



OLLA Project Report

Demonstrate a white p-i-n type OLED

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Executive summary

In order to realise white Organic Light Emitting Diodes (OLEDs) for lighting applications, several different approaches are possible. The main issue is to find an OLED solution based on small molecules which generates highly efficient, long living white light. A possible way which could lead to a stable white OLED solution based on small molecules is to use white p-i-n type OLEDs with doped transport layers and different kind of emitter stacks to generate the white light.

The aim of this report is to demonstrate a proof of principle for this p-i-n OLED approach. This document will demonstrate first white p-i-n type OLEDs produced in the framework of OLLA. Herewith, the feasibility of the p-i-n type OLED approaches should be demonstrated and several routes to reach the next milestones by further optimization of these systems are described in this document.

Furthermore, the most important OLED parameters in terms of general lighting are given in this document to compare the results of these first demonstrators and decide about the future development strategies of small molecules for OLED lighting.

The main parameters which will be discussed later are:

- Power and Current Efficiency
- Lifetime
- Colour point
- CRI
- Operating voltage

This order of parameters above has been fixed for the OLLA project.

1 Technical Summary

In the following sections the technical data of the selected small molecule white OLED approaches are summarized which act as demonstrators in terms of the deliverable.

I) **Blue (Flrpic) and orange (ADS RE76) triplet emitter approach:**

At brightness of 1000 cd/cm²:

- Power efficiency: 6.1 lm/W
- Current efficiency: 6.9 cd/A
- CRI value : 58
- CIE(1931) CCs: (0.32, 0.37), T_c = 5809 K
- Lifetime t_{50%} ~ 2 h (at RT)
- Operating voltage: 3.6 V
- Size: 6 mm²

II) **Blue and yellow state of the art singlet emitter approach:**

At brightness of 1000 cd/cm²:

- Power efficiency: 7.8 lm/W
- Current efficiency: 9.6 cd/A
- CRI value : 65
- CIE(1931) CCs: (0.33, 0.34), T_c = 5858 K
- Lifetime t_{50%} = 1500 h (at RT)
- Operating voltage: 3.9 V
- Size: 6 mm² and 4cm²

III) **Combined triplet and singlet emitter approach:**

a) 2 color device

At brightness of 1000 cd/cm²:

- Power efficiency: 7.8 lm/W
- Current efficiency: 9 cd/A
- CRI value : 54
- CIE(1931) CCs: (0.30 , 0.34)
- Operating voltage: 3.7 V
- Size: 4 mm²

b) 3 color device

At brightness of 1000 cd/cm²:

- Power efficiency: 10 lm/W
- Current efficiency: 13 cd/A
- CRI value : not determined
- CIE(1931) CCs: (0.32 , 0.40)
- Operating voltage: 4.0 V
- Size: 4 mm²

2 Introduction

This document describes the properties and achievements in terms of the OLLA project to present a first physical demonstrator of a white p-i-n type OLED produced in the framework of OLLA.

Different approaches of a white OLED in p-i-n stack are presented and discussed and pictures of the demonstrator are provided. Furthermore, next steps to achieve the first project milestone (the 10 lm/W white OLED without improved light outcoupling enhancement and a lifetime of 2,000h at 1,000 cd/m²) are discussed.

3 Demonstration of white p-i-n type OLEDs

During the first twelve months of the project, Novaled and the IAPP have tested different small molecule (SM) white OLED approaches in a so-called p-i-n type structure. For this purpose, different layer stacks of fluorescent and phosphorescent emitter systems were implemented and characterized in p-i-n type OLED stacks. These structures rely on an electrically intrinsic emission layers, embedded between an n-doped electron transport and a p-doped hole transport layer. By this (and a proper adaptation of interlayers) low operating voltages and high quantum efficiency along with long lifetime are possible.

The Novaled/IAPP standard p-i-n reference system for all presented white OLED approaches is build up by the following stack configuration:

ITO / NHT-5:NDP-2 / EBL / different host :emitter stack / BPhen/ BPhen:Cs / Al

In the following sections selected SM white OLED approaches will be discussed which act all as demonstrators in terms of this report.

3.1 Demonstration of a two color phosphorescent white p-i-n type OLED

First tests concerning a two color white light phosphorescent p-i-n OLED based on Flrpic as light blue and RE076 from American Dye Source, Inc. (ADS) as orange red triplet emitter have been done at NOVALED.

Due to the fact that the CIE 1931 color coordinates of the blue emitter Flrpic is rather light blue, the best achievable color coordinates close to the color point E are (0.32, 0.37) as demonstrated in the following figure. With this diode, a color rendering index (CRI) of 58 could be achieved. Therefore, it becomes quite clear that in terms of color coordinates and CRI values a deeper blue emitter is required to improve this two color phosphorescent white approach.

However, the two color phosphorescent white Flrpic and RE076 based p-i-n OLED shows an operating voltage of 3.6 V at a brightness of 1,000 cd/m². At the same brightness this is related to a current and power efficiency of 6.9 cd/A, and 6.1 lm/W, respectively. The corresponding LIV and efficiency curves of the two color phosphorescent white sample are plotted in the next figure. As one can see, the current efficiency drops to 50% between 100 and 1,000 cd/m², which is a non-usual behavior for such diodes. Usually, the efficiency stays almost constant at least between 100 and 1,000 cd/m². Further investigations have to clarify this.

Furthermore, lifetime measurements of this sample demonstrate that in spite of the low operating voltage and decent efficiencies the two color phosphorescent white approach as it is demonstrated by this OLED is not the best choice. The investigated sample with Flrpic as blue emitter shows a lifetime of $t_{50\%} = 2$ h at $1,000 \text{ cd/m}^2$. Therefore, another blue emitter or related matrix materials which promises a more stable OLED are required to improve the lifetime significantly.

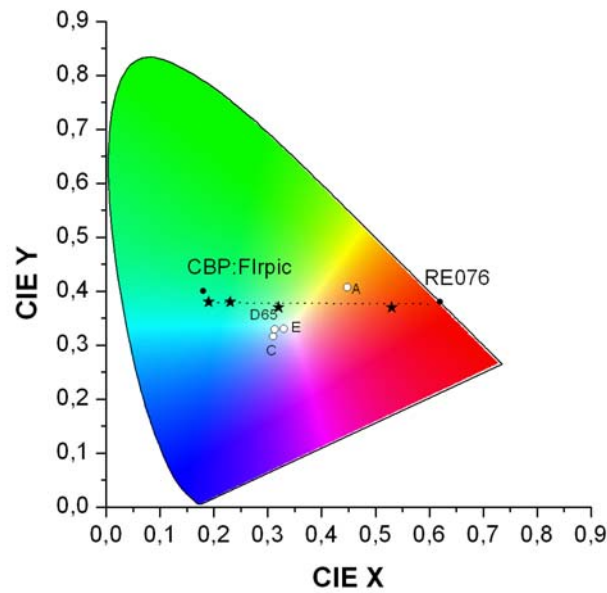


Figure 3.1: Measured CIE color coordinates of two color phosphorescent white p-i-n OLEDs based on Flrpic and RE076 at $1,000 \text{ cd/m}^2$.

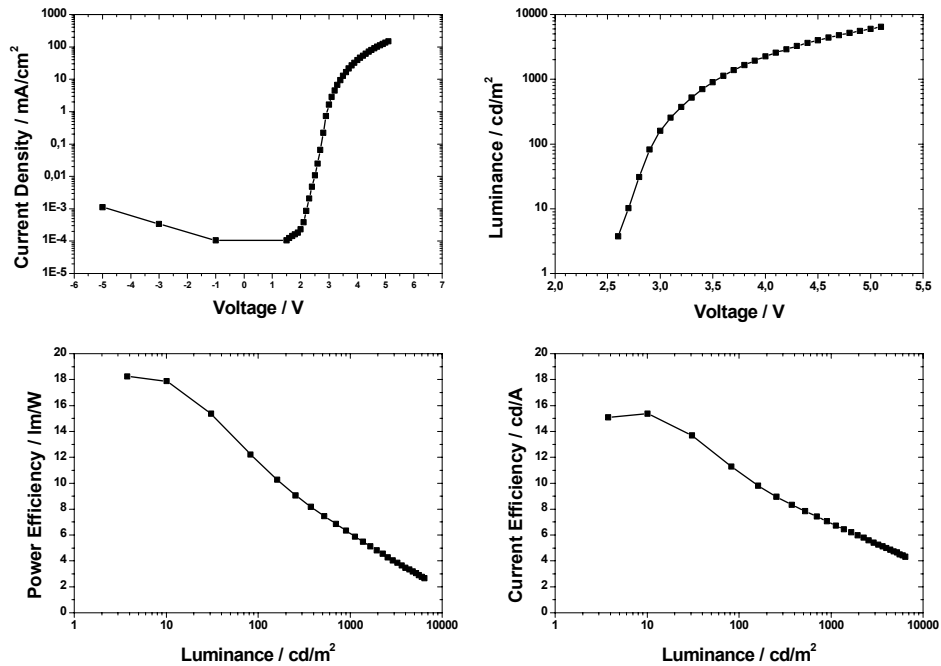


Figure 3.2: LIV and efficiencies curves of the two color phosphorescent white p-i-n OLED.

3.2 Demonstration of a two color fluorescent white p-i-n type OLED

Based on first test structures with two phosphorescent emitter systems described above, further work was done on the development of a two color white fluorescent emitter stack embedded in a p-i-n type OLED architecture at Novaled. For this purpose, matrix and singlet emitter material from a non OLLA Novaled project have been tested as a benchmark for other host:guest systems developed in the framework of the OLLA project.

The overall layer sequence of the p-i-n structure is basically the same as mentioned before:

ITO / NHT-5:NDP-2 / BH-1 / yellow emitter system/ blue emitter system / BPhen/
BPhen:Cs / Al

In contrast to the first experiments with two triplet emitter systems, this time a state of the art commercial yellow and blue singlet emitter system have been used by Novaled. The purpose of this reference system was to

find out which color rendering index (CRI) value can be achieved with a two color white system which contains a yellow and (compared to Flrpic) significantly deeper blue emitter. Furthermore, Novaled knows from pre OLLA experiments that the used matrix materials do not lead to a wrong carrier balance inside of the emission zone and that they are much more stable than the tested triplet emitter system.

As a summary, the following electro-optical characterization and lifetime results of the two color singlet white OLED have been achieved:

As shown in the following figure, the two color singlet white system leads to low operating voltage and good efficiencies, considering the fact that this is a full singlet emitter system. An operating voltage of 2.9 V and 3.9 V at a brightness of 100 cd/m² and 1,000 cd/m² can be reached. At a brightness of 1,000 cd/m² this is related to a current and power efficiency of 9.6 cd/A, and 7.8 lm/W, respectively. The current efficiency shows a small roll-off at higher luminance.

The CIE color coordinates of this device are (0.33, 0.34) at 1,000 cd/m² which is pretty close to the white color point E, and the CRI value is 65.

Lifetime measurements at room temperature show that a lifetime of more than 1,500 h at 1,000 cd/m² can be reached by this OLED. Further optimization is possible here since a similar white system (but with different color coordinates) reached already a lifetime of 10,000 h at brightness of 1,000 cd/m².

A similar OLED device as described above was fabricated for demonstration purpose by Novaled on a substrate with an active area of 2x2 cm². In Figure 3.6, a photo of this OLED demonstrator is shown which runs at a brightness of approximately 1,000 cd/m².

Furthermore, it could be demonstrated that the CIE color coordinates could be shifted by simply tuning the micro cavity of the OLED. As shown in Figure 3.4, the color coordinates can be shifted from (0.33, 0.34) to (0.48, 0.46) at 1,000 cd/m² by a variation of the thickness of the electron and hole transport layer. By doing this, even an OLED with color coordinates of (0.45, 0.40) close to the white point A (at 1,000 cd/m²) could be realized with a CRI value of 60.

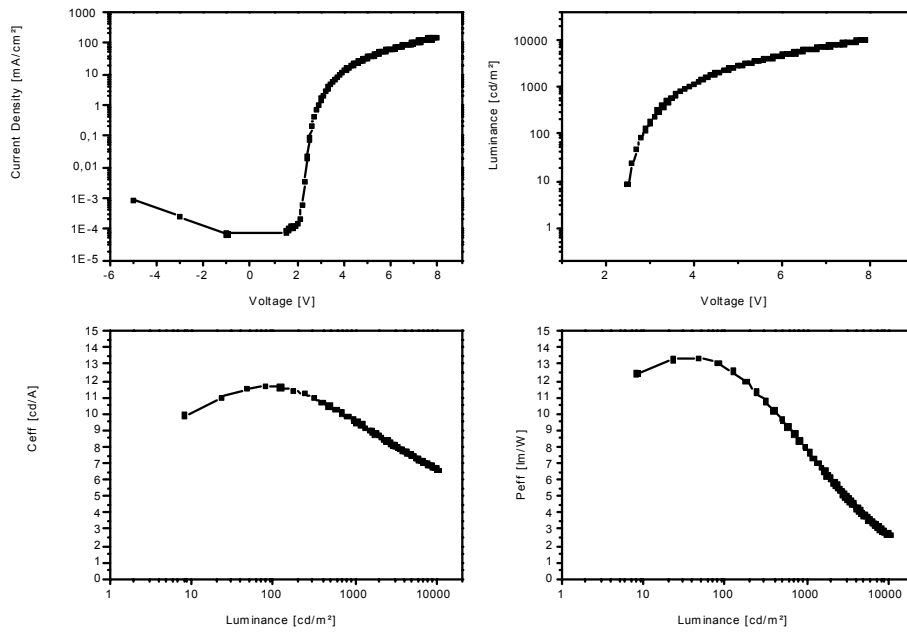


Figure 3.3 LIV and efficiencies curves of the two color fluorescent white *p-i-n* OLED.

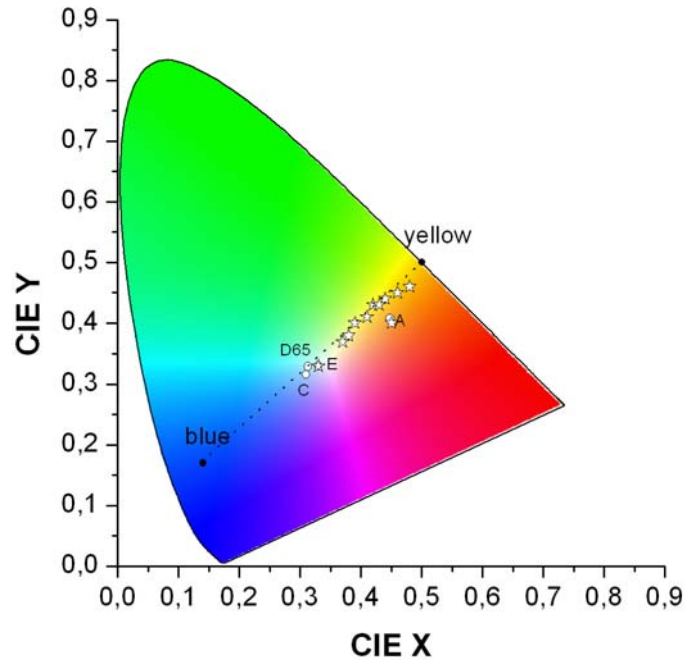


Figure 3.4: Measured CIE color coordinates for cavity tuned two color fluorescent white *p-i-n* OLEDs at 1,000 cd/m^2

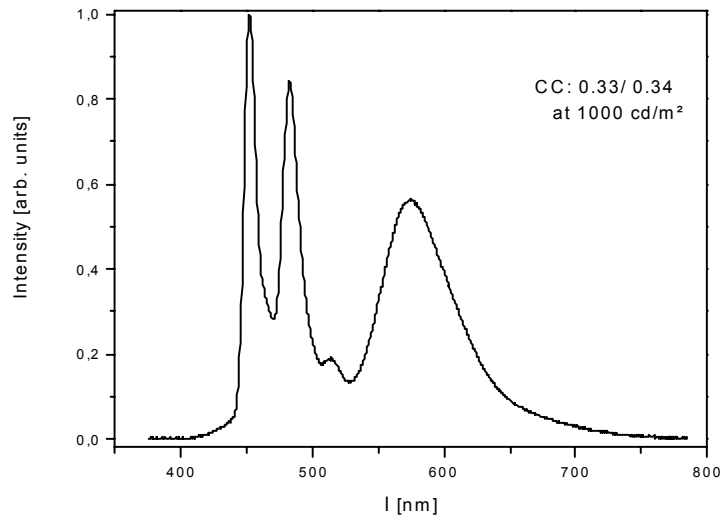


Figure 3.5: Spectrum at 1,000 cd/m² of the two color fluorescent white p-i-n OLED.



Figure 3.6: Picture of the two color fluorescent white p-i-n OLED produced at Novaled.

3.3 Demonstration of a combined phosphorescent and fluorescent white p-i-n type OLED

Until now, blue phosphorescent emitters are still quite problematic, both with respect to device lifetime and emission color, which is usually greenish blue instead of deep blue. Also the results shown in Chapter 3.1 demonstrate these two issues quite well. As a workaround, IAPP has developed an approach which combines deep blue singlet emitters with highly efficient triplet emitters for the green and red. An obstacle with these devices is the usually low triplet energy gap of the blue singlet emitters, which leads to a quenching of triplet excitons created in the red or green emission layers once these excitons reach the interface of the blue singlet emitter. The IAPP solves this problem by introducing a further layer in between the singlet and triplet emission layers, which prevents from triplet exciton diffusion by having a high triplet energy gap. We will call this layer exciton blocking layer (XBL).

The approach as described above is both applicable for two and three color emission devices. In the following, we will discuss an example for each, using the same outer layer structure as Novald for both devices:

ITO / NHT-5:NDP-2 / EBL / white emission layers / BPhen/ BPhen:Cs / Al

Two color device with exciton blocking layer

This device uses a orange-red triplet emitter in combination with a light blue singlet emitter (LBSE) from a non OLLA source. The XBL consists of a very thin nearly ambipolar layer, which blocks the triplet excitons created in the red emitter. The emission system is therefore as follows:

hole side / matrix: red triplet emitter / XBL / matrix: LBSE / electron side

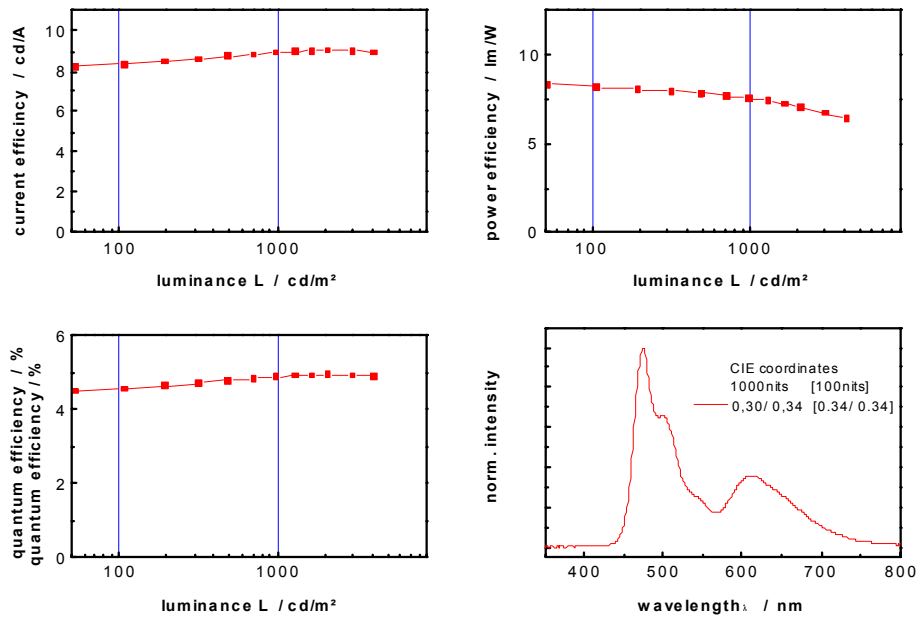


Figure 3.7: Performance of the two color white p-i-n OLED with combined singlet and triplet emitter

Performance data of this device are shown in the figure above. The device needs an operating voltage of 3.2 V and 3.7 V for 100 and 1000 cd/m², respectively. The efficiencies are quite stable for the different luminances. For the lighting-relevant 1000 cd/m², the device reaches a power efficiency of 7.8 lm/W. The CIE color coordinates lie close to the point of equal energy, and reach CIE values of (0,34; 0,34) for 100 cd/m². The CRI of this device is 54, which is close to the one measured for the two color device presented in Chapter 3.1. At higher luminance, the color shifts slightly to (0,30; 0,34), which is still well inside the white range recognized by the human eye.

Adjusting the exact color point depends in a sensitive manner on the nature and thickness of the exciton blocking interlayer as pointed out in the following figure. The XBL material used in this device, which is preferably hole transporting, leads to a lack of electrons at the hole injection side; if the layer becomes too thick, therefore the red emission becomes inefficient. A more ambipolar interlayer would improve this.

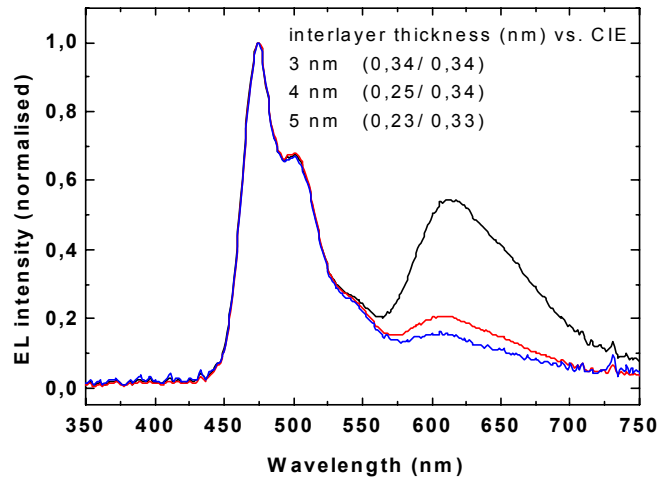


Figure 3.8: Color shift upon change of interlayer thickness

Three color device with exciton blocking layer

For improving the CRI of the devices, IAPP developed a three color stack as well. This device uses an orange-red triplet emitter and the green Ir(ppy)₃ as triplet emitter together with a deep blue singlet emitter (DBSE). Further, the device benefits from the higher ratio of light which is emitted from triplet materials and leads to an increase in efficiencies compared to the corresponding two color device.

Also the XBL has been improved with respect to its ambipolarity and consists now of a co-evaporated hole and electron conductor, both with high triplet gap. The resulting emission system is as follows:

hole side / red triplet emitter / green: Ir(ppy)₃ / XBL / blue: DBSE / electron side

Also this device needs a low driving voltage only, 3.2 V and 4.0 V for 100 and 1000 cd/m², respectively. As expected, the efficiencies are also somewhat increased as compared to the two color device, and reach now up to 10 lm/W at 1000 cd/m² (peak power efficiency 12 lm/W at 100 cd/m²). Figure 3.9 shows the performance data in detail. In the case of this device, the color is somewhat shifted to warm white.

As for the two color device, the exact color point depends on the interlayer. However, in the current OLED, the interlayer consists of a co-evaporated hole and electron transporting matrix material, such that a more balanced charge transport is reached.

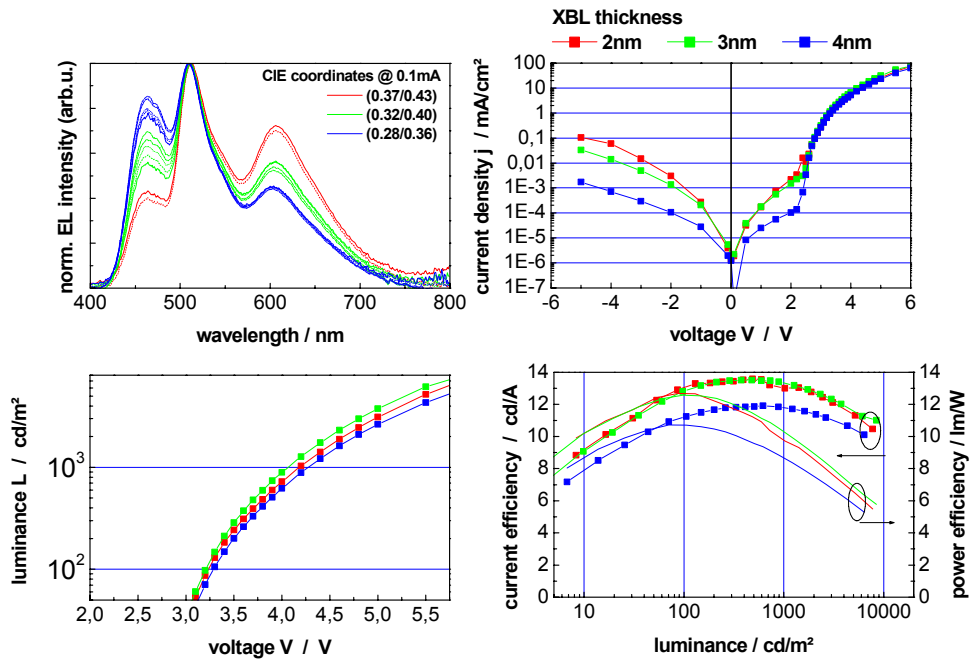


Figure 3.9: Performance of a three color white OLED with combined singlet and triplet emitters. Three devices with different interlayer thickness are shown.

4 Conclusion of white p-i-n type OLED demonstrator

This document describes the properties and different approaches to realise a white p-i-n type OLED. Different approaches of a white OLED in p-i-n stack were presented and discussed. Furthermore, several routes to achieve the 10 lm/W white OLED without improved out coupling enhancement and a lifetime of 2000h at 1000 cd/m² by a p-i-n OLED stack with doped transport layers are introduced.

Apart from optically and electrically stack optimization, further improvement is planned by implementing the new and unique Novaled full organic molecular doping technology also on the n-side of the p-i-n type OLED stack. The replacement of the BPhen:Cs ETL by new and more stable Novaled technology will lead to an increase in lifetime and improved OLED performance.

The aim of this report, to demonstrate a proof of principle for the white p-i-n type OLED approach is fully met.

Furthermore, the most important OLED parameters in terms of general lighting are given as a base for a discussion about further strategies in the development of small molecules p-i-n type OLEDs for lighting applications.