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Toward inkjet printing of small molecule organic light emitting diodes

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ABSTRACT

Thermal evaporation is the current standard for the manufacture of small molecule organic light emitting diodes (smOLEDs), but it requires vacuum process, complicated shadow masks and is inefficient in material utilization, resulting in high cost of ownership. As an alternative, wet solution deposition can provide significant cost savings by enabling high-volume, large-area electronics on flexible substrates at low fabrication costs. In this report we present inkjet printing as a method to produce three active layers in a smOLED stack: a hole-injection layer, a hole transport layer and an emissive layer. The OLED lighting application sets high demands to a uniform light output over an area. This requires homogeneous deposition of the electro-active layers and this poses a significant challenge. OLED device efficiency is greatly influenced by the printed layer morphology and the quality of the deposited layers. Therefore inkjet processed smOLED device efficiency will be compared with reference devices made via vacuum deposition.

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1. Introduction

Unlike all other light sources, organic light emitting diodes (OLEDs) are flat and emit light over an area. OLEDs can form a large area diffuse light source with homogenous light emission. This creates an added value for indoor lighting. OLEDs with efficient light emission offer the benefits of low heat generation, high power efficiency and high color rendering index. It is expected that OLEDs will become an attractive alternative for the traditional lighting technologies such as fluorescent lighting [1].

High-efficiency (20 lm/W light emission with life time up to 10,000 h) OLED panels with an area of 55 cm² are now commercially available from Philips Lighting [2]. These products are manufactured through thermal evaporation and vacuum deposition of phosphorescent small molecules that yield much higher light emission efficiency compared to fluorescent polymer materials. However, the commercially available OLED lighting tiles and panels are still manufactured in a batch to batch way: in small quantities and sizes resulting in high cost of production. This is an important reason why the lighting market is still hardly penetrated by OLED lighting. Reduction of the OLED production costs is therefore essential for OLEDs to conquer a position in the lighting market.

Wet solution deposition can potentially provide an alternative to vacuum deposition techniques. Cost savings can be achieved by enabling high volume production of large-area electronics on flexible substrates [1,3]. For example, semiconducting organic materials, that comprise the active materials in OLED devices, are currently still very expensive and evaporation technology is not a very efficient way to use these materials. Solution processing in a roll-to-roll machine provides a continuous deposition method at atmospheric conditions that are much more efficient in material use [4]. One of the upcoming methods is inkjet printing of the electro-active layers. It is a contactless deposition technology, that can be used to deposit homogeneously patterned layers of a wide range of different materials. In this paper we report inkjet printing of smOLED materials, while avoiding usage of halogenated solvents.

The inkjet printing process has the advantage of being a non-contact technique that offers ease of patterning in various industrial processes and low costs of materials and equipment. Furthermore, the past few years have seen the development of reliable and robust inkjet printer heads/processes, making inkjet technology more and more mature for single-pass production printing [5]. This technological development opens many opportunities for using industrial inkjet technology in manufacturing of printed electronics including OLEDs [6–8] and thin film transistors (TFTs) [9].

Polymer OLEDs generally are less efficient (6–8 cd/A) [10] compared to their small-molecule counterparts (up to 84 cd/A reported in [11]). Over the last decades, there has been an intensive search for the polymer materials for light emitting diodes. However, the market growth of the polymer OLEDs has been limited due to issues with too low power efficiency and limited life time. On the other hand, a smOLED with phosphorescent emissive materials requires a multilayer structure to achieve its highest efficiencies [11]. It is difficult to produce multilayer devices using the solution-processing

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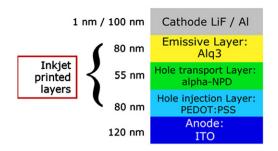


Fig. 1. Schematic representation of an inkjet printed smOLED device.

techniques commonly employed in the fabrication of polymer OLEDs, because the deposition of a second or third layer from solution enhances the risk of degradation of previously deposited layers [12,13].

In this contribution we demonstrate that OLED small molecule materials that are developed for conventional thermal evaporation can be deposited by inkjet printing. More specifically we present inkjet printing of small molecule materials for OLED application, while avoiding the use of halogenated solvents. OLED device efficiency is strongly influenced by the layer morphology and quality of the deposited layers and therefore various stacks of evaporated and inkjet printed layers are tested. Moreover, several requirements for the inkjet deposition of homogeneous electro-active layers are investigated and reported.

2. Experimental setup

2.1. Device stack deposition

SmoLED devices were fabricated on $30\times30~\text{mm}^2$ glass substrates coated with indium tin oxide (ITO, 20– $30~\Omega/\square$). The device consists of a multilayer stack on ITO, coated with a hole-injection layer (PEDOT:PSS, Poly(3,4-ethylenedioxythiophene) doped with poly(4-styrenesulfonic acid), a hole transport layer (α -NPD, N,N'-Di-[(1-naphthyl)-N,N'-diphenyl]-1,1'-biphenyl)-4,4'-diamine) and as an emitting layer (Alq3, Tris-(8-hydroxyquinoline)aluminum), with LiF/Al as the reflective cathode, as illustrated in Fig. 1.

Both PEDOT:PSS type Clevios™ CH8000 from Heraeus and Orgacon HIL1005 from Agfa have been used. The final purpose of high conductive Agfa Orgacon™ HIL1005 is to replace ITO as anode material [14]. Deposition of PEDOT:PSS was done by inkjet printing using a Spectra Galaxy PH 256/50 AAA inkjet printhead. The nozzle pitch is 254 µm, with a drop volume of approximately 50 pl. The lateral separation of neighboring printed lines can be varied by adjusting the azimuthal orientation of the printhead with respect to the direction of substrate motion. Generally, PEDOT:PSS patterns were printed such that the center-to-center distance of neighboring lines was smaller than the line width, guaranteeing sufficient overlap, in order to ensure homogenous layer formation [15]. Stable droplet formation without satellites is obtained by adjusting electric pulse and driving voltage for the printhead used [16].

The hole transport layer (α -NPD, CAS: 123-847-85-8, Sensient, ST16/7.S) was made from an ink composed of 5 mg α -NPD per ml of solution, that consists of a mixture of tetralin (1,2,3,4-tetrahydronaphthalene, Merck) and indane (Aldrich). The emissive material (Alq3, CAS: 2085-33-8, Sensient, ST 1095.S) was deposited with an ink composed of 10 mg Alq3 per ml of a mix of benzylalcohol (Aldrich) and THF

 Table 1

 Refractive index of PEDOT:PSS Agfa Orgacon HILHC4.

Wavelength	Refractive index n	Absorption factor k
455 nm	1.42	0.0264
530 nm	1.41	0.0225

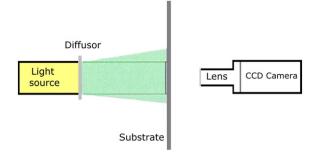


Fig. 2. A schematic outline of the experimental setup for the absorption method.

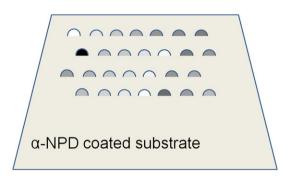


Fig. 3. Schematic of compatibility test of α -NPD, where various solvent drops are placed on an α -NPD coated substrate.

(Merck). Both materials were deposited using a Dimatix DMP2800 inkjet printer, with a 10 pl printhead. Evaporated layers of α -NPD and Alq3 were made by thermal evaporation.

2.2. Measurement of printed layer thickness

We have used the so-called "absorption method" as a contactless method to determine the dry layer thickness of PEDOT:PSS. The height of the PEDOT layer is determined by spatially resolved optical attenuation. The absorption (or attenuation) coefficient of this PEDOT:PSS has to be determined by measuring the complex refractive index (see Table 1).

The experimental setup consists of a light source, a diffuser, the substrate, a CCD camera and a telecentric lens, as shown in Fig. 2. The light source is a LED with a wavelength of 530 nm and a bandwidth of 30 nm. A diffuser is used to homogenize light intensity over the full substrate area. A part of the light from the source will be absorbed by the substrate and another by the PEDOT:PSS layer. The transmitted light is

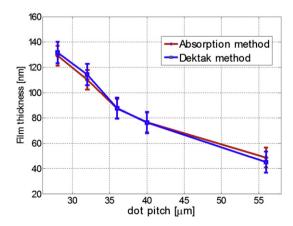


Fig. 4. Film thickness of inkjet printed PEDOT:PSS, type HILHC4, as function of dot pitch. Measurement using the absorption method compared to results determined with a stylus profilometer.

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