50.4: First Hybrid OLED by Organic Vapor Phase Deposition and Its Advantages in Deposition Rate Control for OLED Manufacturing

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Abstract

We are discussing the latest results in the field of Organic Vapor Phase Deposition (OVPD) on basis of our experimental data. In particular the use of carrier gas and its controlled mass flow are adding an additional parameter to control deposition rates in OLED manufacturing by OVPD. Many advantages offered by this key parameter in this technology like stable deposition rates, wide range of adjustable deposition rates and doping concentrations are discussed.

A small molecule hybrid OLED (SM-HLED) consisting of a polymer layer of PEDT:PSS and two small molecule layers of α -NPD and Alq₃ was fabricated by OVPD. For this 3-layer device we observed a turn on voltage of 2.5 V, a luminance brightness of 100 cd/m² at 5.2 V and 300 cd/m² at 6.5 V. Furthermore a passive matrix OLED-display (PMOLED-display) was demonstrated by OVPD. The display had a turn-on voltage of about 3.0 V and showed a very homogeneous light emission.

These device characteristics confirm that OVPD is close to be comparable to Vacuum Thermal Evaporation (VTE).

1. Objective and Background

Figure 1 shows the schematic of OVPD production scale technology using a hot wall deposition chamber equipped with a showerhead. AIXTRON has combined the OVPD technology with its proprietary close coupled showerhead (CCS) and offers a single deposition chamber for multi-layer deposition. The gas phase transport allows to place all organic sources in an external position with respect to the deposition chamber thus the design guarantees no cross contamination of the source material.



Figure 1: Close Coupled Shower Head OVPD-Technology for Industrial Scale Deposition

The hot-walled deposition chamber prevents condensation of the organic material thus leading to low maintenance cycles important for industrial mass production.

Compared to other deposition technologies like Vacuum Thermal Evaporation the OVPD offers essential advantages like high material yield, well defined doping with multiple dopants and cost saving lower downtime and higher throughput.

2. Experimental Results

2.1. General Conditions

We report on process results achieved with OVPD equipment. Prior to the design of the equipment numerical CFD (Computational Fluid Dynamics)-modeling built up the basis for the actual blueprint design. We reported on the computational modeling of the uniform and highly efficient deposition of organic thin films as well as on the thermal management of the deposition chamber and the OVPD technology itself earlier [1].

All organic materials were transported in nitrogen as carrier gas and condensed on substrate that was actively cooled and temperature controlled. Evaporation of organic materials like Tris-(8-hydroxyquinoline)-aluminum(III) (Alq₃) and N,N'-Di-[(1naphthyl)-N,N'-diphenyl]-(1,1'-biphenyl)-4,4'-diamine (α -NPD) from specially designed source containers took place at temperatures between 200-400 °C. Layers were deposited on 6 x 6 inch glass substrates.

Organic material was characterized by High Performance Liquid Chromatography (HPLC) to ensure that no decomposition of organic material takes place during processing by OVPD. Figure 2 shows the comparison of HPLC spectra of the α -NPD material prior to loading and the same material deposited by OVPD. As both spectra are identical no evidence for decomposition was found. This method and photoluminescence experiments were used to ensure working below the decomposition temperature of the organic materials, which is critical to high performance OLED devices.

2.2.Advantageous deposition control by OVPD

For the design of state of the art organic devices the deposition rate control is both critical to thickness and doping concentrations. The use of nitrogen as carrier gas is adding a beneficial parameter in deposition rate control by mass flow controller (MFC) as discussed elsewhere [2], [3], [4].



Figure 2: HPLC of a-NPD as loaded vs. as deposited by OVPD

In order to demonstrate the controllability of the OVPD process we have introduced a quartz crystal monitor into an OVPD chamber in order to measure the deposition rate in-situ. A constant carrier gas flow through an Alq₃ source was adjusted targeting at a deposition rate of 12.5 Å/s and the material flow into the deposition chamber was switched on and off several times. For demonstration of the long-term stability for deposition of thick bulk layers the first deposition was done for 120 seconds. For verification of the reproducibility six subsequent layers were then deposited for 20 seconds each with 15 seconds interruption between the deposition processes.



Figure 3: Deposition Rate of Alq₃ vs. time for two test deposition runs

In Figure 3 the results from two test deposition runs are displayed, demonstrating the advantages of the mass flow control of the carrier gas: the average deposition rates of the runs are 12.27 Å/s and 12.28 Å/s, respectively, which is within 1.8 % of the target value. In addition both runs show excellent stability of the deposition rate, i.e. within the bulk layer, for the full sequence as well as run to run. The overall variation of deposition rate is between 12.22 Å/s and 12.30 Å/s, a stability of \pm 0.32 %, which is outstanding in comparison to known data from conventional deposition rate demonstrate the superior features of OVPD for realizing OLED structures as they become more complex.

Another important factor is the high precision switching of the source material. The two test runs displayed in Figure 3 show the excellent switching behavior: all sequences show very steep on/off edges, which is crucial for realizing sharp interfaces and run to run reproducibility. The excellent match of both sequences further demonstrates the superior reproducibility of the gas phase control used in OVPD.

The demonstration of reproducible deposition rates higher then 10 Å/s by OVPD are proving that this technology shall enable OLED manufacturing with low deposition times thus lowering tact times and cost in volume manufacturing.

2.3. Fabrication of Hybrid OLEDs by OVPD

A hybrid OLED (SM-HLED) consisting of a polymer layer of PEDT:PSS and two small molecule layers of α -NPD and Alq₃ was fabricated by OVPD at TU Braunschweig.

Figure 4 shows the schematic of the layer sequence. On an indium-tin-oxide (ITO) coated glass substrate a layer of PEDT:PSS (Baytron P AI 4083 from H.C.Starck GmbH, Germany) was prepared by spin coating.



Figure 4: Schematic of the SM-HLED structure deposited by OVPD at TU Braunschweig

After transferring the substrate to the AIXTRON OVPD equipment the α -NPD and Alq₃ films were deposited. Finally we evaporated magnesium and silver on top of the organic films in an attached metal deposition chamber without breaking the vacuum.



Figure 5: Current density-voltage characteristic of the SM-HLED.

Electrical and optical characterization was performed on the device: Figure 5 displays the current density-voltage characteristic of the SM-HLED. We observed a turn-on voltage of 2.5 V, a brightness of 100 cd/m^2 at 5.2 V and 300 cd/m^2 at 6.5 V. The

corresponding luminance-voltage characteristic is shown in Figure 6. The characteristics of this first SM-HLED processed by OVPD are promising and comparable to SM-HLED realized with VTE [5].



Figure 6: Luminance vs. Voltage of the OLED

Figure 7 presents the power efficiency of the SM-HLED device. A maximum efficiency of 1.8 lm/W is observed and about 1.4 lm/W were reached at 100 cd/m^2 .



Figure 7: Power efficiency vs luminance for the SM-HLED. (1.4 lm/W @ 100 cd/m²).

Figure 8 displays the current efficiency-luminance characteristic of the SM-HLED. Up to 1000 cd/m^2 were reached with a current efficiency of 2.3 cd/A. The efficiencies for this SM-HLED are comparable to VTE processed devices.

Based on these results for the SM-HLED, we were also able to demonstrate a first large-area OVPD-deposited-OLED displaying the Logo of the University of Braunschweig. The structure deposited is shown in Figure 9.

The ITO substrate was structured by a photolithographic process. For this demonstrator the same parameters were used for the OVPD deposition process to realize the same thickness for the α -NPD (40 nm) and Alq₃ (60 nm) films as for the SM-HLED discussed above. Following the deposition of the organic films the metal layers were deposited accordingly.



Figure 8: Current efficiency-luminance characteristic of the SM-HLED. A steady current efficiency of 2.3 cd/A was achieved up to 1000 cd/m2.



Figure 9: Structure of the large area OVPD-OLED device

The large area OVPD-deposited-OLED with the contacted electrodes is displayed in operation in Figure 10. We observed a homogeneous light emission all over the substrate, with its dimension of $35 \times 50 \text{ mm}^2$. The homogeneous brightness of this logo can be attributed to the superior uniformities with better than 1 % std. dev. achieved by the OVPD process for each film [6].

The next level of process development after demonstration a large area OLED was consequently a low information content (LIC) passive matrix display. The three-step photolithographic process described here is similar to that in [7]. The first photolithographic step defines the ITO anode rows of the display. After the etching process, the remaining photo resist is cleaned off by a solvent. In the next photolithographic step the active areas of the pixels are defined. The third photolithographic step comprises the manufacturing of the cathode separators. For these purposes photo resist barriers with a distinct undercut are applied on top of the passivation, orthogonal to the ITO-rows.



Figure 10: First large area OVPD-OLED displaying the Logo of TU Braunschweig processed on a substrate size of 35 x 50 mm².

After another cleaning process and ozone treatment, the substrate was transferred to the AIXTRON OVPD system. Again two organic layers consisting of α -NPD (40 nm) and Alq₃ (60 nm) were deposited onto the display substrate and the metal layers were evaporated without breaking the vacuum.

The PMOLED display showed a turn-on voltage of 3.0 V and a very homogeneous light emission. The display showed no defects, all pixels were individually addressable. Currently lifetime measurements are under investigations.

3. Summary and Conclusion

In this paper we presented experimental results on the outstanding performance of the OVPD process for deposition of organic materials. Our investigations by HPLC demonstrated the high quality of the deposited organic layers. Deposition rates as high as 12.3 Å/s with excellent reproducibility of better then 0.32% as well as excellent control of thickness and interfaces were shown. A first SM-HLED and a first passive matrix OLED-display (PMOLED-display) were deposited by OVPD and show device characteristics, which are comparable to devices grown by VTE. The excellent uniformity of the OVPD process was demonstrated by a large area OLED device and a passive matrix OLED display, which is demonstrating the vast potential of the OVPD technology with respect to volume manufacturing of OLEDs.

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