OVPD[®] Technology

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Abstract. The precise control of organic thin film processing by Organic Vapor Phase Deposition (OVPD[®]) is presented and analyzed on device level. With its decoupled process parameters, e.g. deposition rate, substrate temperature and deposition pressure, OVPD offers accurate and individual control of deposition layer properties like mixing several materials (co-deposition) and the realization of various morphologies. The benefit of precise co-deposition is demonstrated by an OLED with a sensitive twofold-doped emissive layer and revealed a doping level of level of 0.26% for the red dopant with a std. dev. of 0.38%. The effect of the various morphologies is investigated by optimizing the efficiency of molecular organic solar cells consisting of copper phthalocyanine (CuPc) and C₆₀. With defined process parameters efficiencies of up to 3.0 % were demonstrated.

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1. Introduction

In the year 2000 AIXTRON licensed the OVPD Technology and combined it with its close coupled Showerhead Technology to enable the equipment development for low-cost large-area manufacturing of organic electronic devices like OLEDs for display or lighting application and organic photovoltaics [1-4]. The dry and precise gas phase transport offers a high purity process technology which is essential for reproducible mass production [2]. Additionally, the easily scalable close couple showerhead enables the large area deposition with high material utilization and minimized parasitic condensation [5]. As a member of the EU- and government funded projects OLLA and OPAL 2008, AIXTRON develops various OVPD Gen 4 equipment for the manufacturing of white-light OLEDs [6,7]. Besides this development of OVPD production equipment, AIXTRON observed the ongoing research in the field of organic electronics and started additionally the development of a novel OVPD R&D module with modular sources for highest process flexibility [8]. Here we present the results based on OVPD equipment and its application in OLEDs and organic photovoltaics (OPV).

2. Codeposition

The precise controllability of the organic deposition step is realized by mass flow controllers. Figure 1 show various Alq₃ deposition rates which are realized at steady container temperature by varying only the amount of carrier gas. Deposition rates in the range of 1.8 to 16 Å/s were demonstrated by adjusting the source flow between 50 sccm and 500 sccm. Noteworthy is here the nearly digital on/off performance of the organic deposition, which is essential for precise interfaces and also offers the potential for gradual doping and co-deposition by mixing multiple source flows.

Figure 1: Alq₃ deposition rate as a function of source flow.

For the purpose of investigating the precision of the co-deposition of a multi component film an OLED device structure with an emission layer (EML) consisting of one host and two dopants, a green dopant (GD) and a red dopant (RD) was chosen. The concentration of the red dopant was varied within a range of 0.1-0.8 % and the device was characterized by electroluminescence (EL) measurements.

Figure 2: EL of OLED with varied doping concentration.

Figure 2 shows EL-Spectra of OLED devices. The EL-spectrum is characterized by two dominant peaks: The first peak at 520 nm corresponds to the emission of the green dopant, the second peak corresponds to the emission of the red dopant. The spectra were normalized to the maximum emission peak of the red dopant. As shown in Figure 3 the relative peak intensity GD:RD is sensitive to the doping concentration of the red dopant and decreases with increasing doping concentration.

Figure 3: Dependency of relative peak intensity to doping concentration.

For the investigation of the run-to-run reproducibility and controllability of the sensitive doping concentration a fixed doping concentration of the red dopant was chosen. The device was manufactured eight times at identical OVPD® process conditions. As the result Figure 4 shows 8 nearly identical EL spectra indicating an excellent reproducibility on OLED device level. Calculations based on calibration curves revealed an doping level of 0.26% for the RD with an std. dev. of 0.38% (Figure 5).

Figure 4: Reproducibility of EL-Spectra of 8 OLED devices.

Figure 5: Reproducibility of doping level.

3. Morphology

The precise controllability of the deposition rate in OVPD[®] shown above is also essential for the control of the layer morphology. In OVPD[®] the deposition rate, substrate temperature and deposition pressure are decoupled process parameters and allow realizing different textures and morphologies. Therefore the OVPD[®] technology offers manifold optimization potential to identify the ideal process conditions for defined interfaces, interpenetrating networks or other physical properties [9-11]. We investigated the morphology of CuPc-layers in dependence of the OVPD[®] process parameters like deposition temperature and deposition rates and observed drastically different results. Fig 6 shows scanning electron microscopy (SEM) images of CuPc films on <100> Si as function of substrate temperature and pressure that vary between 96°C and 187°C and 2.5 mbar and 0.3 mbar, respectively. Only for the samples 6d and 6e, grown at 131°C and 151°C in the pressure regime of 0.9 and 0.6 mbar a clear columnar growth of the CuPc crystallites is observed. As described elsewhere, we identified the columnar growth mode to be favorable for charge transport and consequently for high efficiencies [10].

4. Organic photovoltaics

To analyze these effects of precise co-deposition and controlled morphology we investigated simple ITO/PEDOT/CuPc:C₆₀/Mg/Ag bulk hetero-junction solar cells. We adjusted the source flows of CuPc and C₆₀ to generate a homogenously mixed absorber layer with the ratio of 1:1 at the substrate temperature of $T = 151^\circ\text{C}$ and a deposition pressure of 0.6 mbar.

With these optimized process parameters we were able to demonstrate small area OPV with efficiencies of up to 3.0% [11], an open circuit voltage $V_{oc} = 470 \text{ mV}$, a short-circuit current $J_{sc} = 15.1 \text{ mA/cm}^2$ and a fill factor $FF = 0.42$ (Table 1). By transferring these reproducible results to a larger OPV area we observed a drop in short-circuit current and, consequently, in efficiency (see Table 1 and Fig. 7). Lowering of the short-circuit current density is explained by an increased series resistance of the devices with larger area, related mainly to the geometry of the front contact [12].

Figure 6: Cross-section (60° view angle) and top view (insets) SEM images of CuPc layers deposited on Si <100>-oriented wafers at substrate temperatures and deposition pressures of (a) 96 °C and 2.5 mbar, (b) 111 °C and 1.5 mbar, (c) 120 °C and 1.2 mbar, (d) 131 °C and 0.9 mbar, (e) 151 °C and 0.6 mbar, and (f) 187 °C and 0.3 mbar, respectively and $T_{source}=411^\circ\text{C}$, $Q_{source}=400 \text{ sccm}$, and $Q_{total}=1050 \text{ sccm}$ [10].

Table 1: OPV Performance data of ITO/PEDOT/CuPc:C₆₀/Mg/Ag-OPV cells (AM 1.5)

Device	Area (cm ²)	Eff (%)	V _{oc} (mV)	J _{sc} (mA/cm ²)	FF
K01	0.06	3.0 [11]	470	15.1	0.42
I69	0.40	2.3	470	11.2	0.44

Figure 7: I-V characteristics of the OPV-devices in Table 1.

4. Conclusion

The carrier gas concept by OVPD enables easy and precise control of the organic deposition rate which is essential for reproducible processes with precise interfaces and co-deposition of several organic materials. By realizing a twofold-doped OLED with sensitive EL spectrum we were able to demonstrate a reproducible doping level of 0.26% on device level.

Further, with its decoupled process parameters, e.g. deposition rate, substrate temperature and deposition pressure, OVPD controls the texture and morphology of the deposited organic layer. The benefit of controlled morphology was demonstrated by investigating a small molecular organic solar cell. Here the optimized copper phthalocyanine morphology resulted in an organic solar cell efficiency of 3.0%.

The deeper understanding of the physical properties of the organic layers will identify further optimization potential for novel thin film devices.

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OVPD[®] is a registered trademark. OVPD[®] technology has been exclusively licensed to AIXTRON from Universal Display Corporation (UDC), Ewing, N.J. USA for equipment manufacture. OVPD[®] technology is based on an invention by Professor Stephen R. Forrest et al. at Princeton University, USA, which was exclusively licensed to UDC. AIXTRON and UDC have jointly developed and qualified OVPD[®] pre-production equipment.

6. References

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