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Strategies for improving the efficiency of blue OLEDs for a lab-on-chip sensor

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ABSTRACT

A highly emissive blue organic light emitting diodes (OLED) for use as an algae excitation source in a biosensor was designed and optimized to meet spectral filtering requirements of the system. This source needs to exhibit high emission around 470-480 nm (algae absorption) combined with low emission in the algae fluorescence bandwidth (550-600 nm) in order to avoid any overlapping signal in the biosensor's sensitivity range. To address these issues, a microcavity device (MOLED) was studied and optimized. In order to further decrease the residual parasitic emission in the green spectral range, an additional filter was also integrated in the device. An improvement in peak intensity of 2.7 times the reference value was obtained, as well as a significant reduction of the parasitic emission in the green range. These improvements in peak intensity and spectral filtering should lead to a suitable blue OLED excitation source for compact optical biosensors.

Keywords: Blue OLED, integrated filter, biosensor, optical microcavity, multilayers.

1. INTRODUCTION

Nowadays, organic light emitting diodes (OLEDs) are already used in TV screens, smartphones, smartwatches, laptops, but they also generate growing interest for their integration into sensors, biosensors and biomedical applications (health monitoring, therapeutic treatment, optogenetics...) [1]. Indeed, they offer the unique opportunity to adapt the excitation source design to the required application functionalities in terms of: emissive area, wavelength, type of substrates (flexible, biocompatible...). These emerging application fields introduce new challenges to be considered in device fabrication, especially for blue emitting OLEDs. Actually, large area blue organic light emitting diodes (with surfaces much larger than the ones used in displays) stable at high brightness levels ($> 10\,000\text{ cd/m}^2$) with narrower emission spectra than conventional OLED have to be developed for biomedical applications. However, even though phosphorescent, TADF, and hyperfluorescent blue emitters exhibit significantly high efficiency, the first-generation of fluorescent molecules are still used in the displays because they remain more stable [2]. Furthermore, the devices based on phosphorescent and TADF molecules are not suitable for prolonged high luminance uses yet, mostly because its efficiency and stability at high luminance values decreases significantly, this is known as efficiency roll-off [3].

Here, we present the optimization of deep-blue OLEDs for their integration into a lab-on-disc (LOD) based biosensor for *in situ* monitoring of surface water quality. The aim of this LOD is to couple a network of biosensors which will measure the sensitivity of green micro-algae to toxins via its photosynthesis activity. Among them, optical transducers dedicated to chlorophyll-a fluorescence monitoring are needed. As described previously, these transducers lie in an excitation source and a photodetector on the periphery of a disc containing the biosensors (figure 1a) [4]. The constraints related to the LOD system require the development of robust blue OLEDs exhibiting simultaneously high brightness at 470 nm and no electroluminescence in the Chl-a fluorescence bandwidth (650-700nm see fig.1b) with areas comparable to the size of the LOD's analysis tanks ($2.5 \times 2.5\text{ mm}^2$). To address these specifications, the OLED stack was integrated into an optical microcavity and in order to further decrease the residual parasitic emission, an additional filter was integrated in the device.

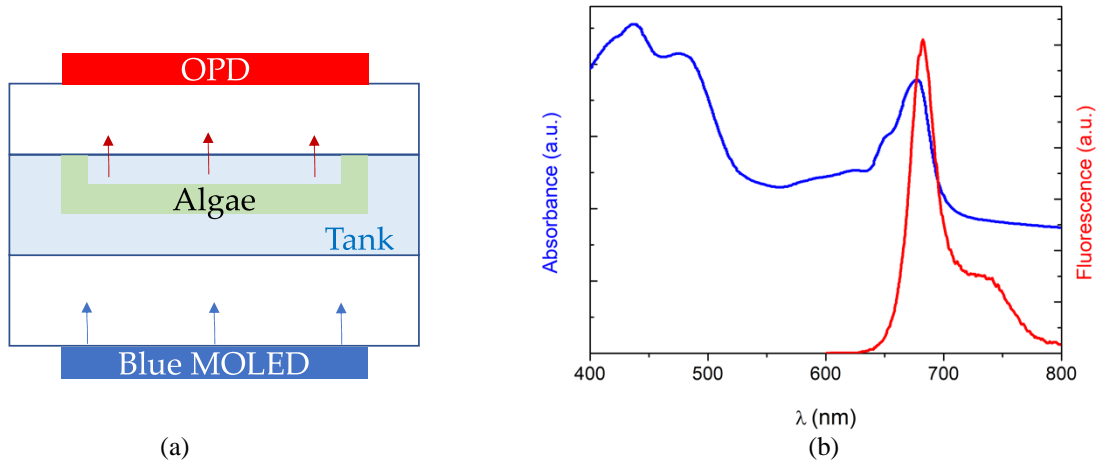


Figure 1(a): Schematic representation of the optical sensor (b) chlorophyll-a absorption and fluorescence spectra measured from *Chlamydomonas reinhardtii* micro-algae.

2. DESIGN OF OPTIMIZED BLUE MOLED

As seen in Fig 1a), in our lab-on-disc system, the algal fluorescence is detected by an organic photodiode (OPD) sensitive only in the NIR spectral range [650-750nm] [4] after excitation by an OLED source emitting in the blue [400-500nm]. As the emission spectrum of a standard OLED is very wide (FWHM~100nm), it overlaps with the sensitivity range of the detector, notably between 500 and 600nm. As a result, the use of a long-pass filter (minimal transmission below 560 nm) placed on the optical path before the OPD is mandatory but not sufficient. A second filter having a minimal transmission above 500 nm has to be added above the OLED. In this study, we investigated the possibility of reducing directly the emission spectral width of the OLED by replacing it by a microcavity OLED (MOLED). Exploiting spectral filtering effects in a low finesse vertical microcavity should lead to a reduced emission bandwidth (<30nm) and at the same time to an increase of the emitted intensity at the MOLED resonance wavelength (470 nm). This wavelength fits with the electroluminescence peak of the available organic materials for an efficient blue emission and with a peak of absorbance of the algae (474 ± 15 nm) (fig. 1b). In this microcavity, the mirrors surrounding the active zone are formed on one hand, by the standard aluminum bottom electrode of the OLED and on the other hand, by a dielectric DBR (distributed Bragg reflector), named here DBR₁ (fig 2a). This mirror is made of alternate thin layers having high index (Nb₂O₅) and low index (SiO₂) deposited on a transparent glass wafer before the ITO electrode deposition. In a standard DBR, the thickness of each layer is equal to a quarter wave of the design wavelength, that is to say, for 470nm: 49.8nm for the high index layer (H) and 79nm for the low index one (L). Doing so, we obtain for the DBR a band of high reflectivity centered at 470nm and for the whole MOLED a maximal transmission peak of the organic layers' emission through the glass wafer at 470 nm, provided the active organic zone thickness is chosen to be a multiple of a half-wave at the same wavelength and that the EML is placed at an anti-node of the stationary optical field to get constructive interferences at this wavelength [4].

As seen in fig. 3a, the calculated reflectivity spectrum $R(\lambda)$ of such a MOLED (structure 1) exhibits a reflectivity dip positioned at 470nm, as aimed. It is in principle possible to reach a reflectivity value close to zero at 470 nm, and thus a 100% MOLED top transmission, by playing on the thicknesses of the layers in the DBR and choosing them slightly different from usual high (H) and low (L) index quarter waves thicknesses. We chose here to design a specific stack (glass/H/L/H/L/1.5H/1.22L/ITO/organic layers/aluminum) leading to a minimal reflectivity of 5% at 470nm, because we added a second DBR, named DBR₂, on the back side of the glass wafer (fig. 2b). This additional DBR (glass/H/L/H/L/1.34H/0.84L/1.03H/air) is centered at 556 nm and its role is to suppress the parasitic transmission

of the MOLED in the green region corresponding to the edge of DBR₁'s stop band (fig. 3a). Figure 3b shows the theoretical reflectance of the global stack including DBR₁ and DBR₂ and the result of their interplay. As seen, the reflectivity of the MOLED with the back filter is now close to zero at 470nm and at the same time, it is strongly increased around 556 nm, as aimed for our lab-on-disc application. It is worth noting that this optical modeling takes into account the optical properties of all layers in the stack, including the organic layers and the electrodes (ITO and aluminum). Their refractive index and absorption coefficient were previously measured by spectroscopic ellipsometry on this purpose.

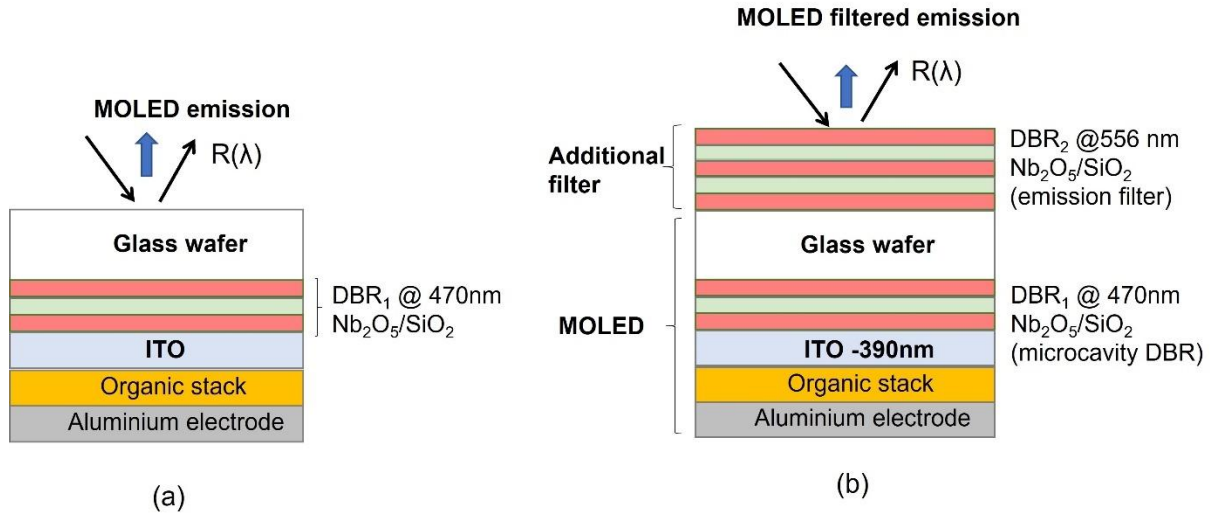


Figure 2 (a): Description of structure 1: MOLED stack with DBR₁ (air/glass/H/L/H/L/1.5H/1.22L/ITO/organic layers/aluminum); thicknesses: air/glass 1mm / 49.8nm / 79.0nm / 49.8nm / 79.0nm / 74.7nm / 96.2nm / ITO 390nm. (b) Structure 2: MOLED with additional filter DBR₂: glass/H/L/H/L/1.34H/0.84L/1.03H/air); thicknesses: glass 1mm / 60.6nm / 93.7nm / 60.6nm / 93.7nm / 81.2nm / 78.4nm / 62.5nm / air.

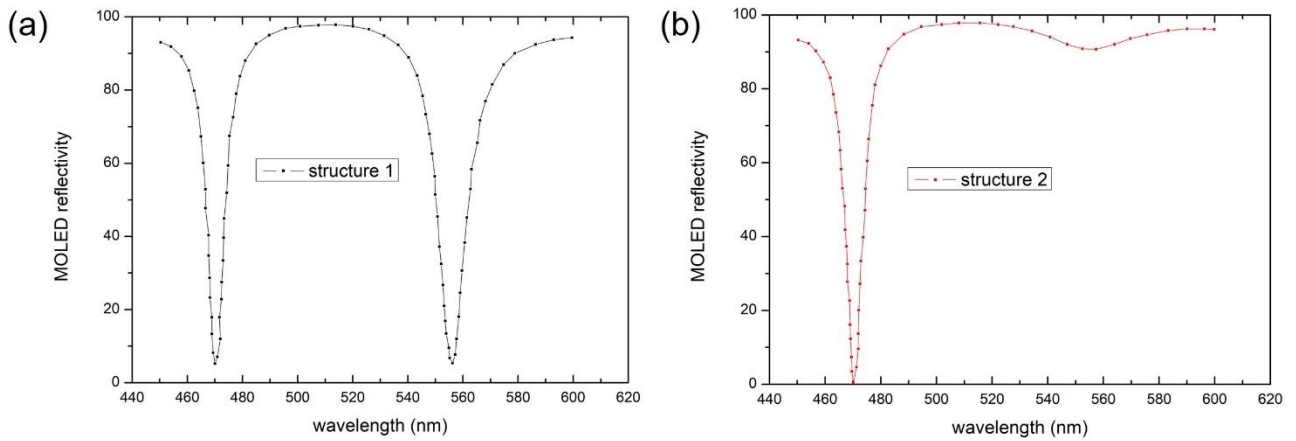


Figure 3: Calculated reflectivity spectra of (a) structure 1: MOLED with DBR₁ (470nm) (b) structure 2: MOLED with DBR₁ and DBR₂ (back filter at 556 nm).

3. EXPERIMENTAL RESULTS AND DISCUSSION

MOLEDs (structure 1) and MOLEDs with back filter (structure 2) designed above were fabricated using the following process. The 1mm-thick glass wafer substrates were first cleaned with acetone and isopropanol 10 min each in an ultrasonic bath before DBR deposition by plasma assisted reactive magnetron sputtering (Bühler HELIOS). In this work, we used a standard ITO electrode deposited by sputtering (390 nm; sheet resistance of 7 ohms/square). The organic layers were evaporated at 5.10^{-7} mbar with a rate of 0.03nm/s. The OLED stack was: HAT-CN (5nm) / NPB (55 nm)/ Tris-PCz (10 nm)/ MADN:DPAVBi (20 nm) / BmPyPhB (40 nm)/ Liq (1.5 nm)/Al (100nm). The MADN:DPAVBi emitting layer was obtained by the co-evaporation of MADN and DPAVBi with a 4% doping level, controlled by the deposition rate of the two molecules (0.1 and 0.004 nm/s respectively). Finally, the devices were encapsulated with a cover glass sealed with epoxy adhesive inside the glove box. A standard OLED was also fabricated using the same organic stack to serve as a reference.

The measured J-L-V (current density-luminance-voltage) characteristics of the devices are shown in Fig. 4. As seen, the turn-on voltage is similar for all of them, as expected. However, the luminance is found slightly lower for the MOLEDs than for the reference OLED. This is explained by the fact that the contribution to the emission of the undesired green wavelengths is significantly suppressed in the MOLEDs, especially in the MOLED integrating the back filter, whereas luminance measurements are performed with a photometer (calibrated with the photopic luminous efficiency function) having a maximal sensitivity at 555 nm i.e. in the green range [8] (see inset in fig. 4).

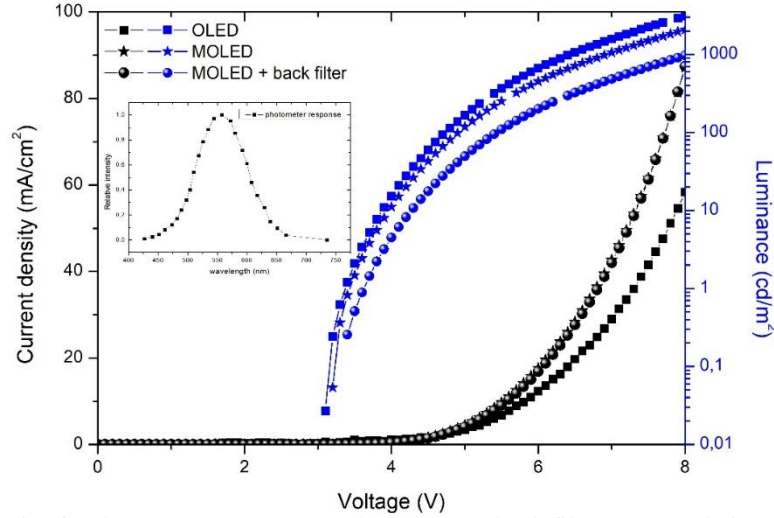


Figure 4: J-L-V characteristics for the OLED, MOLED, and MOLED + back filter. Inset: relative optical sensitivity of the photometer used for luminance measurements.

To confirm this explanation, we also measured the electroluminescence (EL) spectra of the devices (fig. 5). As seen, the maximal emission of the MOLED (structure 1, peak at 473 nm) is found higher (x3) than the one of the standard OLED (peak at 469 nm). A secondary emission peak is still observed in the green region but smaller than for the reference. These results are in quite good agreement with our modeling. As for the MOLED with the back internal filter (structure 2), it exhibits a nice Gaussian-like shape with a negligible emission in the range (550-600 nm), confirming the origin of lower luminance value and fulfilling well the requirements of our lab-on-disc. The emission peak is however detuned at 477 nm and slightly less intense than for the bare MOLED. This means the reflectivity is not exactly balanced between the two mirrors in the structure. We account this difference with the expected theoretical behavior to remaining residual uncertainties on the refractive indices and on the thicknesses of the organic layers. Nevertheless, the peak emission of structure 2 remains ~ 2.7 times higher than the one of the standard blue OLED. Furthermore, we calculated the integrated area of the different spectra shown in Fig. 5, in the blue spectral range [400-500] nm and in the green one [500-600] nm, respectively. As seen in table 1, the ratio of the light emitted in the blue range (algae excitation) over the one emitted in the green one (parasitic detection) is significantly increased by a factor

2.35 for the MOLED (from 0.73 to 1.72), and by a factor 5 for the MOLED with the back filter (from 0.73 to 3.51). These results clearly demonstrate the interest of using a MOLED combined with an integrated back filter for optimizing a blue source for lab-on-chip applications.

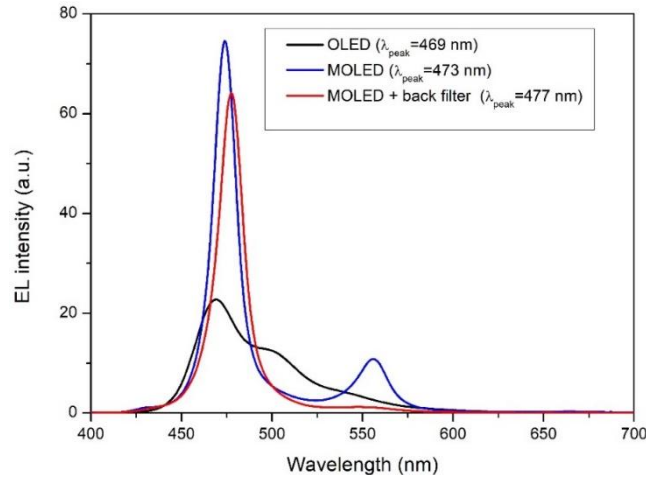


Figure 5: Relative EL spectra for the OLED, MOLED (structure 1) and MOLED + back filter (structure 2).

Table 1: Summary of OLED and MOLED parameters extracted from J-L-V characteristics and electroluminescence spectra

Device	V _{on} (V)	L _{max} (cd/m ²) @ 8V	λ _{peak} (nm)	I _{blue} / emission in algae absorption range [400-500] nm (integrated area, u.a.)	I _{green} / emission in parasitic detection range [500-600] nm (integrated area, u.a.)	Ratio I _{blue} /I _{green}
OLED reference	3.1	3100	469	64858	88851	0.7
Structure 1 MOLED	3.2	2084	473	110319	64452	1.7
Structure 2 MOLED + back filter	3.4	972	477	92270	26282	3.5

4. CONCLUSIONS

In this work, we demonstrated the possibility of producing blue OLEDs with reduced FWHM, increased emission in the blue range and suppressed light contribution around 550 nm to make their integration in a biosensor possible. By exploiting the optical properties of a MOLED and of a complementary DBR back filter, the peak emission intensity was increased by 2.7 times, while the ratio of light emitted in the blue range to the parasitic one in the green one was improved by a factor 5. Future work will consist in optimizing the stability of such device with the inclusion of additional layers in the organic OLED stack and in testing such device in the final lab-on-disc configuration.

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REFERENCES

- [1] Murawski, C., and Malte C. G. "Emerging biomedical applications of organic light- emitting diodes." *Advanced Optical Materials* 9.14 (2021): 2100269. DOI: 10.1002/adom.202100269
- [2] Monkman A. Why Do We Still Need a Stable Long Lifetime Deep Blue OLED Emitter? *ACS Appl Mater Interfaces* 2021;14. https://doi.org/10.1021/ACSAMI.1C09189/ASSET/IMAGES/LARGE/AM1C09189_0001.JPEG.
- [3] Murawski, C., Karl Leo, and Malte C. G. "Efficiency roll- off in organic light- emitting diodes." *Advanced Materials* 25.47 (2013): 6801-6827. DOI: 10.1002/adma.201301603
- [4] L.A. Lozano-Hernández et al, Design and optimization of a blue fluorescent microcavity-organic light-emitting diode (MOLED) on AZO anode for an algae excitation light source application, *Materials Today: Proceedings*, 2023. [//doi.org/10.1016/j.matpr.2023.07.173](https://doi.org/10.1016/j.matpr.2023.07.173).
- [5] Eynaud, Quentin, et al. "Toward Air Stability of Efficient Filter- Free Band- Selective Organic Photodetectors Based on Bulk Heterojunction: Avoiding Environmental Degradation with Atomic Layer Deposition Encapsulation." *Advanced Energy and Sustainability Research* (2024): 2300262.<https://doi.org/10.1002/aesr.202300262>