OLLA Project Report

Manufacturing of the MS3 OLED lighting tiles

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Executive summary

Scope of this public report is parts of the fabrication, integration and scaling of the MS3 OLED stack.

It described the reference OLED stack development and the transfer on the fabrication machine. The strategy to achieve highest possible efficiency was based on the combination of two parts:

- Highly efficient hybrid OLED systems with stacked and interlayer approach
- Out-coupling improvement by using glass substrates with high refractive index and thick electron transport layers

Only in this combination highest efficiency could be achieved. The stack development has been done on low index substrates due to the fact that the handling of high-index substrates is difficult as well as to separate different parameters.

The OLED stack development was realized firstly on the experimental deposition systems installed at Novaled and Philips in parallel. The reference system has been then applied to a high-n glass substrate and further optimized for outcoupling.

The stacked OLED approach generated the highest efficacies on high-index substrates. It can be seen that the power efficiency with a flat out-coupling solution is 50.7 lm/W at a luminance of 1000 cd/m².

Due to time limitations only the hybrid approaches could be implemented on large area substrates. Based on a complete new structuring process specially developed for high-n substrates, the successfully scaling on high index substrates (15x15cm²) could be demonstrated.

1 Technical Summary

The reference systems based on the hybrid approach were optimised firstly on low index substrate. The optimized combination leads to color coordinates of (0.43/0.45) and a luminous efficacy of 26.1 lm/W at 1000 cd/m² without outcoupling improvement.

For the transfer on high-index substrates the ETL thickness was optimized to achieved highest efficiency. Novaled achieved 39.6 lm/W on small dots, Philips 37-48 lm/W efficiency. Using a half sphere lens Novaled achieved 67.5 lm/W and Philips 55-80 lm/W based on the same stack systems, but different machines.

Based on the result that the interlayer seems to be a critical issue concerning the OLED lifetime an alternative OLED stack approach was tested at Novaled. Instead of an undoped bipolar interlayer between the fluorescent and phosphorescent emitter layers a p- and n-doped layer was used to enable the carrier transport between the different emitter layers. Based on this optimized approach Novaled achieved 50.7 lm/W (including outcoupling) and 84.7 lm/W using a half sphere outcoupling system, in combination with a long lifetime.

The hybrid approach on high-index substrate was chosen to scale up. Starting with the recipe supplied by Novaled, device optimization was carried out on the larger deposition system in order to reach the target C.I.E. coordinates. An efficacy of 23.0lm/W and colour coordinates of 0.44/0.46 was reached after device optimization on low-index glass without outcoupling enhancement.

To implement this stack onto high-index substrates a new structuring technique had to be developed. High-index substrate is very sensitive against chemical etchants, so a complete lithography free printed process flow was developed. The process flow was implemented but shows a lot of improvement options due to the substrate material. For the large area implementation a new optimized grid layout was developed. From the simulation, a homogeneity of jmin/jmax=0.55, corresponding to a non-uniformity (defined as contrast) of 0.29 was be expected.

Lighting tiles with this grid design were produced by the screen printing process and the OLED stack was deposited. The fabricated tiles show the possibility to scale-up but also show a large inhomogeneity. The large inhomogeneity was analysed and is mainly based on a contact resistance between the metal layer and the ITO.

Electrical measurements at high total current furthermore showed that this contact resistance becomes unstable. This problem needs to be addressed by optimizing the screen printing process in combination with the high index glass substrate supplier.

As a result the scaling up of the high efficient OLED on high index substrates was successfully demonstrated but open process issues needs to solved to achieve compared efficacy and homogeneity on large substrate sizes.

2 Introduction

Scope of this report is the fabrication, integration and scaling of the MS3 OLED stack. It described the reference OLED stack development and the transfer on the fabrication machine. Also the results for the substrate preparation and scaling are summarized in this document.

3 OLED stack development

In this chapter the OLED stack development for the MS3 deposition was described. The strategy to achieve highest efficiency was based on the combination of two parts:

- Highly efficient hybrid OLED systems with stacked and interlayer approach
- Out-coupling improvement by using glass substrate with high refractive index and thick electron transport layers

Only in this combination highest efficiency could be achieved. The stack development has been done on low index substrates due to the fact that the handling of high-index substrates is difficult as well as to separate different parameters. The OLED stack development was realized firstly on the experimental deposition systems installed at Novaled and Philips in parallel, which is described in Section 3.1. The reference system has been then applied to a high-n glass and further optimized for outcoupling (Section 3.2). An additional stacked approach is described in Section 3.3. The transfer of the stack to the larger area deposition systems at Fraunhofer IPMS is described in Section 3.4.

3.1 OLED reference system (hybrid approach)

Not made public.

3.2 OLED system transfer and optimization on high-n substrates

The three-colour hybrid-white stack developed has been transferred to the substrates with a high refractive index. In order to optimize the outcoupling efficiency and colour coordinates, samples with different ETL thicknesses and cathode materials have been processed.

OLED reference system 1

Not made public.

OLED reference system 2

Parallel to the investigations at Novaled a second reference system was developed by Philips. For the red layer Philips used a phosphorescent red emitter doped into a suitable host, for the green layer Philips used a phosphorescent green emitter from Merck doped into a mixed matrix, and for the blue layer Philips used a commercial fluorescent emitter doped into a suited matrix.

The RGB layer stack is shown in the figure below. The stack was optimized optically using Lightex simulation program.

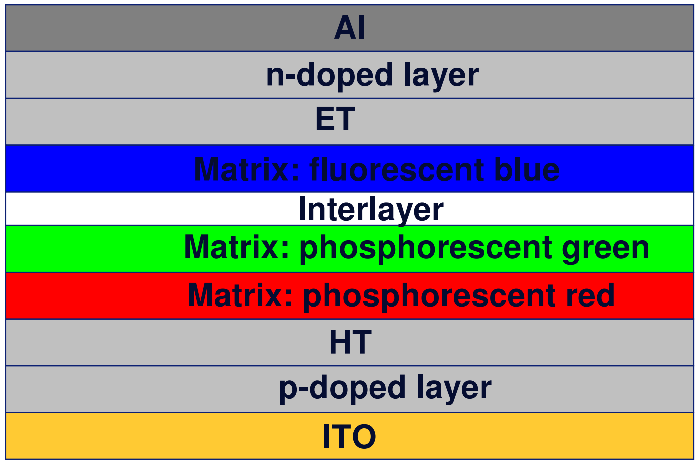

Figure 1: *Hybrid white OLED stack with interlayer (Philips).*

In the following the stack was optimized experimentally for maximum efficacy on normal glass mainly by varying the thickness of the electron transport layer (ET). The interlayer thickness was varied, and optimized.

Subsequently the stack was transferred to high index glass. For optical out-coupling on high index glass Philips used pyramid structures that were coupled to the OLED my means of a high index matching fluid. The figure below shows an OLED on high index glass with a pyramid out coupling structure.

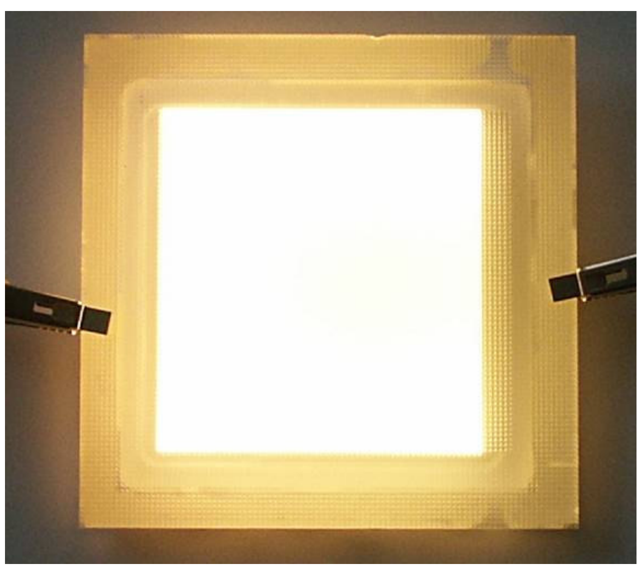

Figure 2: *Hybrid white OLED on high index glass with pyramid out-coupling structure*

The following table shows the results of different runs on high index glass measured in an integrating sphere. The efficacy without out-coupling was between 22 and 28 lm/W at 1000 Cd/m2 depending on colour. With a micro-lens out-coupling structure Philips finds 37 – 45 lm/W at a brightness of 1000 Cd/m2 without emission at the edges of the devices. If emission from the edge of the glass substrate is included, efficacy values between 39-48 lm/W are found. Using a large hemisphere to extract practically all light from the substrate Philips found efficacy values between 55 and 80 lm/W. The best device has an efficacy of approximately 48 lm/W at 1000 Cd/m^2 including emission from the edges of the substrate. This value has been reproduced several times on different devices.

Table 1: *Efficacy values in lm/W and CIE colour x, y coordinates of hybrid white OLED devices with interlayer on high index glass. The symbols (coloured triangles) can also be found in the colour diagram below.*

Substrate	OLED 1000 Nit	With out-coupling 1000 Nit	With out-coupling + edge emission	Half sphere	x, y
10 cm^2 ▲	22	37	39	63	0,432, 0,434
10 cm^2 ▲	26	42	46	69	0.462, 0.455
10 cm^2 ▲	22	37	40	55	0.461, 0.430
0.2 cm^2 ▲	28	45	48	80	0.425 0.453
10 cm^2 ▲	24	40	42	68	0.499, 0.423

The following figure shows the colour coordinates of the hybrid white OLED stacks on high index glass. Also shown are the colour coordinates of the three red, green and blue emitters.

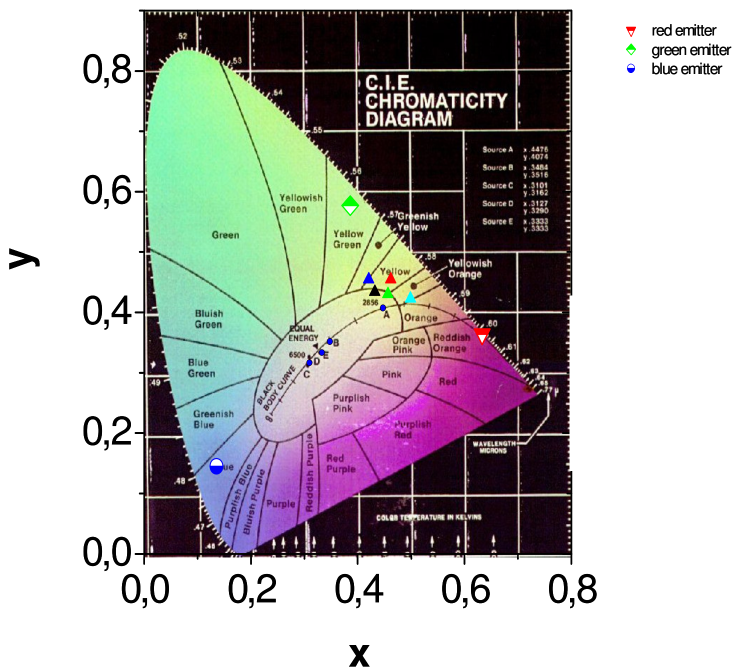

Figure 3: *CIE Colour coordinates of the OLED hybrid white devices on high index glass (from table above).*

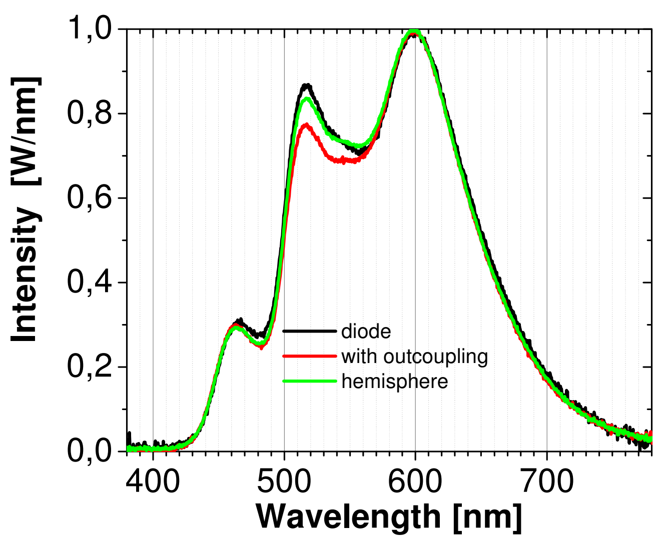

Figure 4: *Normalized spectrum of hybrid white OLED on high index glass.*

The Figure 4 above shows that out-coupling is a bit less efficient for the green spectral range.

Philips achieved for hybrid white stacks with interlayer 45 lm/W with a micro-lens out-coupling structure on high index glass excluding emission at the edges of the substrate. If edge emission is included 48 lm/W can be achieved. Using a large hemisphere for light extraction 80 lm/W efficacy can be achieved. Colour temperature was 3600 K, CRI was 83. These results were obtained both on small and large diodes.

3.3 OLED reference system (stacked approach)

Based on the result that the interlayer seems to be a critical issue concerning the OLED lifetime an alternative OLED stack approach was tested by Novaled. Instead of an undoped bipolar interlayer between the fluorescent and phosphorescent emitter layers a p- and n-doped layer was used to enable the carrier transport between the different emitter layers.

The general OLED structure is sketched in the figure below.

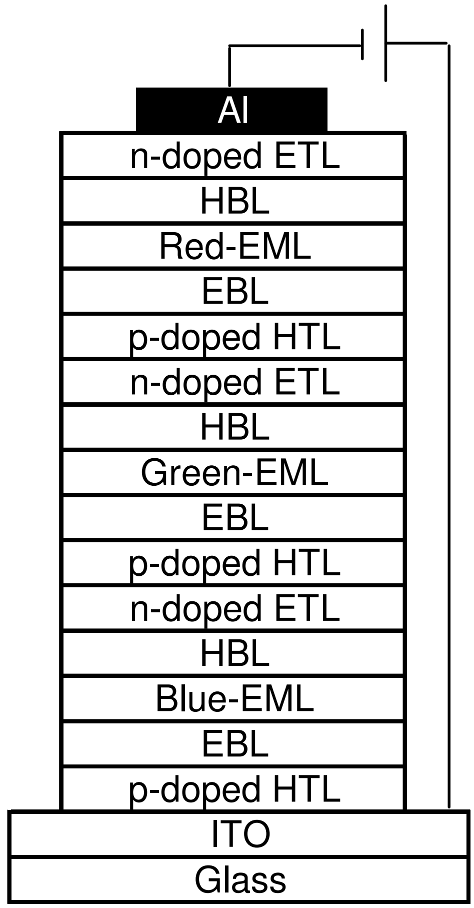

Figure 5: *Stack for stacked-white OLED.*

Since each PIN OLED starts from a highly p-type electrical doped HTL and ends with a highly n-type doped ETL, we can just simply stack one PIN OLED on top of another PIN OLED without any intermediate layer. Figure 5 shows an example of stacked OLED with red, green and blue emission units to generate white light. The n-type doped hole transport layer in previous emission unit is in contact with the p-type doped electron transport layer in the next emission unit to form a doped so called organic p-n junction.

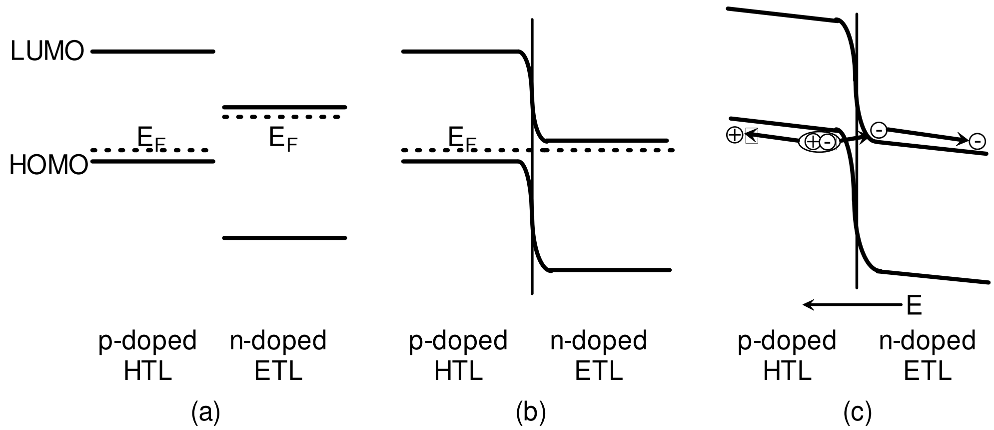

Figure 6: *Sketched energy level diagram of p-n junction: (a) without contact, (b) no external bias, and (c) under reverse bias.*

The understanding how such an organic p-n junction works is very important for the device design and further improvement. Figure 6 (a) sketches the energy levels of doped transport layers qualitatively. With effective doping, the Fermi level within the p-doped HTL is close to the highest occupied molecular orbital (HOMO), while the Fermi level within the n-doped ETL is close to the lowest unoccupied molecular orbital (LUMO). If they contact each other, a common Fermi level throughout both layers is required by equilibrium (see Figure 6 (b)).

When an external bias is applied on a stacked OLED, the p-n junction works actually under a reverse bias as shown in Figure 6 (c). The reverse bias lowers the LUMO level of ETL with respect to the HOMO level of HTL. Therefore the electrons may tunnel through from occupied HOMO states of HTL to unoccupied LUMO states of ETL. Then these electrons will immediately be driven away from the interface by the external electric field and be injected into emission layer. The holes left in the HOMO states of the HTL are in the same way injected into the emission layer of its own emission unit.

It was found that properly selecting host and dopant materials of transporting layers plays an important role to achieve efficient charge separation at the interface of p-n junction.

In order to enhance the light which is coupled into the substrate the whole OLED stack is build on an ITO coated high-index glass substrate.

The electroluminescent (EL) spectra (measured in an integrating sphere) of the stacked white OLED at 1000 cd/m^2 are illustrated in Figure 7. The emission covers a wide range of the visible region. After attaching a flat outcoupling foil on top of the white OLED, the spectrum slightly changes. More green and red emission is coupled out. This effect if even more pronounced if the spectrum is measured with a high refractive index matched half sphere lens in order to get all the light out of the substrate as described in the chapter above.

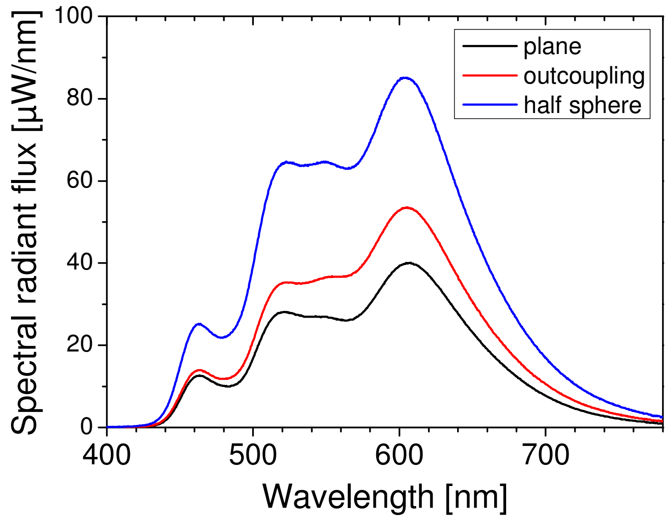

Figure 7: *Comparison of spectra without out-coupling, flat outcoupling and half sphere. The spectra are measured in the integrating sphere at a current corresponding to a luminance of 1000 cd/m² without outcoupling.*

Table 2 shows the luminous efficiency without, with plane out-coupling structure, and with a half sphere lens. The luminous efficiency values are measured in an integrating sphere according the OLLA white paper. It can be seen that the power efficiency with a flat out-coupling solution is roughly 50 lm/W at a luminance of 1000 cd/m². However, the measurement with the half sphere lens demonstrates that just 60% of the light which is trapped in the high-index glass substrate is coupled out by the flat out-coupling and demonstrates the further potential of this OLED stack.

Table 2: *Comparison of luminous efficacies and color coordinates for OLED without outcoupling, with flat out-coupling and with half sphere. The measurements with and without out-coupling were performed at the same light flux, corresponding to a luminance of 1000 cd/m² (measured without out-coupling). The edges of the substrate were covered by tape.*

	Luminous efficacy [lm/W]	Color coordinates
Plane	35.8	(0.39, 0.42)
Out-coupling	50.7	(0.44, 0.45)
Half sphere	84.7	(0.43, 0.45)

3.4 OLED system transfer to larger area deposition system

To scale the OLED deposition, an OLED reference system by Novaled (hybrid approach, chapter 3.2) was installed at Fraunhofer IPMS on a Sunic Sunicell 200 plus system. Starting with the recipe supplied by Novaled, device optimization was carried out in order to reach the target C.I.E. coordinates. An efficacy of 23.0lm/W and colour coordinates of 0.44/0.46 was reached after device optimization on standard glass (not high n). The corresponding L-I-V data are shown below. Device parameters are estimated from forward emission based on the approximation of Lambertian emission and are obtained without out-coupling enhancement.

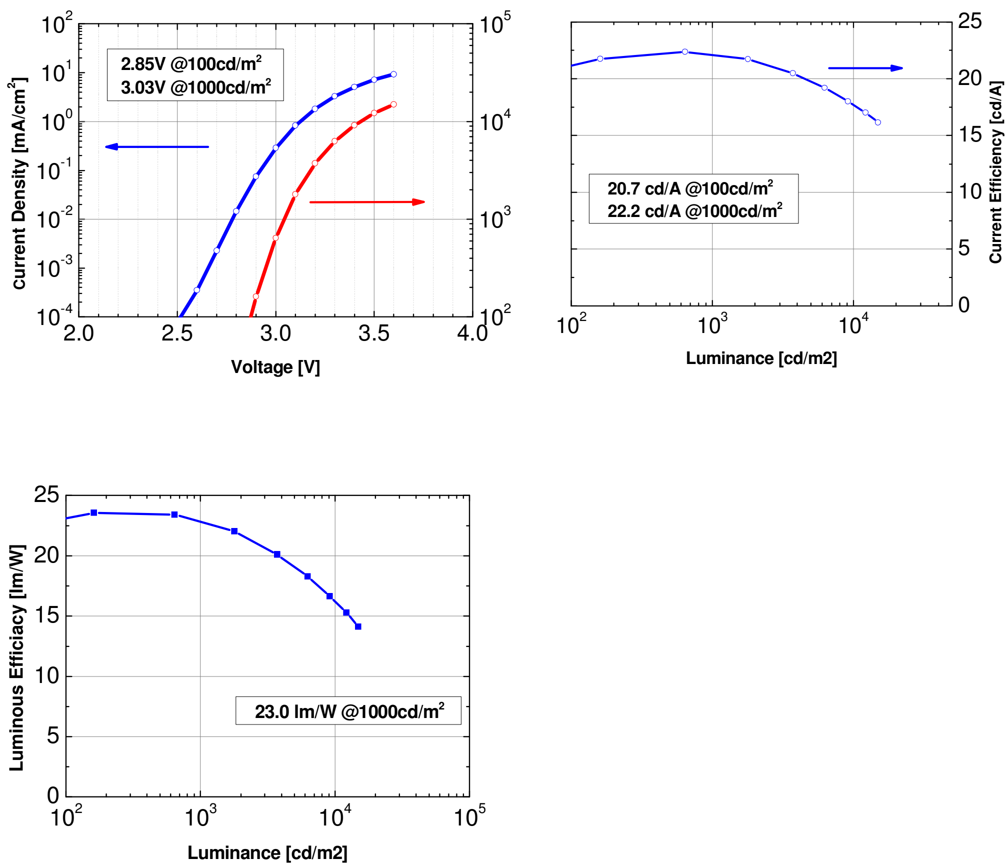

Figure 8: *Current density, luminance vs. voltage curve, current efficiency and luminous efficacy vs. luminance curves for a hybrid white OLED. Active area of the device is 19.9mm^2 (all data measured in forward emission and without out-coupling enhancement).*

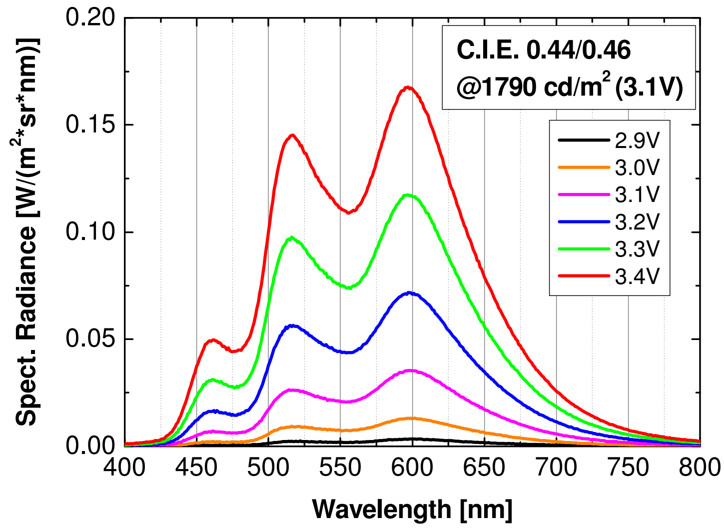

Figure 9: *Emission spectra at different driving voltages.*

The Novaled reference stack is based on a rather thin hole transport layer (HTL). The HTL thickness is known to be an important parameter for the yield of large area OLEDs. In order to realize the 300mmx300mm OLED demonstrator the HTL thickness was increased.

The hybrid approach white stack with thicker HTL is the base for large area implementation for the MS3 demonstrator. The device has to be implemented onto high-index substrates to improve the light out-coupling. The implementation requires a lithography free structuring process based on the fact that the high index substrate cannot be used for standard lithography vendors. In the following chapter the process for integration will be described.

4 Substrate fabrication

To achieve highest luminance efficiencies a high index substrate material for scaling has to be used. The base for the fabrication was Schott high-index substrate.

Due to the sensitivity of the high index substrate against etchant materials, the substrate has to be prepared using alternative method. For this reason, screen printing technique was used for the preparation of passivation layer and metal grid with OLLA MS2 layout on high index substrate. The screen printing process was done in a clean room. Prior to the screen printing, the ITO-coated high index substrates were cleaned with deionised water and organic solvents in an ultrasonic bath. A conductive silver for the metal gird and a dielectric material for the passivation material were used, respectively. First, metal grid was deposited on the structured ITO substrate following a passivation layer coating over the metal grid. The screen printing materials require a thermal curing after each deposition process. After the thermal curing, these materials become stable so that the substrates were cleaned with standard cleaning process. The substrates were annealed prior to being installed in the OLED evaporation chamber.

Firstly the process was tested on low-index substrate.

The initial sample shows that large area OLED substrates can be provided by the screen printing process. The remaining problems are mainly caused by handling procedures in the printing process and should be avoidable after further process optimization.

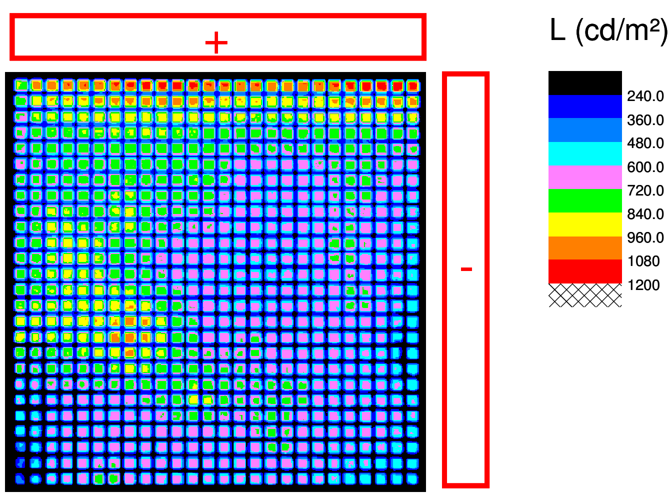

Figure 10: *Luminance map (video photometer) of screen printed 150x150 mm² panel on standard glass.*

5 OLED scaling

For the layout of the MS3 demonstrator, a combination of 4 lighting tiles with outer dimensions of 150x150 mm² each was chosen. In order to minimize the non-active area between the tiles, contact pads are positioned only at the corners and at the two outside edges of the tiles (see Figure 11). For an efficient current distribution to the active area, a new grid concept has been developed. Instead of the regular square grid from the MS2 layout, a radial grid with a high degree of design freedom was employed. Within some practical restraints, the optimum grid geometry was found by a series of FEM simulations in which the homogeneity was optimized for a given grid fill factor. The optimization was based on the electrical OLED-parameters of the MS2 stack.

The final grid geometry and the simulated distribution of the OLED current density is shown in Figure 11. The OLED data in the simulation of Figure 11 are taken from an IPMS MS3 reference stack on small area an low index substrate with an inverse slope of the j-V-characteristics of dV/dj = 29 Ωcm² at 500 cd/m². From the simulation, a homogeneity of jmin/jmax=0.55, corresponding to a non-uniformity (defined as contrast) of 0.29 would be expected. The simulated homogeneity is smaller than in the original optimization since the new MS3 OLED stack has a significantly steeper j-V characteristics compared to the MS2 stack on which the grid optimization was based.

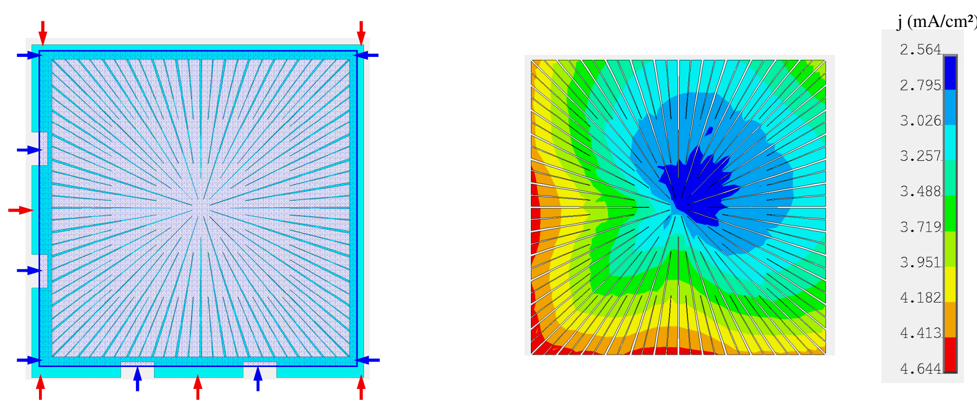

Figure 11: *Left: Grid layout and contacting scheme for the MS3 demonstrator tile (outer dimensions 150x150 mm²). Red arrows: anode contacts, blue arrows: cathode contacts (cathode contact pads not shown), cyan area: busbar region, blue shaded area: cathode. Right: Simulated distribution of the OLED current density for a total current of 550 mA. The maximum current of 4.6 mA/cm² would correspond to a luminance of 1040 cd/m² from the reference OLED.*

Lighting tiles with this MS3 grid design were produced by the screen printing process and the OLED stack was deposited.

Conclusion

The OLLA MS3 demonstrator has been made fabricated using our best known stack, best possible outcoupling and high-n glass substrates. Due to processing and time issues, it has not been possible to successfully reproduce the final OLLA results obtained on a large substrate. Nevertheless on smaller substrates a white OLED with an efficacy result of 50.7 lumen per Watt with an expected lifetime of over 10.000 hours have been delivered, as originally was promised in the OLLA project proposal back in 2003.

Figure 12: *The final MS3 demonstrator with 4 sub panels, as photographed during the final event.*