

High Resolution Ink-Jet Printed OLED for Display Applications

Daniel Walker¹, Hamish Leith¹, Lisa Duff¹, Li Wei Tan¹, Hsin-Rong Tseng², Thorsten Schenk², Peter Levermore²

1) Merck Chemicals Ltd, Chilworth Technical Centre, University Parkway, Southampton, SO16 7QD, UK
2) Merck KGaA, Frankfurter Strasse 250, Darmstadt, 64293, Germany

Abstract

We report performance of an inkjet printed OLED device suitable for a 55inch display at 4k2k resolution. Device efficiency, voltage, emission spectra and lifetime are presented and the effect of the uniformity of the printed layers on these parameters is discussed. The manuscript then goes on to discuss the readiness of inkjet print technology and the requirements of the inks for producing high resolution OLED displays.

Introduction

Inkjet printed (IJP) organic light-emitting diode (OLED) technology has accelerated dramatically in recent years, with IJP OLED performance metrics and manufacturing processes now approaching the requirements for large area displays [1, 2]. Here we examine in detail some of the features of inkjet printed devices that effect the final performance. Results from printed OLEDs are demonstrated with all materials in the device layers developed by Merck KGaA. Figure 1 shows the device stack for a green IJP OLED, although the results discussed here are equally applicable for the printed layers in red and blue devices. The hole injection layer (HIL), hole-transport layer (HTL) and green emissive layer (GEML) are printed. Subsequent device layers are evaporated through a common mask. These evaporated layers include the hole blocking layer (HBL), electron transport layer (ETL) and cathode.



Figure 1. Printed OLED Device Architecture. HIL, HTL and GEML are inkjet printed. HBL, ETL and Cathode are deposited via vapour deposition

In this manuscript we will present the performance of such an inkjet printed device and continue with a discussion on the effect the topography of the layers has on the device performance. These results will be extrapolated to a discussion on the salient issues for printing at higher resolutions.

Results

All devices were fabricated in full at Merck KGaA laboratories in Darmstadt, Germany. Bottom emission IJP substrates with 80ppi resolution were used as a platform, with each substrate having 4 individually addressable pixels. Each pixel is itself divided into an array of commonly addressable sub-pixels. Each sub pixel has an emissive area defined by a 212 μm long axis and a 64 μm short axis. Total emissive area for each pixel is 4.606 mm^2 . A resolution of 80ppi was selected because this corresponds to ultrahigh definition (UHD) resolution (4K2K) for 55" panel size. The target of this work is the large area (TV) OLED display market. Illuminated sub-pixels are shown in Figure 2. IJP substrates were cleaned with deionized water and dried with nitrogen. Substrate and bank design is such that no further pretreatment is necessary. Ink wets the ITO surface, but remains fully contained by the bank material. All IJP OLED inks (HIL, HTL, GEML) were printed in air, dried under vacuum to form device layers, and residual solvent removed by annealing. Two examples of devices are shown. These examples are printed with identical materials and processes, except the EML vacuum-drying pump-down profile which was altered resulting in a change to the dried film topography. The process flow was as follows:

HIL: Print in Air \rightarrow Dry in vacuum \rightarrow Anneal in Air

HTL: Print in Air \rightarrow Dry in vacuum \rightarrow Anneal in N_2

GEML: Print in Air \rightarrow Dry in vacuum \rightarrow Anneal in N_2

After processing the printed layers, devices were transferred to a vacuum thermal evaporator (VTE) chamber for vapour deposition of common layers, including HBL, ETL and aluminium cathode. Devices were encapsulated using a UV-curable epoxy edge seal. Electroluminescence (EL) spectra, current-voltage-luminance (UIL) and device lifetime were measured, with the results shown in Figure 3 and the data at 1000 cd/m^2 summarised in Table 1. For lifetime testing, initial luminance was set at 8000 cd/m^2 with an acceleration factor of 1.9. External quantum efficiency (EQE) data for IJP OLEDs is plotted against luminance in Figure 2(a) and EL spectra are shown in Figure 2(b), with the lifetime shown in Figure 2(c). Optical micrographs of the illuminated sub-pixels are shown in Figure 3. Figure 3(a) shows that very good emission uniformity can be achieved on the sub-pixel scale. Uniformity of emission indicates that the device layers are uniform and this results in strong device performance. Figure (b) demonstrates the common case of one or more of the layers being non-uniform. It clearly shows an uneven EL emission across the subpixel. Whilst the UIL results are largely unaffected, with both devices exhibiting similar performances with an EQE of 17.5 and 18.3%, a luminance efficiency of 64.1 and 66.9 cd/A and an operating voltage of 5.5 and 5.4V respectively, there is a dramatic influence on lifetime, with the LT95 of the non-uniform pixel being just 40% of that of the device with uniform sub-pixels. A non-uniformity in emission of this nature is dependent on the

topography of all the printed layers together, but most critically on the GEML. The bright emission is an indication of a higher current density in the EML at that point. As the EML typically has a higher resistivity than the hole-transport layers it is usually a sign of a thinner EML at the point of bright emission. This is demonstrated in Figure 4 by the profiles of the GEML layer of the subpixels shown in figure 2, taken across the short axis in the middle of the sub-pixel, with an Alphastep D600 stylus profilometer.

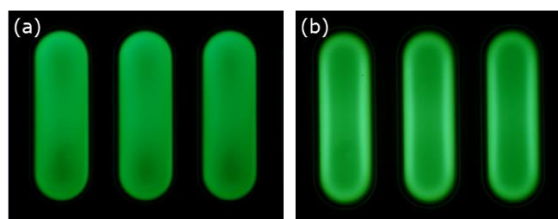


Figure 2. EL micrograph of inkjet printed OLED with (a) uniform layers. (b) non-uniform layers

Discussion

All the data presented in this work is for OLEDs at 80ppi, which is suitable for a 55" display with a 4k2k resolution, keeping in mind that each pixel contains three sub-pixels in a traditional RGB format. However, a number of companies, including Panasonic and LG Display have demonstrated 55" panels at 8k resolution, which if it were to be inkjet printed, would require 160ppi with each subpixel being 4 times smaller than those at 80ppi. Furthermore, with the popularity of tablets around 10" in size, there is a strong motivation to inkjet print these at up to 4k2k resolution, which would require a pixel density of around 430ppi. This kind of resolution is likely to be the limit of inkjet printing for the foreseeable future as at the time of writing the inkjet hardware is not available to extend printing beyond 500ppi. Furthermore, printing at high resolution has significant implications for the inks used and the film uniformity becomes even more critical.

Details	λ_{max} (nm)	At 1000 cd/m ²			LT95 Normalized
		LE (cd/A)	EQE (%)	Voltage (V)	
Uniform sub-pixel	522	64.1	17.5	5.5	1
Non-uniform sub-pixel	522	66.9	18.3	5.4	0.4

Table 1. Summary of performance of inkjet printed OLED at 1000 cd/m²

Technology readiness for high-resolution inkjet printing

State of the art printheads are available with drops sizes down to 1 picolitre (1pl). Such a drop has a diameter of 12.6 μm in flight, essentially defining the minimum feature which can be printed. Whilst it is possible to force a printhead to eject drops smaller than its native resolution this can be at the expense of stability, which is not acceptable for mass-production. Even though low drop volume printheads exist, for example, Fujifilm Diamtix (FFD) Samba, Konica Minolta KM1024 or KM128SNG, and the recently released XAAR 5601, at the time of writing they are usually marketed for graphics or textiles use, and not easily integrated into current R&D printers. It is worth mentioning the FFD Dimatix 16 nozzle R&D cartridges. These come as 10 and 1pl and serve a valuable purpose on R&D printers. However, the reproducibility is barely acceptable for

OLED inkjet device manufacture and the frequency is severely limited, so demonstrating the inks functionality at comparable frequencies as mass production is a challenge.

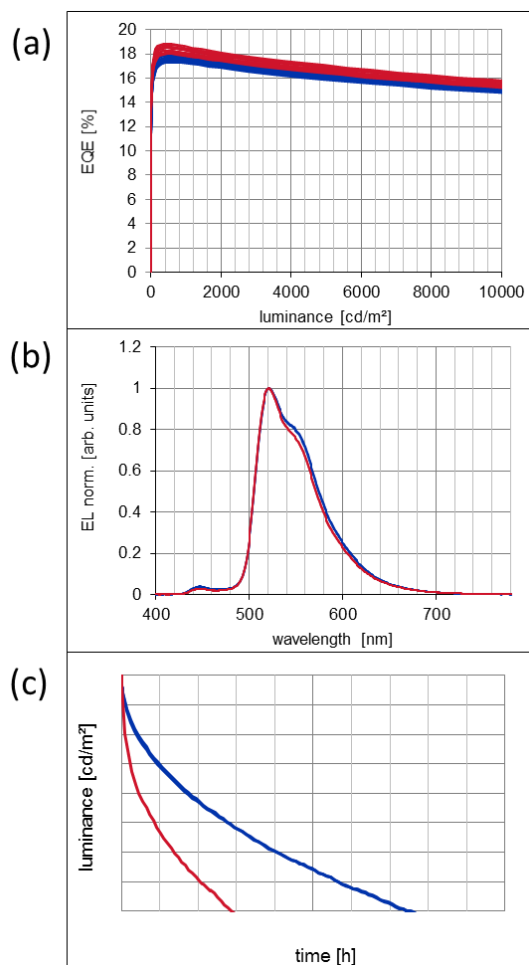


Figure 3. (a) EQE, (b) EL spectra, and (c) normalised lifetime, for devices with both uniform and non-uniform subpixels.

The frequency required to print is dependent on the resolution required, R (in this case defined by the distance between neighbouring drops), the quality factor Q , and the stage speed V according to the following equation:

$$F \text{ (Hz)} = R \text{ (dpi)} \times V \text{ (mm/s)} / 25.4 \times Q$$

The 25.4 constant is a conversion factor to account for the different conventions in units. It turns out that this frequency is not strongly affected by reducing the sub-pixel size. This is because, as the size reduces the volume of ink required reduces dramatically. For example a sub-pixel for an 80ppi display, may be $220 \times 64 \mu\text{m}$ in dimension and be able to hold a volume of 300pl before overspill. A sub-pixel for a 400ppi display might be more like $40 \times 10 \mu\text{m}$, but tests in our lab have shown that such a pixel would hold less than 4 pl before overspill. One sensible print strategy is to evenly space the drops required along the length of the sub-pixel, so for an 80ppi sub-pixel the drop spacing is 6 μm with a 10 pl printhead and for the 400ppi case, the drops spacing is 10 μm with a 1 pl printhead.

The next consideration is the tact-time of a printed substrate which is ideally as short as possible, however around 120 seconds is a likely to be required for mass production. A gen-

8 substrate is 2160×2460 mm in size. Assuming the printer used to print this has printheads along the entire width of the substrate so no x-direction scanning is required, then the print speed must be a minimum of 18mm/s to print the whole substrate in 120s. This would give a frequency requirement from the printhead of just 3 kHz, well within the operating specification of current industrial printheads, which will operate in ranges of 40 – 100 kHz depending on the model. This estimation does not take into account mura-correction techniques, but even including these the operating of the printhead is unlikely to be a problem in inkjet printing OLEDs.

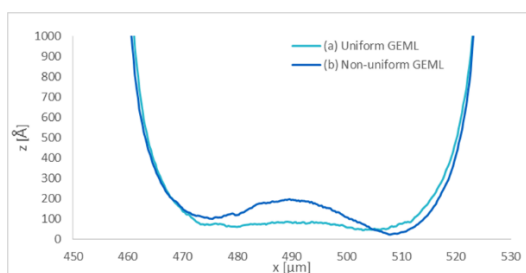


Figure 4. Stylus profilometer traces across the small dimension of the two devices shown in Figure 2.

As a final consideration in this section, the required stage accuracy should be estimated. For the 80ppi display pixels used in this manuscript the sub-pixel size is 220×64 μm . A repeat stage accuracy of 10 μm should be sufficient to print such a substrate. In reality the stage accuracies of R&D printers are already around 10 times better than this, with Ceradrop quoting an x and y stage accuracy of 1.5 μm and a reproducibility of 0.5 μm and Unijet, who manufacture printers up to mass production scale quoting 3 μm accuracy and 1 μm reproducibility. In the authors experience these figures are usually conservative.

Ink considerations for high-resolution inkjet printing

As the resolution required increases, the printhead drop size decreases and typically the viscosity requirement of the printhead also narrows. For example a 10 pl printhead may be perfectly able to jet inks over a wide range of 1 – 20 cp, however a 1 pl industrial printhead, may only reliably jet 1 – 5 cp. This has significant implications on the ink. This is compounded at high resolution where less ink per unit area can be contained by the sub-pixel. This is not immediately obvious, but as the active area scales for high resolution, so does the distance between the pixels. It is common for the as deposited ink to bulge outside the dimensions of the sub-pixel and this is significantly limited as the resolution increases. Therefore, to achieve the same layer thickness for higher resolutions the concentration of the ink used must increase, without the viscosity rising. This gives Merck small molecule inks, which have almost no viscosity–concentration dependence, a significant advantage when printing high-resolution OLEDs.

The topography of the layers becomes more sensitive at high resolutions. It was shown earlier that the topography has a very strong influence on the lifetime of the device. The mechanism causing the shape of the layers is the same regardless of the resolution and is related to the ink pinning at a certain point on the substrate bank material. If non-level ink was only within 5 μm of the bank material, this would be a good result for a 64 μm wide 80ppi device, but catastrophic for a 10 μm wide 400 ppi device, which would have only a very small percentage of the active area of the sub-pixel at the optimum thickness. Therefore, the ink and process need to be very carefully designed together to give flat film formation at any resolution.

Finally, high-resolution designs would require a shift to top-emission OLED architectures. These typically use second-node designs, which require much thicker cavity tuning layers, sometimes up to 250 nm, in contrast to bottom emission designs which are typically < 100 nm. This pushes the ink concentration to much higher values than is required for bottom emission, so Merck small molecule OLED inks are carefully designed in conjunction with a process, specifically for high resolution printing.

Conclusions

IJP OLED device data reported for a full stack of Merck KGaA materials and inks is very promising and demonstrates what can be achieved at resolutions suitable for 55 inch displays with 4k2k resolution. The technology is largely in place to be able to research and produce IJP OLEDs up to 500ppi, although the industry is waiting on an easily available 1pl printhead solution for research purposes. With the development of Merck small molecule inks for OLED, it seems as though the ink technology will not be a limiting factor for printing high resolution, however great care must be taken in designing an ink and process to give flat films when dried.

References

- [1] Takuya Sonoyama, Masahiro Uchida, Takumi Sago, Shotaro Watanabe, Kohei Ishida, Masaki Ito, Yuta Okawa, Minoru Yamada, Masaya Ichida, Seichi Tanabe and Hiroshi Kiguchi, “OLED Device Fabrication by Ink-Jet Printing Technology,” 21st International Display Workshops, Niigata, Japan, (2014).
- [2] Conor Madigan and Christopher Brown, “Inkjet Technology for OLED SSL Mass Production,” US DOE 12th annual Solid State Lighting R&D Workshop, San Francisco, USA, (2015).

Author Biography

Daniel Walker received his PhD in material science from the Technical University of Darmstadt (2012). Since then he has worked in the OLED research and development team at Merck based in Southampton, UK. The work focuses on developing processes and formulations for use in inkjet printed OLEDs for display applications.