Project:

HyperOLED

(Grant Agreement number 732013)

"Development of high-performance, hyperfluorescence OLEDs for use in display applications and solid state lighting"

<u>Funding Scheme</u>: Research and Innovation Action <u>Call</u>: ICT-02-2016 "Thin, Organic and Large Area Electronics" Date of the latest version of ANNEX I: 12/10/2016

D5.7 Report on deep blue OLEDs

Project Coordinator (PC):	Dr. Christof PFLUMM Merck Kommandit Gesellschaft auf Aktien
Project website address:	www.hyperoled.eu
Lead Partner for Deliverable:	Merck
Report Issue Date:	23/12/2019

Document History (Revisions – Amendments)		
Version and date	Changes	
1.0 – 23/12/2019	First version	

Dissemination Level			
PU	Public	Х	
PP	Restricted to other program participants (including the EC Services)		
RE	Restricted to a group specified by the consortium (including the EC Services)		
СО	Confidential, only for members of the consortium (including the EC)		



The H2020 HyperOLED project is a three-year EC funded project entitled "Development of high-performance, hyperfluorescence OLEDs for use in display applications and solid state lighting". The project will run from January 2017 to December 2019.

The overall goal of the HyperOLED project is to develop materials and matching device architectures for high performance, hyperfluorescence organic light emitting diodes (OLEDs) for use in display applications and solid state lighting. The innovative OLEDs will be realised by combining thermally activated delayed fluorescence (TADF) molecular hosts with novel shielded fluorescence emitters, targeting saturated blue emission of very high efficiency, especially at high-brightness levels.

Further efficiency gains will be achieved through molecular alignment to enhance light outcoupling from the hyperfluorescence OLEDs. Using shielded emitters will enable simpler device structures to be used, keeping drive voltages low to be compatible with low voltage CMOS backplane electronics. This will enable demonstration of the concept's feasibility for high-brightness, full-color OLED microdisplays as one application example.

To develop the hyperfluorescence OLEDs, the following scientific and technical objectives will be targeted:

- Objective 1: Develop shielded emitters
- Objective 2: Develop TADF hosts
- Objective 3: Photo-physically characterise the shielded emitters and TADF hosts
- Objective 4: Anisotropic molecular orientation for enhanced performance
- Objective 5: Design and test prototype hyperfluorescence OLEDs
- Objective 6: Fabricate and evaluate demonstration hyperfluorescence microdisplays

To show the project's overall goal has been achieved, blue and white stack unit prototypes will be integrated into a high-brightness microdisplay demonstrator (based on MICROOLED's 0.38" WVGA CMOS backplane) and tested to demonstrate significant improvements in functionality, performance, manufacturability and reliability.

LEGAL NOTICE

Neither the European Commission nor any person acting on behalf of the Commission is responsible for the use, which might be made, of the following information.

The views expressed in this report are those of the authors and do not necessarily reflect those of the European Commission.

© HyperOLED Consortium 2017



1. Introduction

Organic light emitting diodes (OLEDs) are devices which convert electrical energy into light. The technology has made tremendous progress in recent years and is now found in most high-end smartphones, but also high-end TVs, where it replaces the still dominant liquid crystal display (LCD) technology. Though beautiful lighting applications are also possible with OLEDs¹, this area is still a niche market.

While OLED based displays are excellent in terms of display quality, there's still room for improvement in terms of efficiency, which is now roughly on the same level as or better than that of LCDs (depending on the exact application and content which is displayed). Most significantly, the blue emission is not as efficient as it could be, the reasons for this will be explained in the following. The HyperOLED project consequently targets the improvement of the blue efficiency of OLEDs by a novel approach.

1.1. Basics of OLEDs

OLEDs are very thin devices, usually their thickness is around 100 nm, which is roughly 1000 times thinner than a human hair. However, OLEDs have to be supported by a substrate (usually a glass or metal sheet) with a thickness of a few tenths of a millimeter and they have to be encapsulated to protect them from air which adds to the thickness of the display. Materials used in OLEDs are highly specialized organic molecules and the main focus of the HyperOLED project is the improvement of these materials.

The production of OLED based displays (and displays in general) is quite complex², but the basic process of forming an OLED is to heat the organic materials under vacuum³. The materials evaporate at a certain temperature (usually in the range of 250-350°C) and create a vapor which condenses on the substrate where a solid layer is formed. Usually, multiple layers (around 6-20 depending on the application) consisting of different materials are coated one after the other in this way to form the OLED stack. Some of the layers, especially the emissive layer which is responsible for light emission, contain two or three materials. The basic structure of a typical OLED device is shown in Figure 1⁴.

1.2. Basic Working Principle of Emission and Efficiency

In very simple terms, if current passes through an OLED, it emits light. Going more into the details, the current creates so-called excitons in the emissive layer of the OLED. These excitons contain energy which is released as light. The excitons come in two flavors, singlet and triplet excitons. 25% of the formed excitons are singlets and 75% are triplet excitons. The issue is that only singlet excitons can be used for light production in classical, light emitting organic materials (so-called fluorescent emitters). This means that most of the energy is lost.

¹ See for example https://www.oledworks.com

² You can for example visit the website https://www.oled-info.com/lg-announces-its-85-genoled-tv-fab-guangzhou-now-production to obtain some information on a typical display factory.

³ In Principle, printing of OLEDs is also possible, but not relevant for big scale mass production at the moment, see e.g. https://www.oled-info.com/joled-starts-sampling-printed-oled-panels-its-55-gen-nomi-site

⁴ For more information on how OLEDs work, you can visit the website from which the picture is taken: https://www.merckgroup.com/en/expertise/displays/solutions/oled-displaymaterials/oled-technology.html

You can also find information about OLEDs on Wikepdia: https://en.wikipedia.org/wiki/OLED

However, it was found around 20 years ago⁵ that it is in principle possible to make use of the triplet excitons by using phosphorescent emitters. In simple terms, triplet excitons are converted into singlet ones using an effect called spin-orbit coupling in such materials. Development of sufficiently stable and efficient materials was a complex task requiring considerable effort from the whole OLED industry. But it was successful and since a few years, the red and green emission in all OLED displays and lighting panels comes from phosphorescent emitters which greatly improved their efficiency compared to fluorescence based devices.



Figure 1: Structure of a typical (bottom-emission) OLED.

1.3. Improving the Blue Efficiency

However, despite huge efforts in the industry, no blue emitting phosphorescent materials with sufficient stability have been found, and so blue emission in OLEDs still comes from fluorescent emitters. As a result, more than half of the energy consumed for light emission is required to produce blue. If blue had the same efficiency as red and green, the whole display (not only counting the OLED, but also the electronics etc.) could be around 15-20% more energy efficient, which would be a further significant improvement.

In the last years, another way to improve the efficiency of OLEDs has been investigated extensively in the scientific community. It is based on the "thermally delayed activated fluorescence" (TADF) mechanism. The effect was described by Parker in the 1960s already (and named E-type delayed fluorescence), but only in 2012 Prof. Adachi's research group discovered that highly efficient OLEDs can be made based on the effect⁶. Basically, a very special molecular configuration is employed in order to transform triplet into singlet excitons and thus make them available for light emission. Starting from that point, many materials that show the effect have been discovered. Also, efficient blue TADF materials have been developed and in the HyperOLED project, we also investigate this type of materials to obtain efficient blue OLEDs.

⁵ DOI: 10.1038/25954

⁶ DOI: 10.1038/nature11687



Both TADF materials and phosphorescent emitters make triplet excitons available for light emission, however there are some drawbacks. In the case of TADF materials, saturated colors are very difficult to achieve due to the nature of the underlying physical mechanism. This is partially also true for phosphorescent emitters and in addition, stability of the materials is an issue in both cases. A way to tackle these problems is the combination of a TADF material or phosphorescent emitter with a conventional fluorescent emitter. These approaches are called hyperfluorescence (TADF combined with a fluorescent emitter) or hyperphosphorescence (a phosphorescent emitter combined with a fluorescent emitter). The idea is to use the TADF or phosphorescent emitter material to convert triplet into singlet excitons. The material responsible for the conversion is called a "sensitizer".

However, instead of using the TADF of phosphorescent emitter material for emission, the singlet exciton is transferred to the fluorescent emitter in a process called Förster energy transfer. The advantage is that fluorescent emitters can give very saturated colors and that they are well known, stable materials. However, there's one problem: The fluorescent emitter can "steal" triplet excitons from the sensitizer, which means they are lost for emission. This "stealing" process is also known as Dexter energy transfer.

In order to prevent this loss mechanism, the HyperOLED project investigates a unique and novel approach: The fluorescent emitter is modified on a molecular level in order to prevent the Dexter energy transfer. We also call this "shielding" the fluorescent emitter. The chemistry to achieve this and the scientific methods to investigate the underlying physics are highly complex and had to be developed during the project which lead to a lot of advancements in the understanding of TADF materials and the energy transfer processes involved.

2. Color Coordinates, Efficiency and Lifetime

In photometry, color is usually defined using "color coordinates" in a "color space" diagram. Most often, the CIE 1931 color space is employed⁷, the corresponding diagram is shown in Figure 2. In such a diagram, two numbers, called the CIE x and CIE y coordinates, define the color. For example, x = 0.67, y = 0.33 indicates a red color, while x = 0.21, y = 0.71 is green, x = 0.14, y = 0.08 is blue and x = 0.33, y = 0.33 white. A display usually contains pixels of the three primary colors red, green and blue. The brightness of the pixels can be adjusted individually and depending on their relative brightness, different mixed colors are obtained. These colors all lie inside the triangle which is defined by the primary colors, an example of this is also shown in Figure 2. It is desirable to make this triangle as big as possible so that a wide range of colors, the so-called "color gamut", can be displayed.

We target blue colors with high efficiency in the HyperOLED project. In modern displays, color coordinates of x = 0.14, y = 0.05 are usually considered to be excellent, but being a research and innovation project working on novel materials and concepts, we do not directly target such deep blue colors. We rather try to demonstrate high efficiency in combination with a sufficiently deep blue color to demonstrate the potential of the project's ideas.

Another important concept besides color is the brightness of a display. To define the brightness, it must be realized that the eye has different sensitivity for different colors. Green is perceived brighter than blue or red, and most of the electromagnetic spectrum is not visible at all (UV light for example, or infrared). To take this eye sensitivity into account, the "luminous function" V(λ) is used (see Figure 3). It describes how sensitive the eye is to

⁷ For a more detailed explanation see e.g.

https://en.wikipedia.org/wiki/CIE_1931_color_space



different wavelengths (colors) and there's a curve for day and night vision, but only the one for day vision (the red curve) is relevant for display applications. It can be seen that the eye is most sensitive for wavelengths around 550nm, which is in the green color region. This also means that a display which shows green content is perceived brighter than one with blue or red content if the same input power is consumed. When the brightness is measured by taking the eye sensitivity into account, the most commonly used measure is called "luminous intensity" with unit candela $(cd)^8$.







Figure 3: Luminosity function $V(\lambda)$ for day (red) and night (blue) vision.

Source: https://commons.wikimedia.org/wiki/Fi
le:V-lambda-phot-scot.svg

There are a lot of ways to define the efficiency of light sources in general. For displays, the so-called current efficiency is a common measure. It is obtained by dividing the luminous intensity by the current which is flowing through the display, its unit of measurement is cd/A. The higher this number the better, but it has to be noted that due to the eye's sensitivity curve, the current efficiency for blue or red is lower than for green.

Using the current efficiency, deeper blue devices would always appear to be less efficient than lighter blue ones, but this is not really the case, because to achieve the same overall display brightness, less luminous intensity is required in the case of deeper blue. Due to this, the external quantum efficiency (EQE) is often employed. It is a measure for the number of photons that leave the device divided by the number of electrons passing through it. In simple terms, it is a measure that is proportional to the color-independent light output divided by the current. It is usually given in percent and the higher it is, the better. Using the EQE, devices with different color can be compared more easily than using the current efficiency. Green and Red OLEDs in today's displays reach around 25% EQE, while blue ones have slightly more than 10%.

To judge the stability of OLEDs, life time measurements are carried out. The devices are driven with a constant current density and the emitted luminous intensity is recorded over time. The light output drops over time and the time it takes until the brightness has dropped

⁸ For more on the subject, see e.g. https://en.wikipedia.org/wiki/Luminous_intensity





to a certain fraction of the initial value is called the lifetime of the device. The higher the driving current density (the higher the brightness), the lower the lifetime. In the following, we present lifetime data for devices which were driven with a current density of 5 mA/cm² which is roughly what is used as a maximum in display applications. We define the time for which the brightness has dropped to 70% of its initial value as lifetime of the devices.

3. Optimisation Results

In order to obtain blue OLEDs that demonstrate the potential of the project's approach and materials, we chose the most promising material set that we developed and modified the device structure to obtain optimum performance. We focused on high efficiency in combination with as deep blue color and as high lifetime as possible. In a first step, the composition of the OLED emission layer and the other layers in the stack were adjusted for optimum performance in a standard test device. Subsequent to that, we employed two different device setups for further optimization, a bottom-emission OLED with thick electron transport layers and a top-emission OLED.

Bottom-emission OLEDs emit light through the transparent substrate on which they are deposited (see Figure 1). Such an architecture is used in today's OLED TVs. It has been shown previously that color purity can be optimized using an electron transport layer (ETL) of appropriate thickness⁹ for bottom-emission OLEDs. Top-emission architectures are used in OLED smartphone displays. The light is emitted away from the substrate which is usually non-transparent. The big difference to bottom-emission devices is that a thin, semi-transparent cathode is used which lets the light pass, whereas the cathode in bottom-emission devices is thicker, non-transparent and acts as a mirror.

3.1. Devices with Optimized ETL Thickness

In Figure 4, left, you can see how the efficiency depends on the thickness of the electron transport layer (ETL). A maximum EQE of 20% is reached for a thickness of around 180 nm.





Figure 4: Efficiency, given as EQE for various devices driven at 0.1 mA/cm² with different ETL thickness.

Figure 5: Current-voltage characteristics of an optimized device.

For one of the devices with top performance, more details are given in the following. Figure 5 shows the IV characteristics which is obtained when different voltages are applied and the current density through the device is measured. In Figure 6, left, you can see the

⁹ Böhm et al, IDW '09 - Proceedings of the 16th International Display Workshops



dependence of efficiency on driving current density while the graph on the right hand side shows the decay of the brightness over time when the device is driven with a constant current density of 5 mA/cm². From this curve, the lifetime can be determined to be 345 h (drop to 70% of the initial value). This is a good value for such a highly efficient blue device but clearly below state-of-the art fluorescence-based blue OLEDs (which however have much lower efficiency). The device shows blue emission with CIE color coordinates of x = 0.13 and y = 0.30. This is not sufficiently deep blue for typical display applications which is why we also employed a top-emission architecture for which better color coordinates can be obtained.



Figure 6: Efficiency versus current density (left) and decay of emission over time ("lifetime curve") for one of the top performing devices with optimized ETL thickness.

3.2. Top Emission Devices

Because layer thicknesses have a much higher impact on device performance in top emission devices as compared to bottom emission ones, we started the optimization by carrying out optical simulations of the OLED stack. The project partner Fraunhofer IOF determined the necessary input parameters and carried out the simulations. The goal was to find a configuration which gives good efficiency and color coordinates under the condition to use an aluminum cathode. This is not the ideal material for this application, usually silver or magnesium silver alloys are used. However, due to restrictions of the processing equipment we used in the project, this was not possible. This also means that we have not yet obtained the optimum performance and further improvement is in principle possible. From optical simulations we estimate that around 15% more efficiency is feasible without compromising other performance parameters if a silver cathode is employed.

The simulations show that the stack given in Figure 7 should give good performance. However, thicknesses in real devices do not correspond 100% to the optical simulations, and so it is necessary to fine tune the layer thicknesses experimentally. To this end, the thickness of the hole and electron transport layers are varied deliberately in small steps. This yields a variety of devices with differing performance. In a production process, the layer thicknesses that result in the best performance would be chosen. The thickness of the aluminum cathode was kept at a constant thickness of 14 nm.

Figure 8 shows the efficiency and color coordinates for several of the manufactured devices. As can be seen, a wide range of colors and efficiencies is obtained from the thickness variation. However, for deeper blue colors with CIE y around 0.20, the maximum achievable EQE is 17% which is realized for devices with a hole transport layer (HTL) thickness of 27 nm and an electron transport layer (ETL) thickness of 31 nm which is close to the values predicted from simulations.

The CIE coordinates of the corresponding device are x = 0.11, y = 0.21 which would be suitable for displays with limited color gamut only, but in the HyperOLED project the main



goal is to demonstrate feasibility of the investigated concepts, which is clearly achieved by the performance data we obtained. The device shows a lifetime 260 h which is lower than that of the bottom-emission device presented above. This is due to the fact that we used a slightly modified emissive layer composition which was optimized for efficiency and deep color at the expense of lifetime.



Figure 7: Basic layout of the top-emission OLED stack. ITO is Indium-Tin-Oxide, a very common material used for transparent electrodes in displays.

Figure 8: CIE y versus efficiency for a number of top emission devices driven at 0.1 mA/cm² with varying hole and electron transport layers.

As for the bottom-emission devices, more details of the device performance are presented in Figure 9, where the IV characteristics and the dependence of the efficiency on driving current density are shown. As can be seen, the drop of efficiency with increasing current density ("roll-off") is quite small. This is an important aspect because a lot of power is consumed when the device is bright (the current density is high), and so a high efficiency is crucial here.



Figure 9: Current-voltage characteristics (left) and efficiency versus current density (right) of a topemission device with optimized performance.

4. Conclusion

We have demonstrated blue OLEDs with good efficiency, color and decent lifetime using materials and concepts developed in the HyperOLED project. The efficiency is much higher than that of blue OLEDs used in today's products, but color purity and lifetime are not on the same level. Though the performance is not sufficient for immediate use in display or lighting applications, our results clearly demonstrate the potential of the HyperOLED approach for real world applications.