OLED Lighting Fabricated by Roll-to-Roll

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ABSTRACT

We have been developing OLED lighting by Roll-to-Roll. The substrates are plastic film and ultra-thin glass. For plastic films, a high gas barrier layer was deposited in the first step. In addition, we have developed new application products that take advantage of the thinness of the OLED.

1 Introduction

We have been developing OLED lighting by roll-to-roll [1], [2], [3]. The substrates are plastic film and ultra-thin glass. Plastic films are flexible and easy-to-handle substrates, but the gas barrier properties of plastic films are the most serious problem. On the other hand, ultra-thin glass has various advantages such as optical and bas barrier properties and surface smoothness, but its only drawback is that it is easily broken.

In this report, the barrier layer deposition process on plastic substrates by roll-to-roll and the fabrication process of OLED lighting by roll-to-roll will be introduced, and finally, fun products using OLED lighting will be introduced.

2 High Gas Barrier Film by Roll-to-Roll PECVD

2.1 Policy for Improving Barrier properties and standard for barrier measurement method

Since OLEDs do not emit when exposed to water vapor, a high gas barrier layer is required on the film. The water vapor barrier is usually evaluated using the water vapor transmission rate (WVTR), and ISO 15106-5 [4], 15106-6 [5], and 15106-7 [6] have been published as evaluation methods for high gas barrier films. However, in high-gas barrier films used in OLEDs, water vapor does not permeate through the barrier layer by diffusion, but through defects in the barrier layer. Therefore, it is more practical to measure barrier defects as an evaluation of barrier properties.

The standard established based on this idea is SEMI D78 [7]. In SEMI D78, calcium is deposited on the barrier film and the corrosion of calcium by water vapor passing through defects in the barrier layer is evaluated. The evaluation items are the corrosion area ratio and the number of defects (=corrosion number) per unit area, and these changes over time are the barrier properties. The WVTR can be calculated from the rate of change in corrosion area per day. We considered that improving barrier properties would reduce barrier defects. Then, based on this idea, the high gas barrier film was improved.

2.2 Deposition Process of Gas Barrier Layer on Film

The film was PEN or PET.

The deposition process on the film is as follows.

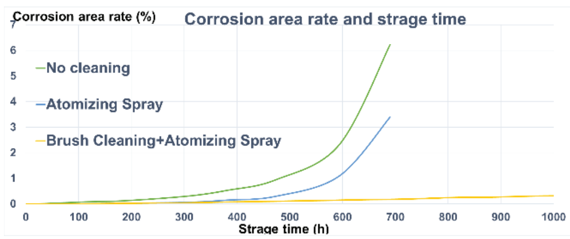
1) Clean the film with a wet cleaning equipment
2) Coat the planarization layer with slit die equipment
3) Clean the film with a wet cleaning equipment
4) Deposit barrier layer by PECVD
5) Deposit IZO layer by sputter

2.3 Results and discussion

This report describes barrier deposition technology on PEN film.

2.3.1 Clean the film

The film was washed with roll-to-roll wet cleaning equipment. This equipment has brush cleaning unit, atomizing spray cleaning unit, shower cleaning unit, air knife, and IR drying unit. We evaluated the difference in barrier properties due to cleaning of PEN film [8]. The steps were 2.2.1) and 2.2.4), the precursor was hexamethyldisiloxane (HMDSO), and the barrier layer thickness was about 600 nm. Evaluation of these barrier films using SEMI D78 showed that the cleaning effect of the brush was significant. The WVTR of the barrier film deposited with the barrier layer after cleaning with brush cleaning + atomizing spray was $1X10^{-5}$ g/(m^2・24h).



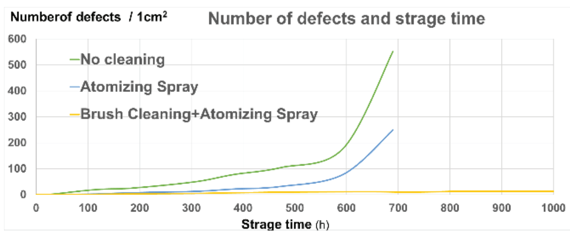
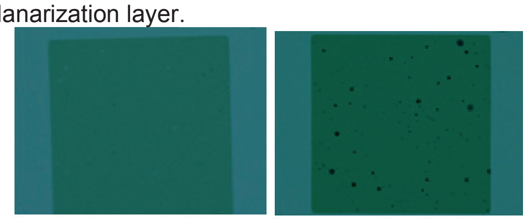


Fig.1 Relationship between cleaning and barrier properties

2.3.2 Planarization Layer and IZO

IZO was deposited on the film of 3.3.1. we thought that IZO would improve the barrier properties of the film since it is an inorganic oxide layer, but the result was the opposite. Barrier film sample with only barrier layer and barrier film sample with barrier layer and the IZO were evaluated by SEMI D78 [9]. The evaluation result of barrier films with or without IZO is shown in Fig.2. These samples were stored under 40 °C and 90 %RH for 595 hrs. It was confirmed that the defects of the barrier layer increased dramatically by the deposition of IZO. On the surface of PEN film before barrier layer deposition, small protrusions were observed. The height of the protrusions was only a few tens of nm, so they were sufficiently covered by the barrier layer, and the barrier properties were enough. We considered that the stress of IZO caused defects in the barrier layer at the protrusions, so we coated the organic planarization layer before depositing the barrier layer. As shown in the Fig.3, the barrier properties after IZO deposition were greatly improved due to the effect of the planarization layer.



Only barrier layer Barrier layer and IZO

Fig.2 Ca corrosion images of barrier films with or without IZO after 595 hours under 40 ° C and 90 %RH.

Fig.3 Ca corrosion images of planarized barrier films with IZO after 640 hours under 40 ° C and 90 %RH.

2.3.3 Structure of Barrier Layer

For the deposition of the barrier layer, PECVD roll coater manufactured by Kobe Steel was used. Schematic of PECVD roll coater is shown in Fig. 4.

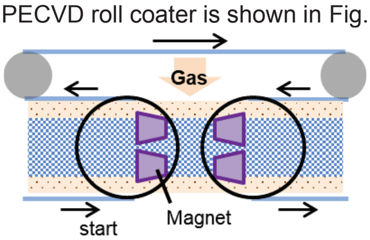


Fig.4 Schematic of deposition roll

The properties of the barrier layer depend on the deposition parameters [10]. The parameters are oxygen: HMDSO flow rate and ratio, pressure, input power, substrate transfer speed and number of passes. SiOx-based films were deposited by the oxidation of HMDSO, and the composition varied depending on the deposition conditions. In case of insufficient SiOx conversion, the barrier properties were not good and water vapor permeated through the barrier layer. When the conversion to SiOx was advanced, it became a barrier layer with many defects. The inorganic oxide layer is brittle, so the defects were caused by stresses in the roll-to-roll process. The cross section image and element mappings of the barrier layer with the best barrier properties is shown in Fig. 5. The multilayer structure having periodicity in the depth direction was observed in the barrier layer by TEM. As a remarkable point of this TEM image, stack of 8 layers per 1 pass was observed. Such a multilayer structure was important for improving barrier properties. Since our PECVD roll has two magnets, the precursor was converted to SiOx near the magnets. On the other hand, at the position away from the magnet, the conversion of the precursor to SiOx was not sufficient, and the C concentration in the barrier layer was high.

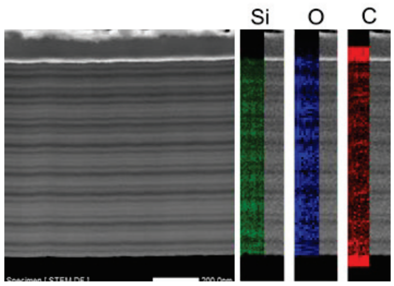


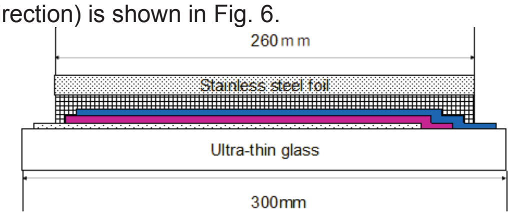
Fig. 5 Cross section of barrier layer using HMDSO as a precursor by roll-to-roll (Left: TEM imaging. Right: EDS element mapping.)

3 Roll-to-Roll Fabrication for OLED Lighting

Explaining OLED lighting fabricated by roll-to-roll at Yamagata University and Fraunhofer FEP [3]. The explanation will focus on the case of ultra-thin glass, but the process is the same for barrier film. The process of patterning electrodes on transparent electrodes (IZO) was also done in a roll-to-roll process at Yamagata University. Deposition of Organic and Cathode Layers and Encapsulation were fabricated in Fraunhofer FEP by R2R process.

3.1 Device Structure

The device structure (cross section in the width direction) is shown in Fig. 6.



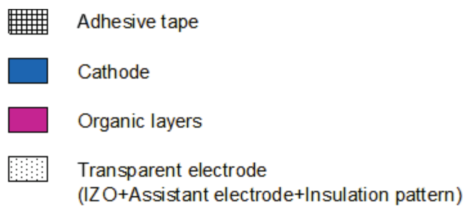


Fig. 6 Device Structure (cross section image)

3.2 Transparent Electrode

The first process was the deposition of IZO on ultra-thin glass. IZO was deposited on roll-to-roll sputtering machine. Next, IZO was etched using the etching paste. The etching paste was printed by special roll-to-roll screen printing machine and the paste solved the IZO. After the paste was washed off with water, the IZO pattern was completed. Large-area OLEDs did not emit uniform due to insufficient resistance of IZO. Therefore, assistant electrodes for current supply were required. The assistant electrode was printed by conductive ink. Finally, the insulation pattern was printed with insulating ink.

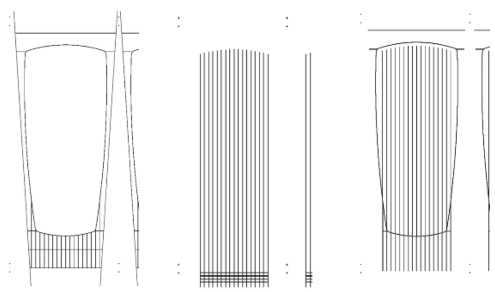


Fig. 7 IZO pattern Fig. 9 Insulation pattern
Fig. 8 Assistant electrode

3.3 Deposition of Organic and Cathode Layer, and Encapsulation

The OLED deposition was carried out in roll-to-roll vacuum equipment at Fraunhofer FEP. The thermal evaporation of the organic and metal cathode films was done continuously using organic linear sources and metal point source. Encapsulation process using stainless steel foil laminated with PSA film (Pressure Sensitive Adhesive) was done in one process using the substrate and interleaf winder in vacuum after the cathode deposition to prevent front-back side contact.

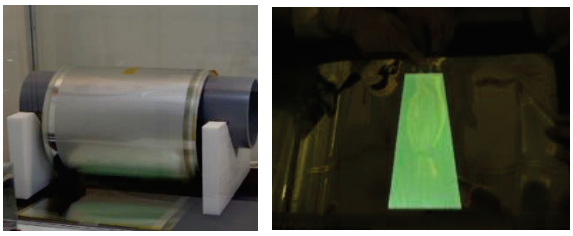


Fig. 10 Roll sample after encapsulation

3.4 Cutting and Improved Mechanical Durability of OLED Devices

The roll sample was roughly cut at a pitch of 320 mm so that two pieces (one set) of OLED devices would be placed on one sheet. This sheet was cut on the cut line indicated by the IZO pattern and divided into individual devices. First, the glass surface was scribed with a mechanical tool [11], then the sheet was inverted and the stainless steel foil surface was irradiated with a laser and then separated. The cut OLED device has exposed ultra-thin glass on both sides. This part was very fragile and needed to be handled with care. Two pieces of stainless steel foil were attached to the back side of the exposed ultra-thin glass for reinforcement.


Fig. 11 Divided OLED devices

To further improve the impact resistance of the ultra-thin glass, a special silicone gel (OPTαGEL) was laminated onto the glass [12]. The table shows the results of the drop impact test. When a ballpoint pen was used, cracks tended to occur at lower points than with versus stainless steel ball. When a ballpoint pen was used, cracks tended to occur at lower points than with a stainless steel ball. Although the ultra-thin glass did not crack by the 64 g stainless ball impact when OPTαGEL was installed.

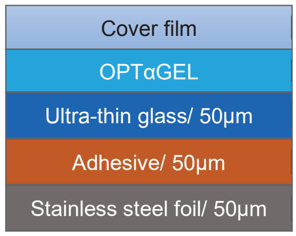


Fig.12 Configuration of the test sample

Table 1 Drop impact test result

Protection layer		Height glass cracked [cm]		
Cover film /Thickness [μm]	OPTαGEL Hardness /Thickness	Stainless ball 64 g	Stainless ball 230 g	Ballpoint pen
-	-	60~80	20~40	10
PET/100	-	90~100	60~80	20~50
PC/200	-	50~60	Passed	50~70
PET/100	n130/250	Passed	70	50~60
PC/200	n130/250	Passed	90	50~70
PC/200	n90/250	Passed	Passed	80
PC/200	n50/250	Passed	100	50~60
PC/200	n25/250	Passed	80~90	60

4 Fun product using OLED Lighting

The final goal of this technology is general lighting, but it is too expensive compared to LEDs. Therefore, we are developing fun products that take advantage of the thinness.

4.1 Fabrication process for design OLED Lighting

The process up to the IZO pattern process was the same as 3.2. After cutting the substrate, the process changed from roll to sheet. The insulating pattern was printed on the IZO according to the design by screen printing machine. The part of IZO without insulation pattern emitted light. By sharing the IZO pattern, it is possible to handle a wide variety of production.

4.2 Luminescent necktie in collaboration with traditional crafts

We will be able to introduce some fun products, but here we will describe the luminescent necktie.

Our section of Yamagata University is located in Yonezawa City, Yamagata Prefecture. In Yonezawa, there is a craft called “Harakata-sashiko”. “Sashiko” is a handicraft in which a geometric pattern or other design is embroidered with thread and sewn into a fabric. We combined "Sashiko" and OLED lighting on a tie. When switched on, a pattern of hemp leaves blinks repeatedly. The OLED lighting device can be easily removed from the necktie and the necktie is washable.

Fig. 13 Neckties fitted with OLED lighting

5 Conclusions

We have developed a roll-to-roll fabrication technology for OLED. The main process technologies were high gas barrier layer deposition technology on plastic films, IZO deposition and patterning technology on plastic films and ultra-thin glass, printed electrode technology for large area OLED, evaporation technology for organic layer and cathode metal, stainless steel lamination technology for encapsulation, cutting technology from roll to sheet, and protection technology for OLED elements. Furthermore, we are developing fun products.

6 Acknowledgement

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References

[1] T. Furukawa, K. Mitsugi, S. Akiyama, H. Itoh, D. Kobayashi, T. Suzuki, H. Kuroiwa, M. Sakakibara, K. Tanaka, N. Kawamura, M. Koden, “ITO Pattern Formed by Roll-to-Roll Process on Ultra-thin Glass”, IDW’14, FLX3-4L, pp 1428-1429 (2014).

[2] T. Furukawa, N. Kawamura, T. Noda, Y. Hasegawa, D. Kobayashi, M. Koden, ”Novel Roll-to-Roll Fabrication Processes of Transparent Electrodes on Ultra-Thin Glass”, IDW ’17, FLX6-2, pp. 1535-1538 (2017).

[3] T. Furukawa, J. Hauptmann, T. Nakagaki, R. Ikeuchi, M. Sagawa, D. Nagata, J. Nakatsuka, ” Roll-to-Roll Fabrication for OLED Lighting Using Ultra-Thin Glass Substrate and Encapsulating Stainless Steel Foil”, IDW ’21, FLX5/FMC6-1, pp. 949-951 (2021).

[4] ISO 15106-5:2015 Plastics - Film and sheeting - Determination of water vapor transmission rate – Part 5: Pressure sensor method

[5] ISO 15106-6:2015 Plastics - Film and sheeting - Determination of water vapor transmission rate - Part 6: Atmospheric pressure ionization mass spectrometer method

[6] ISO 15106-7:2015 Plastics - Film and sheeting - Determination of water vapor transmission rate - Part 7: Calcium corrosion method

[7] SEMI D78 - Test Method of Water Vapor Barrier Property for Plastic Films with High Barrier for Electronic Devices

[8] T. Furukawa, N. Kawamura, M. Koden, H. Itoh, H. Kuroiwa, K. Nagai, “Gas Barrier Film for OLED Devices”, LOPEC (2017)

[9] K. Taira, T. Furukawa, N. Kawamura, M. Koden, T. Takahashi, “High Gas Barrier Film for OLED”, IDW’17, FLXp1-8L, pp 1572- 1574 (2017).

[10] K. Taira, T. Suzuki, W. Konno, H. Chiba, H. Itoh, M. Koden, T. Takahashi, T. Furukawa, “Development of High Gas Barrier Film Using Novel Precursor by Roll to Roll PECVD”, IDW’18, FLX2 - 4L, pp 1492-1495(2018).

[11] T. Nakagaki, T. Kawabata, H. Takimoto, T. Furukawa, “Scribing Tool and Cutting Method for Ultra-thin Glass”, IDW ’19, FLXp1-9L, pp. 1638-1641 (2019).

[12] M. Natsuka, Y. Ono, H. Mataki, S. Usui, H. Suzuki, M. Abe, T. Furukawa, “Protection of OLED Lighting with Ultra-Thin Glass by Special Silicone Gel”, IDW ’21, FLX5/FMC6-2, pp. 952-955 (2021).