Present status of Roll-to-Roll Fabrication for OLED lighting

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Abstract

Roll-to-Roll OLED device manufacturing on flexible substrates will have high potential for lower fabrication costs. Different kind of flexible substrates: plastic barrier films, metal foils or ultrathin glass films open a huge variety of OLED lighting applications. However, the challenge for the roll-to-roll OLED fabrication is the defect generation caused by web handling and proper pre-treatments for removing the residual water. Plastic barrier films, metal foil and ultrathin glass web have their own advantages and disadvantages for roll-to-roll processing. This paper will outline the present status and will discuss how the gap for a robust and stable roll-to-roll OLED manufacturing on different kind of substrates can be closed.

Introduction of the Roll-to-Roll OLED line at Fraunhofer FEP

An attempt has been made to start OLED fabrication by roll-to-roll process in high vacuum to study the high potential of cost reduction in contrast to sheet-to-sheet fabrication [1], [2]. Major cost reducing factors will be high organic material yield determined by performance of wide scalable organic linear sources, low substrate costs and high throughput. Recently, the roll-to-roll mass production on barrier films have been launched [3].

The roll-to-roll infrastructure allows OLED developments starting from the uncoated web till full integrated OLED lighting systems. In Figure 1, the process flow of the roll-to-roll R&D prototype line is schematically outlined.

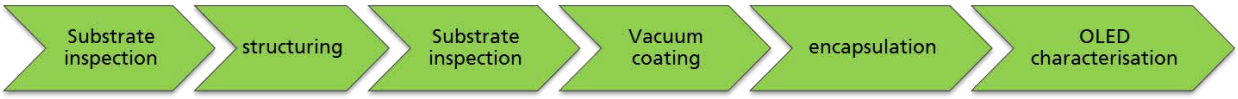

Figure 1: Process flow of the R2R prototype line from substrate inspection up to the final electro-optical characterisation of OLED devices.

For the deposition of small molecule OLED devices unique vacuum coater is equipped with 14 organic linear evaporators with alternating single and co-evaporation sources. This allows highly efficient OLED stacks, for e.g. p-i-n OLED technology [4], as seen in Figure 2.

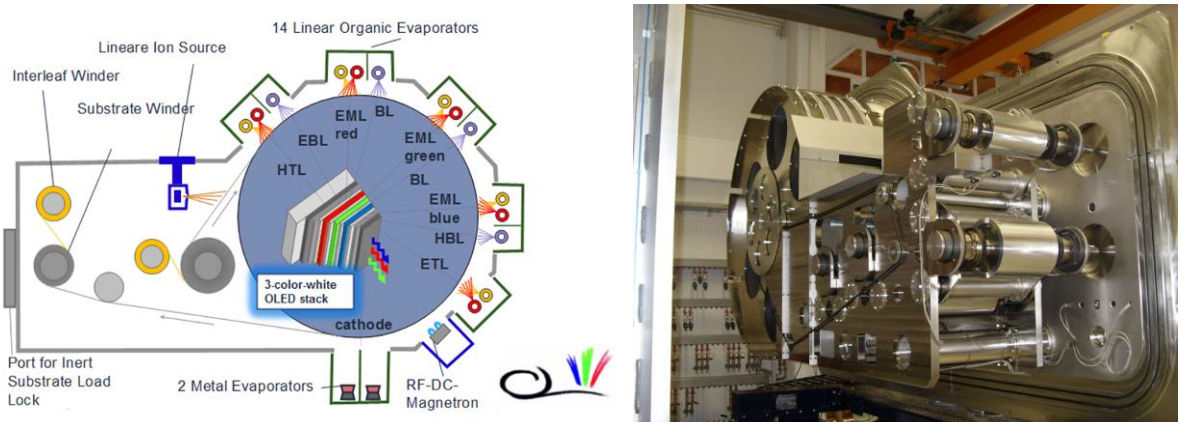

Figure 2: On the left hand side a schematic drawing of the R2R vacuum coater and the indication of the white p-i-n OLED stack processing which requires 14 organic linear evaporators is depicted. On the right hand side shows a photograph of the substrate winding unit and the deposition drum of the R2R vacuum coater with winders for substrate and protective liner foil to minimize substrate damage after the OLED deposition process.

After the OLED coating process the coated coils are extremely sensitive to moisture. Therefore, the OLED coated coil must be transferred and encapsulated under N_2 protective environment.

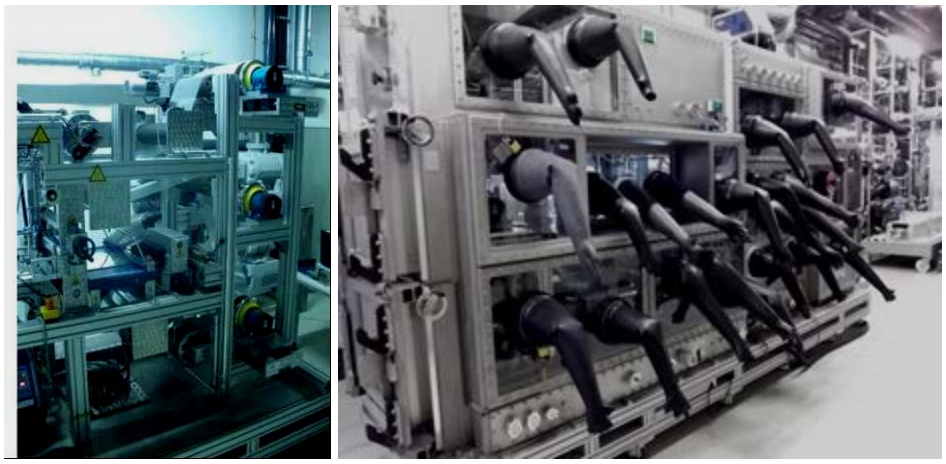

Figure 3: Photograph of R2R lamination system (on the left) which is encased in an inert box at Fraunhofer FEP (on the right).

Figure 3 shows the photograph of the roll-to-roll lamination tool for OLED encapsulation which operates in a protective nitrogen environment with H_2O and O_2 of < 10 ppm, respectively. This tool is directly linked to coating and curing units to allow developments of the encapsulation technology by barrier film lamination.

In general, the processed roll is monitored by optical defect inspection starting with the pre-delivery check up till to the backtracking of the OLED device parameters. Figure 4 shows the roll-to-roll inspection system encased in a clean room carbine. The inspection system consists a winding unit and an inspection table. Over the inspection table a 100% inspection with CCD line scan camera and a moveable optical microscope is performed.

Figure 4: Roll-to-Roll inspection system encased in a clean room carbine ISO6.

The following inspection modes are possible and are illustrated in Figure 5:

(A) A 100% inspection with CCD line scan camera with top and bottom light source for the detection of surface defects and non-topographic defects in transparent films, respectively. A pixel resolution of 14 µm is possible (≈ 40 µm particle resolution).
(B) Defect review after the 100% inspection using the moveable optical microscope is possible to analyze defects of interests.
(C) Automatic image recording on a homogenous web with the moveable microscope based on a before defined xy position table. In the recorded images defects with a point resolution of 1µm (depend on the applied objective in the microscope) can be displayed. The following binarization of the recorded images by filtering the threshold of pixel grey values. This allows a defect spot counting with a determination of the defect density and bin size distribution.
(D) Automatic recording of patterned web to recognize pattern defects or the quality of printed layers.

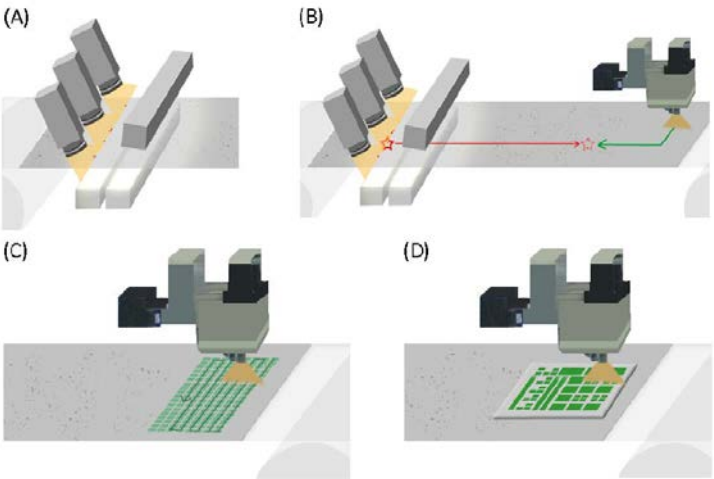

Figure 5: Overview of inspection possibilities of the R2R inspection system. (A) 100% inspection, (B) Defect review to specify further defects of interest, (C) Spot counting mode to determine the defect density on homogenous webs with a defect resolution far below the 100% inspection of 14 µm and (D) Quality control of patterned web and substrates.

Potential and challenges for the different kind of flexible substrates

Different kind of flexible substrates for roll-to-roll processing have in particular their advantages and disadvantages regarding, barrier properties, surface quality and web handling issues. In Table 1 summarize the substrate properties which have been evaluated for roll-to-roll OLED fabrication.

	metal	ultra-thin glass	plastic
bendability	o	o	√
permeation barrier	√	√	o
roll-to-roll process-ability	√	(√)	√
surface roughness	o	√	√
cost	√	o	o
advantages	good barrier thermal conductivity	good barrier surface quality transparency	transparency high bendability
disadvantages	top emission additional treatment of reducing surface roughness	brittle device separation	barrier coating pin-holes thermal stability residual water possible pinholes

Table 1: Comparison of different kind of substrates which have been evaluated for roll-to-roll OLED fabrication.

Metal foil seems to be suitable candidate, because they already fulfill one important requirement as a substrate for OLEDs regarding impenetrability to water and oxygen. However, for the top-emitting devices the emitted light is strongly affected by the micro cavity effects and this results in high angular dependency of the out-coupled light [5]. Therefore, there is a growing interest to perform the roll-to-roll OLED coating on transparent substrate for highly efficient bottom-emitting devices. A challenge for flexible barrier film encapsulation is to guarantee sufficient barrier properties against water and oxygen over several 100 meters of roll materials. At present semi-commercial barrier films on rolls (large area coatings) are available on the market which could already meet the requirements for OLEDs with low life time specification, but still the WVTR is 2 order of magnitude higher than in case of lab scale samples [6]. Therefore, a great potential could be the roll-to-roll manufacturing on ultra-thin glass (50 – 200 µm). Because for flexible glass no further barrier film coatings are needed to reach the required barrier requirements for long lifetime OLED devices. Especially, flexible glass is suitable for sputtering of conductive ITO layers by applying a temperature above 200°C in order to reach highly conductive TCO layers with optical transparency > 90 %. However for winding of ultra-thin glass much more attention needs to be drawn for the equipment and a big challenge is to guarantee for the edge stability of the glass without any initial micro cracks over several 100 meters on coils.

OLED device properties on different kind of substrates

For stable roll-to-roll OLED device manufacturing a very important entry requirement is the surface quality of the flexible substrate, including the coated electrode. The root-mean-square roughness of ≈ 1 nm and peak to valley roughness of ≈ 10 nm for ITO coated glass has been published for needed surface requirements and it has been pointed out that the leakage will be influenced mainly by the peak to valley roughness value [7].

As an alternative, the surface roughness can be smoothed by thicker hole transport layer (HTL) to a 2nd or higher light out-coupling maximum. Figure 6 illustrates the smoothing effect by increasing the HTL layer from 1st to a 2nd maximum which results in factor 10 lower leakage current. However, the overall OLED stack thickness is in the order of several hundred nm and makes the device still sensitive against particle contaminations.

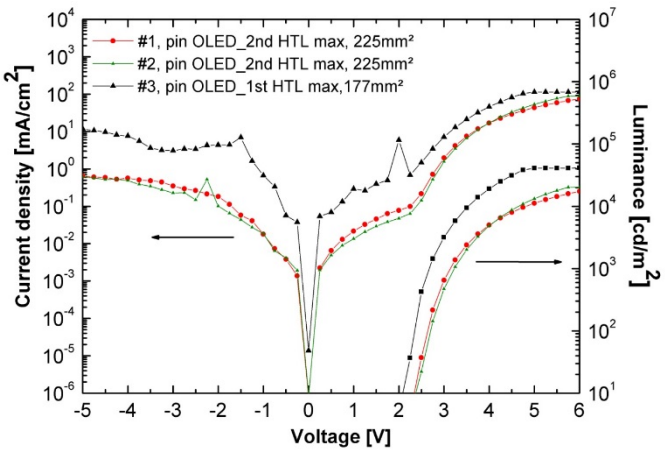

Figure 6: Current-voltage characteristics of 177 mm² and 225 mm² OLED areas on metal substrate. The leakage current can be reduced by an order of magnitude to increase the HTL layer thickness from the 1st to the 2nd maximum.

Therefore, particles with a size down to 0.2 -1 µm will negatively affect the OLED power efficacy and leakage current. Particle levels on substrates and defect level controls during the OLED fabrication process are essential to stabilize and improve the yield for a given window of OLED device behavior, different types of particle contaminations have different consequences [8]. A local thinning and local electric field concentrations in the organic layers can result in higher leakage currents.

The defect level have been monitored for different kind of available plastic barrier films. As illustrated in Figure 7, the defect levels for different barrier films and the defects level on the same roll at different running meter show a high fluctuation of the defect density (defect resolution ≈ 4 µm).

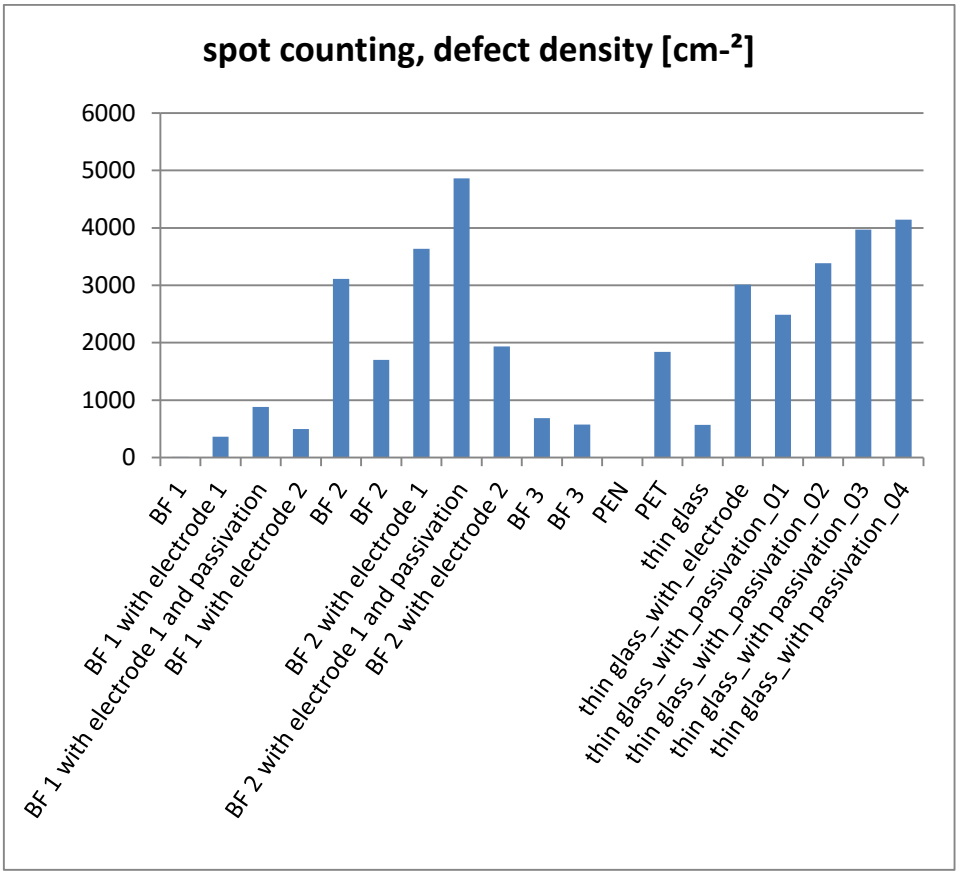

Figure 7: The diagram shows the defect density in 1/cm² level for different barrier films determined as described in Figure 5 (C). It is obvious that the defect level varies significantly at a deterrent running meter position on the same roll and on different coils.

After the roll-to-roll OLED run the devices have been characterized by measuring the current-voltage behavior on 10 x 10 mm² test OLED structure. The leakage current of OLED devices is evaluated over a length of 15 m to gain enough statistics. The comparison of metal substrate with plastic barrier film reveal that a fabrication of stable OLED devices is possible on both kind of substrates to reach sufficient low leakage currents of 10^{-4} mA/cm^2, as shown in Figure 9. However, the value for the leakage current varies from 10^{-1} – 10^{-4} mA/cm² for plastic barrier film and 10 – 10^{-4} mA/cm² for metal foil. This derives a high fluctuation of the surface quality of the substrate. Additionally, as shown in Figure 8 (C), the exclusion of the web handling during the roll-to-roll coating process results in significant increasing amount of stable OLED devices. This means that the winding process is important and have a high impact getting a high yield of stable OLED devices.

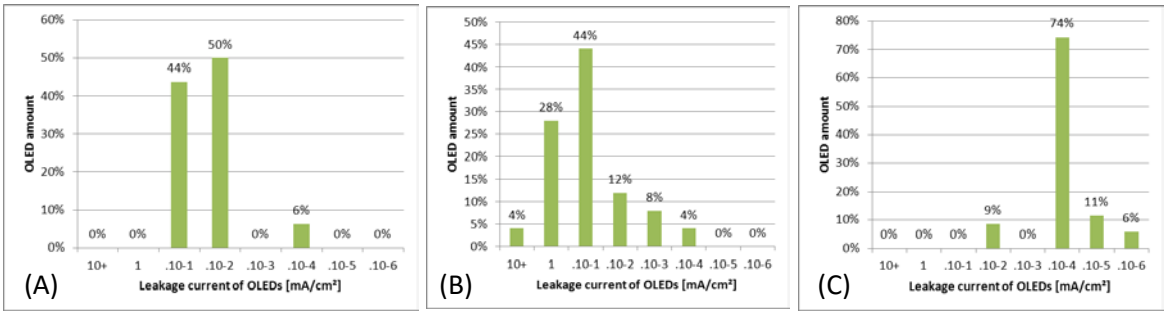

Figure 8: A comparison of the leakage current distribution from OLED devices fabricated in a roll-to-roll process on metal substrate (A), plastic barrier film (B) and on plastic barrier film sheets, excluding possible web handling issues (C).

OLED devices on ultra-thin flexible glass, encapsulated with ultra-thin flexible glass

Plastic foils for OLED substrates require high barrier layers to protect the devices from water. These layers are very sensitive and defects caused by roll-to-roll process reduce the barrier properties. These defects can also accelerate degradation or release short circuits. In contrast, ultra-thin glass substrates are excellent barriers for water transmission with a high light transmission. To avoid cracks, the glass substrate have higher demands for winding accuracy than plastic films, but still the barrier itself is more robust for roll-to-roll processes than high barrier stacks on plastic foils. Another advantage of thin glass is high surface smoothness which allows to reduce defects in deposited layers above. Fraunhofer FEP has successfully applied OLEDs on flexible ultra-thin glass which are then encapsulated with plastic barrier foils. Further Fraunhofer FEP succeeded also to encapsulate the ultra-thin glass devices using an additional thin glass foil laminate in a single manufacturing step – all in a continuous roll-to-roll process.

Figure 9: The photograph shows two 250 x 100 mm² working OLED devices on ultra-thin flexible glass (G-leaf®) encapsulated with ultra-thin flexible glass (G-leaf®).

To ensure process ability 300 mm wide and 50 µm thick ultra-thin flexible glass was laminated on a PET foil, width 310 mm and thickness 75 µm, by Nippon Electric Glass. An IMI electrode was deposited and structured by printing of a passivation layer. A single unit white OLED stack was evaporated, OLED encapsulation was realized by 250 mm wide and 125 µm tick glass/PET foil laminate. Encapsulation is enabled through a high-performance adhesive, which was applied over the entire surface of the encapsulating glass laminate.

Conclusions and Outlook

The OLED roll-to-roll fabrication line allows stable deposition of OLED devices on metal, plastic barrier film and ultra-thin glass substrates. At present 15 m roll-to-roll OLED coating campaigns are in focus to evaluate the OLED device behavior on different kind of substrates. A 15 m roll of working OLED devices on 50 µm ultra-thin glass have been demonstrated, as seen in Figure 9 and Figure 10 with a minimum of dark spot nucleation.

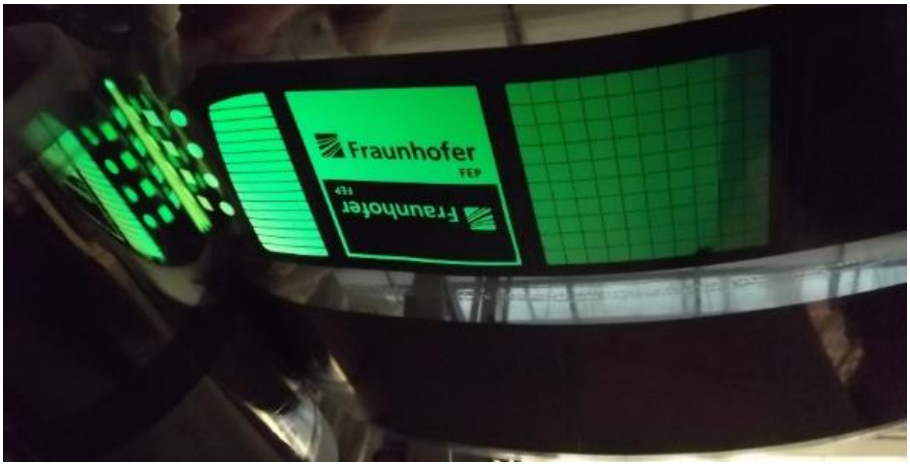

Figure 10: The photograph shows 100 x 100 mm² working OLED devices on a several meter long and 300 mm wide OLED demonstrator on 50 µm ultra-thin glass.

For the future work a several developments and evaluations are in focus:

- The overall quality of flexible substrates must be guaranteed over meters on coils.
- For plastic barrier film a very low pinhole density is required to minimize darkspot growth on large area flexible OLED devices or large area OLED displays.
- Minimization of the substrate damage during the winding process to minimize the leakage current and in particular for plastic barrier film no damage of the ultra-high barrier properties.
- An efficient roll-to-roll drying process or dry storage concepts for roll materials is essential to allow the introduction of roll-to-roll OLED production.
- Damage of barrier film layers during TCO coating needs to be further understood and optimized. Ultra-thin glass could be a promising alternative to plastic barrier film to maintain the ultra-high barrier properties after several windings.
- Stable and reliable electrical contacts with low contact resistance on flexible substrates.

The most critical issue for getting high OLED lifetime on flexible substrate is the nucleation of

dark spots. Those dark spot growth can have different root causes and are present under investigation. It is important to correlate the defect level from the substrate to the dark spot nucleation to gain a better understanding of yield relevant defects. Unfortunately, the pin holes in plastic barrier films cannot be directly monitored by optical inspection methods. For determination of pinhole densities a calcium tests on large areas needs to be further promoted.

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