



Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/gmcl19>

Novel Techniques for Characterization of Degradation in Polymeric Light Emitting Devices

Lee-Mi Do^a, Taehyoung Zyung^a, Do-Hoon Hwang^a,
Hye-Yong Chu^a & Seong Hyun Kim^a

^a Electronics and Telecommunications Research
Institute, P.O. Box 106, Yusong, Taejeon, 305-600,
Korea

Version of record first published: 24 Sep 2006

To cite this article: Lee-Mi Do, Taehyoung Zyung, Do-Hoon Hwang, Hye-Yong Chu & Seong Hyun Kim (1999): Novel Techniques for Characterization of Degradation in Polymeric Light Emitting Devices, *Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals*, 337:1, 43-48

To link to this article: <http://dx.doi.org/10.1080/10587259908023373>

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use: <http://www.tandfonline.com/page/terms-and-conditions>

This article may be used for research, teaching, and private study purposes. Any substantial or systematic reproduction, redistribution, reselling, loan, sub-licensing, systematic supply, or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae, and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand, or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

Novel Techniques for Characterization of Degradation in Polymeric Light Emitting Devices

LEE-MI DO, TAEHYOUNG ZYUNG*, DO-HOON HWANG,
HYE-YONG CHU and SEONG HYUN KIM

*Electronics and Telecommunications Research Institute P.O. Box 106, Yusong,
Taejon 305-600, Korea*

Durability of the polymer light emitting device is a key for the industrial application. The durability depends on the degradation procedure of the device. The degradation of the device initiates with the dark spot that is the non-emissive portion of the polymer layer. Formation process of dark spots during the device operation in polymeric light-emitting diodes (PLEDs) has clarified by *in-situ* measurement with the interferometer, time-resolved confocal laser scanning microscope, and other various analytical instruments.

Keywords: Degradation; Light emitting device; Electroluminescence; Surface defects; Organic/inorganic Interfaces

INTRODUCTION

After the first report of organic/polymeric electroluminescence (EL) device by Tang and Burroughes^[1-2], organic/polymeric EL devices have been studied extensively due to their commercially attractive advantages including low cost, full color, and feasibility of the flexible large area flat panel display. As far as quantum efficiencies, brightness, and response times are regarded, organic/polymeric light emitting devices have reached a level, where technical applications seem to be the next step. A drawback, however, is the limitations due to their relatively short life- and operating

* Corresponding author: thz@ard.etri.re.kr

times. Long operating time of the device for commercial development at a sufficient brightness has not been achieved yet. The reliability and the durability of organic or polymeric multilayer EL devices still remain as a bottleneck for the practical use. Therefore, it is important to investigate the origin of the degradation in the EL device resulting in the device failure.

Even though many reports have described the possible degradation processes in the organic/polymeric EL devices^[3-7], one of the major difficulties is that *in-situ* sequential change of the degradation progress during the operation is undetectable due to the mechanical limitation of various analytical instruments. In this paper, we discuss the novel characterization techniques of the device degradation including *in-situ* sequential measurement of the initial stage of dark spot formation in degradation process.

EXPERIMENTAL SECTION

The studied EL device had a typical single polymer layer structure of ITO/poly(2-methoxy-5-(2'-ethylhexyl-oxy)-p-phenylenevinylene (MEH-PPV)/Al. The 100 nm thick polymer layer was formed by spin coating onto ITO coated glass. Al top electrode was thermally deposited with the rate of 1-2 Å/sec. The characterization techniques used here for the investigation of the degradation are the optical spectroscopies such as UV-Vis, PL, EL, FT-IR and *in-situ* measurements using time-resolved confocal laser scanning microscopy, interferometry. All the experiments were performed under ambient conditions.

RESULTS AND DISCUSSION

It is not easy to investigate the short term degradation behaviour of the device under operation. The degradation process during device operation can

be observed by *in-situ* sequential measurements of the PL image variation with an inverted Karl Zeiss time resolved confocal laser scanning microscope (CLSM) and the interferometric image change upon device operation. Fig. 1 shows the sequence of time resolved CLSM PL images taken at the different operating times. As indicated by the arrows in Fig. 1, there are two kinds of spots in the PL images: the completely dark one (position 1 in Fig.1, hereafter we call it type I defect) and the circular shaped one with dark magenta color (position 2 in Fig.1, type II defect). Type I defect is initiated from the impurities or the inhomogeneities with less than a few μm size in the pristine device as shown in Fig. 1. Type I grows continuously during the EL operation and coalesces gradually each other. In contrast, type II defect appears suddenly and the size does not change during the operation.

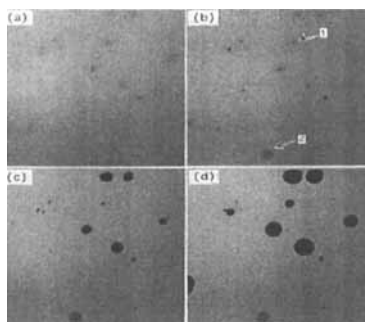


FIGURE 1. The sequential time-resolved CLSM PL images in ITO/MEH-PPV/Al devices. (a), (b), (c), (d) are taken at the operating time of the devices for 0 sec, 400 sec, 540 sec, 630 sec, respectively.

Fig. 2 shows the surface and the cross-sectional SEM images of two types of spots. As shown in Fig. 2(a), Al electrode at type I defect is sunk in the depth of 100 nm yielding the dented circle shape and its morphology is

seriously changed. Some part of the Al electrode is burnt out depending on the applied electric field strength. These degradation progresses, i.e., burning in and/or out are the typical phenomena in single layered polymeric EL devices. According to the atomic force microscopy study of the type I defect as seen in Fig. 2(c) and (d), we find that sharp peak at the center of every dented circle. Radial size of the peak does not change while the dented circle grows. When the type I defects are observed by a transmission optical microscope, the center of type I defects is transparent. It may suggest that the center of Al electrode is changed into the optically transparent Al_2O_3 during the device operation.^[3,8] In contrast, the type II spot has a dome shape. Their cross-sectional image as seen in Fig. 2(b) clearly shows that the polymer layer is detached from the ITO electrode. The formation mechanism of type II defect is not clear yet. It may be related to the gas evolution at the interface between ITO and the polymer due to the electrochemical reaction with moisture.^[9]

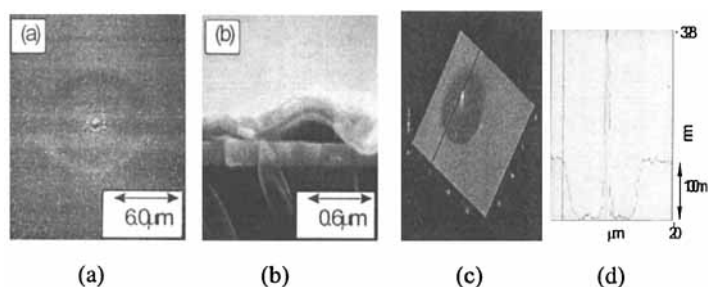


FIGURE 2. SEM images of a partially degraded ITO/MEH-PPV/Al device (a) top view of a type I defect and (b) cross-sectional view of a type II defect. (c) the AFM images of dented site (a), and (d) the cross-sectional profile along the line in (c).

Fig. 3 shows four *in-situ* sequential cross sectional profiles of the interferometric images at the dark spot in PL images. The dark spot at

position 1 in Fig. 3 (a) is observed with interferometer after applying 8 V for 1 min. In Fig. 3 (a), the dip at position 2 in the cross sectional profile indicates the pinhole at the surface. As shown in Fig. 3, the morphology of Al electrode is changed depending on the device operation time. By comparing the profiles at position 2, the images are differently changed even under the same applied voltage. Fig. 3 (b), (c) and (d) show the change in cross sectional profiles of the pinhole obtained after operating for 45, 77 and 100min at 8 V, respectively. The cross sectional profile of Al electrode changes with time from dip to sharp con-shape peak. Therefore, we found that the morphological change of Al electrode was initiated from the pinhole of Al surface.

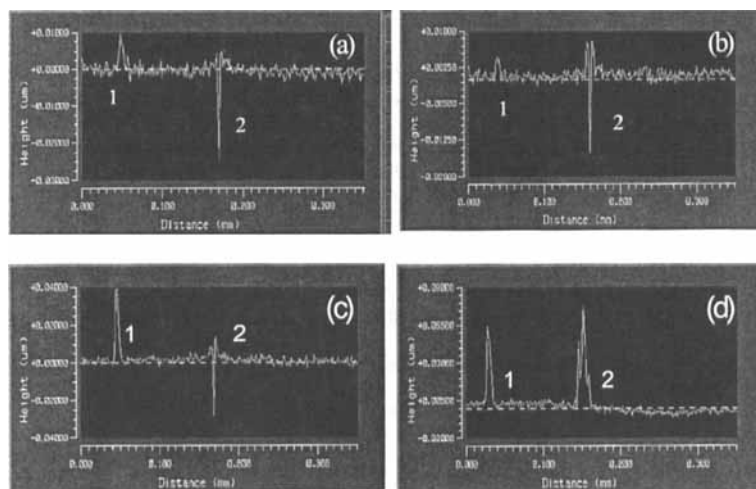


FIGURE 3. The change of cross sectional profiles of interferometer images in EL devices during the constant bias voltage. The sequential images are taken at 1min (a), 45min (b), 77 min (c), and 100 min (d) at 8 V. The samples are stored for 6 hrs in air condition (humidity, $40 \pm 1\%$).

CONCLUSION

In summary, we observe the degradation process with *in-situ* measurement techniques. According to the interferometric images, it is clear that the dark spot originates from the pinhole leading to the severe damages on the microstructure of polymer the layer, polymer/metal interface, and metal electrode. Interferometry and time-resolved confocal laser microscopy are both powerful tools for the investigation of the short term degradation behaviour of the polymeric EL devices.

Acknowledgments

This research is supported financially by MOC in Korea.

References

- [1] C. W. Tang and S. A. Vanslyke, *Appl. Phys. Lett.* **51**, 913 (1987).
- [2] J. H. Burroughes, D.D.C. Bradley, A. R. Brown, R. N. Marks, K. Mackey, R. H. Friend, P. L. Burns, and A. B. Holmes, *Nature*, **347**, 539 (1990).
- [3] L. M. Do, E. M. Han, Y. Niidome, M. Fujihira, T. Kanno, S. Yoshida, A. Maeda, and A. J. Ikushima, *J. Appl. Phys.* **76**, 5118 (1994).
- [4] T. Zyung and J. J. Kim, *Appl. Phys. Lett.* **67**, 3420 (1995).
- [5] J. R. Sheats, H. Antoniadis, M. Hueschen, W. Leonard, R. Moon, D. Roitman, and A. Stocking, *Science* **273**, 884 (1996).
- [6] J. McElvain, H. Antoniadis, M. R. Hueschen, J. N. Miller, D.M. Roitman, J. R. Sheats, R. L. Moon, *J. Appl. Phys.* **80**, 6002 (1996).
- [7] L. M. Do, K. Kim, T. Zyung, H. K. Shim, and J. J. Kim, *Appl. Phys. Lett.* **70**, 3470 (1997).
- [8] P.E. Burrows, V. Bulovic, S.R. Forrest, L.S. Sapochak, D.M. McCarty, and M.E. Thompson, *Appl. Phys. Lett.*, **65**, 2922 (1994).
- [9] B.H. Cumpston, and K. F. Jensen, *TRIP*, **4**, 151 (1996).