPIE **1910**, 38 (1993).

[12] T. Sano, M. Fujita, T. Fujii, Y. Nishio, Y. Hamada, and K. Shibata, Extended Abstracts, The 41st Spring Meeting of The Japanese Society of Physics and Related Societies, 28 (1994).

[13] V. I. Adamovich, M. S. Weaver, R. C. Kwong, and J. J. Brown, Curr. Appl. Phys. **5**, 15 (2005).

[14] S. A. Van Slyke, C. H. Chen, and C. W. Tang, Appl. Phys. Lett. **69**, 15 (1996).

[15] C. C. Wu, C. W. Chen, and Y. T. Lin, Appl. Phys. Lett. **79**, 3023 (2001).

[16] C. C. Wu, C. W. Chen, and T. Y. Cho, Appl. Phys. Lett. **83**, 611 (2003).

[17] Z. Popovich, G. Vamvounis, H. Aziz, and N. X. Hu, Proc. SPIE, 4800 (2002).

[18] C. W. Chen, T. Y. Cho, and C. C. Wu, Appl. Phys. Lett. **81**, 1570 (2002).