

# Effect of Contaminant Particles on Folding of Encapsulating Organic-Inorganic Multilayer for Foldable OLEDs

**Yun Taek Park, Sang Woo Kim, Gui Young Han, Sung Min Cho**

School of Chemical Engineering, Sungkyunkwan University, Korea

Keywords: Thin film encapsulation, Folding, Particle

## ABSTRACT

The effect of contaminant particles on the folding of encapsulating organic-inorganic multilayer was investigated. The stability of the organic-inorganic multilayer thin films was evaluated when they were folded inward or outward in 1 mm radius according to the size of the contaminant particles and the thickness of the multilayer thin films. It was confirmed that the organic-inorganic multilayer films deposited by the atomic layer deposition and plasma-enhanced chemical vapor deposition methods completely covers the contaminant particles regardless of the size of the particles. However, due to the angle of curvature in the resulting encapsulation structure caused by the presence of the contaminant particles, higher stress is generated around the particles and the cracking due to folding is more easily occurred. In this study, we proposed the stability criteria for the thickness of encapsulation layer upon folding even in the presence of contaminant particles

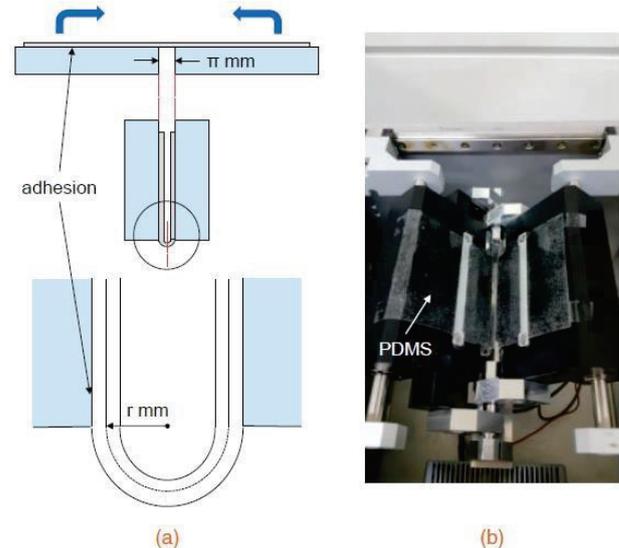
## 1 INTRODUCTION

We have secured the stability of OLED devices by encapsulation with thin film of organic / inorganic composite layer by vitex method. However, as the research of foldable display or rollable display becomes more active, the mechanical properties of encapsulation layer will be seriously deteriorated by contaminant particles, and it is time to identify it. The objective of this study is to investigate the particle coverage of the encapsulation thin film when the contaminant particles are present on the surface of the OLED devices and to evaluate the effect of the covered particles on folding of the encapsulation thin film. This study is very important in the implementation of foldable OLEDs and ultimately aims to provide a criteria for stable thickness of thin film encapsulation for folding.

## 2 EXPERIMENT

This section describes the details about the text content of the manuscript. An  $\text{Al}_2\text{O}_3$  thin film grown by atomic layer deposition (ALD) was used as the thin film encapsulation layer for this study. The organic layer used in this study caused plasma phenomenon by using n-hexane. The plasma polymer layer fragments the organic material from the polymerization by-products of the radical. First, a 1- $\mu\text{m}$ -thick poly (methyl methacrylate) (PMMA) layer was spin-coated onto a 50- $\mu\text{m}$ -thick polyimide (PI) substrate for

surface planarization. Since a poly (ethylene



**Fig. 1. Folding condition**

(a) Schematic diagram of 1 mm diameter folding.

(b) Photographic image of folding apparatus.

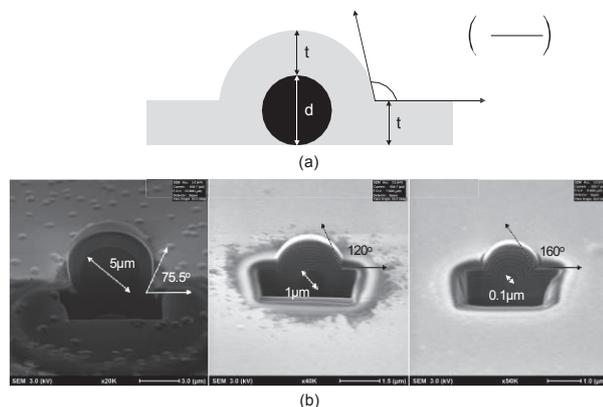
Two folding plates are spaced  $\pi$  mm apart and designed to have a spacing of 2 mm at the folding. The sample is tightly attached to the PDMS surface on the folding plate during the folding.

terephthalate) substrate of the same thickness is damaged by repetitive folding, we utilized a PI substrate causing no damage under the same condition owing to the relatively low Young's modulus. Polystyrene was used as contaminated particles, and diluted 1: 1000 with IPA. An spin coating was carried out at 4000 RPM conditions for application.  $\text{Al}_2\text{O}_3$  and plasma polymer layers were deposited on the contaminated substrate by ALD and plasma-enhanced chemical vapor deposition, respectively. For the deposition of the  $\text{Al}_2\text{O}_3$  layer, TMA and ozone were used. Each precursor was injected into a deposition chamber maintained at a pressure of  $3 \times 10^{-2}$  Torr for 2 s. After the injection of each precursor, the chamber was purged for 10 s using argon (Ar) gas. The substrate temperature was maintained at 90 °C and the  $\text{Al}_2\text{O}_3$  growth rate per cycle was 1.2 Å. The plasma polymer layer was deposited in Ar plasma using n-hexane and the deposition rate was 20 nm/min. The water vapor transmission rate (WVTR) of the fabricated encapsulation layers was measured by the Ca test. The measured WVTRs of contaminated 10 dyad multilayer (1

dyad: 5-nm-thick  $\text{Al}_2\text{O}_3$  layer and 20-nm-thick organic layer) was  $7 \times 10^{-4} \text{ gm}^{-2} \text{ day}^{-1}$ , respectively. Here, the  $\text{Al}_2\text{O}_3$  layer grown by ALD has excellent moisture resistance, whereas the plasma polymer has poor moisture resistance and thus does not significantly affect the WVTR. Therefore, the thickness of the  $\text{Al}_2\text{O}_3$  layer was fixed at 50 nm, and the folding stabilities of the inorganic single layer and organic-inorganic multilayer with the same total thickness of the  $\text{Al}_2\text{O}_3$  layer were evaluated. However, in the case of the organic-inorganic multilayer barrier, a lower WVTR is usually obtained because the path of the moisture penetrating through the defects becomes tortuous. Nevertheless, it was confirmed by the WVTR ( $8 \times 10^{-4} \text{ gm}^{-2} \text{ day}^{-1}$ ) that the barrier properties were maintained when 1R folding was performed after contaminant particles were applied. In addition, the side state was checked by using a Focused Ion Beam (FIB) to check whether the barrier was broken during 1R folding in the presence of contaminant particles.

### 3 RESULTS

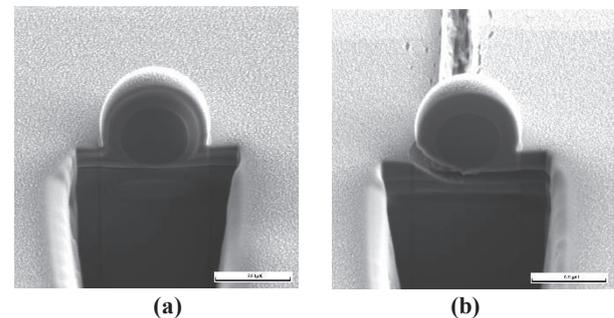
In this paper, we can recognize thin film encapsulation for stable foldable OLED display is advantageous as the thickness of encapsulation layer is thinner. If contaminant particles are present in the foldable OLED display, when the encapsulation layer is thick, it can cover the particles sufficiently and be stable even when folded. On the other hand, if the thickness of the encapsulation layer is very thin, the contaminant particles may not be covered enough, encapsulation layer crack due to folding. As the structure of the organic-inorganic multilayer encapsulation layer, a structure in which 5nm  $\text{Al}_2\text{O}_3$  inorganic layer and 60nm plasma polymer layer was alternately stacked 10 dyads was always stable to 1R folding.



**Fig. 2. (a) Schematic diagram representing 100% step coverage of an encapsulating thin film (thickness  $t$ ) onto a particle (diameter  $d$ ); (b) cross-sectional SEM images of particles encapsulated with organic-inorganic multilayer thin film.**

If the organic-inorganic multilayer encapsulation layer with a total thickness of ( $t$ ) completely covers 100% of spherical contaminant particles with diameter ( $d$ ), it must

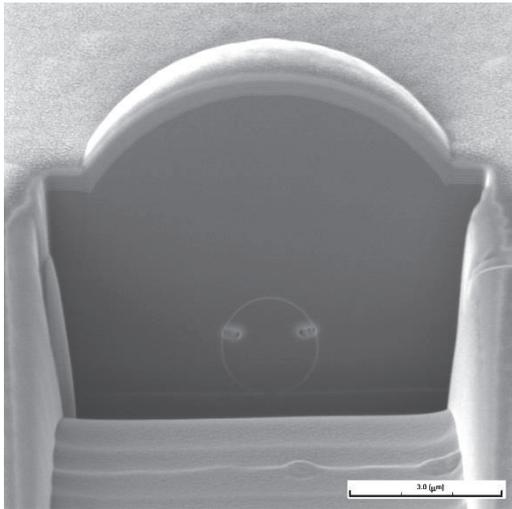
have a covering angle as shown in Fig 2 (a). In fact, the organic-inorganic multilayer encapsulation layer was deposited on spherical particles having a diameter of 5, 1 and  $0.1 \mu\text{m}$ , and the coating angles were found to be in close agreement with the expected coating angles. The larger the size of the contaminated particles, the lower the coating angle, and in the case of in-folding toward the top of the particles, cracking is most likely from the periphery of the polluted particle with the lowest coating angle. Therefore, the occurrence of cracking by folding is expected to depend on the size ( $d$ ) of the contaminant particles and the thickness ( $t$ ) of the encapsulation film. The effect of contaminant particles on the crack formation of the encapsulation film was investigated when the contaminant particles were present in the fold.



**Fig. 3. (a) Cross-sectional SEM image showing around a contaminant particle before 1R in-folding. (b) Cross-sectional SEM image showing a crack originated from a perimeter position around a contaminant particle after 1R in-folding.**

Fig. 3 is a SEM photograph of cracks generated after 1R repeated folding after depositing spherical particles having a diameter of  $2 \mu\text{m}$  by depositing 5 nm  $\text{Al}_2\text{O}_3$  inorganic thin film and 60 nm plasma polymer thin film alternately. The total thickness of this organic-inorganic multilayer encapsulation thin film structure is 590 nm. Calculating the cover angle using the equation shown in Figure 2 shows that it is about  $75^\circ$ . The actual measured coating angle was about  $82^\circ$ , and it was confirmed that the crack occurred in the circumference of the polluted particle with the steepest coating angle when 1R in-folding occurred at the coating angle.

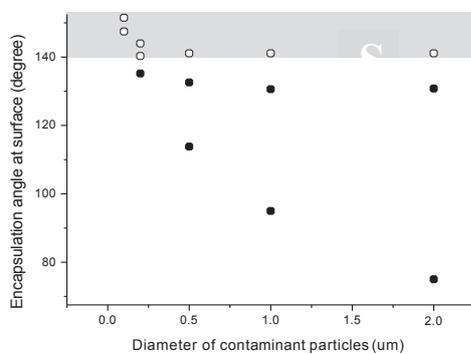
Fig. 4 is a SEM photograph of cracks generated after 1R repeated folding after depositing spherical particles having a diameter of  $2 \mu\text{m}$  by depositing 8  $\mu\text{m}$  planarization layer by PECVD. After planarization layer, deposited encapsulation a structure in which 5nm  $\text{Al}_2\text{O}_3$  inorganic layer and 60nm plasma polymer layer was alternately stacked 10 dyads. The total thickness of this organic-inorganic multilayer encapsulation thin film structure is 8590 nm. Calculating the cover angle using the equation shown in Figure 2 shows that it is about  $141^\circ$ . The actual measured coating angle was about  $146^\circ$ . After 1R folding, it can't find crack around contaminant particles.



**Fig. 4. Cross-sectional SEM image showing a contaminant particle with planarization layer after 1R in-folding**

#### 4 DISCUSSION

In fact, there is a possibility that large contaminant particles are generated in the manufacturing process of OLED display, and if the large contaminant particles exist on the surface before the thin film encapsulation process, the surface is flattened before the thin film encapsulation to minimize the occurrence of cracks in the repeated folding process. It is necessary to loosen the surface coating angle of the thin film to  $140^\circ$  or more by performing the procedure.



**Fig. 5. 1R folding stability of organic-inorganic multilayer encapsulation structure when contaminant particles exist on the folding surface.**

As shown in Fig. 10, when the coating angle was more than  $140^\circ$ , it was found to be stable to 1R in-folding.

#### 5 CONCLUSIONS

From the above results, organic-inorganic multilayer encapsulation layer deposited by ALD and PECVD is confirmed that the coating uniformity of contaminant particles is about 100%. When contaminant particles are present and folding occurs, cracks are generated near the perimeter of the contaminant particle. The occurrence of

cracks is closely related to the covering angle, which can occur more often in-folding than out-folding. If the coating angle was looser than about  $140^\circ$ , it was found to be stable even for folding in 1R. Even if there are contaminant particles, it is very important to planarization before deposited thin film encapsulation layer so that the coating angle is loosened to more than  $140^\circ$  around particles. This research is looking forward to be used not only for the foldable OLED display under development but also for the encapsulation layer for rollable or stretchable OLED display.

#### REFERENCES

- 1 Y. C. Han, E. G. Jeong, H. Kim, S. Kwon, H. G. Im, B. S. Bae, K. C. Choi, Reliable thin-film encapsulation of flexible OLEDs and enhancing their bending characteristics through mechanical analysis, *RSC Adv.* 6, pp. 40835-40843 (2016).
- 2 J. Lewis, Material challenge for flexible organic devices, *Mater. Today* 9, pp. 38-45 (2006).
- 3 S. Kim, H.-J. Kwon, S. Lee, H. Shim, Y. Chun, W. Choi, J. Kwack, D. Han, M. S. Song, S. Kim, S. Mohammadi, I. S. Kee, S. Y. Lee, Low-power flexible organic light-emitting diode display device, *Adv. Mater.* 23, pp. (2011) 3511-3516.
- 4 B. Hwang, S. Lim, M. Park, S. M. Han, Neutral plane control by using polymer/graphene flake composites for flexible displays, *RSC Adv.* 7, pp. 8186-8191 (2017).
- 5 S.-W. Seo, H. Chae, S. J. Seo, H. K. Chung, S. M. Cho, Extremely bendable thin-film encapsulation of organic light-emitting diodes, *Appl. Phys. Lett.* 102, pp. 161908 (2013).
- 6 P. F. Carcia, R. S. McLean, M. H. Reilly, M. D. Groner, S. M. George, Ca test of  $\text{Al}_2\text{O}_3$  gas diffusion barriers grown by atomic layer deposition on polymers, *Appl. Phys. Lett.* 89, pp. 031915 (2006)
- 7 M.-H. Park, J.-Y. Kim, T.-H. Han, T.-S. Kim, H. Kim, T.-W. Lee, Flexible lamination encapsulation, *Adv. Mater.* 27, pp. 4308-4314 (2015).
- 8 S. Li, Y. Su, R. Li, Splitting of the neutral mechanical plane depends on the length of the multilayer structure of flexible electronics, *Proc. R. Soc. A.* 472, pp. 20160087 (2016).
- 9 Z. Suo, E. Y. Ma, H. Gleckova, S. Wagner, Mechanics of rollable and foldable film-on-foil electronics, *Appl. Phys. Lett.* 74, pp. 1177-1179 (1999).
- 10 C.-C. Lee, Y.-S. Shih, C.-S. Wu, C.-H. Tsai, S.-T. Yeh, Y.-H. Peng, K.-J. Chen, Development of robust flexible OLED encapsulations using simulated estimations and experimental validations, *J. Phys. D: Appl. Phys.* 45, pp. 275102 (2012).
- 11 Y.-F. Niu, S.-F. Liu, J.-Y. Chiou, C.-Y. Huang, Y.-W. Chiu, M.-H. Lai, Y.-W. Liu, Improving the flexibility of AMOLED display through modulating thickness of layer stack structure, *J. SID* 24, pp. 293-298 (2016).

- 12 H. Lee, J. Han, S. B. Ham, C. Jang, M. S. Huh, S. M. Cho, Folding stabilities of encapsulation layers at positions off the mechanical neutral plane, *Appl. Phys. Express* 11, pp. 086502 (2018).
- 13 W. Kim, I. Lee, D. Y. Kim, Y.-Y. Yu, H.-Y. Jung, S. Kwon, W. S. Park, T.-S. Kim, Controlled multiple neutral planes by low elastic modulus adhesive for flexible organic photovoltaics, *Nanotechnology* 28, pp. 194002 (2017).
- 14 J. H. Kwon, Y. Jeon, S. Choi, J. W. Park, H. Kim, K. C. Choi, Functional design of highly robust and flexible thin-film encapsulation composed of quasi-perfect sublayers for transparent, flexible displays, *ACS Appl. Mater. Interfaces* 9, pp. 43983-43992 (2017).
- [15] S. H. Jen, J. A. Bertrand, S. M. George, Critical tensile and compressive strains for cracking of Al<sub>2</sub>O<sub>3</sub> films grown by atomic layer deposition, *Appl. Phys. Lett.* 101, pp. 234103 (2012).