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Cost-Efficient Lighting devices based on Liquid processes and ionic Organometallic complexes

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1. Introduction

In this dissemination kit, apart from some of the most informing figures/pictures generated during the course of CELLO also some background information is presented to explain the nature of Light-emitting electrochemical cells (LECs) and their difference with organic light-emitting diodes (OLEDs).

In the next page also an executive summary of the achievements of the consortium during the last three years is included.



2. Executive summary of the project CELLO.

The European Project "Cost-Efficient lighting devices based on liquid processes and ionic organometallic complexes" (CELLO) has led to a strong improvement of the understanding, performance and market potential of light-emitting electrochemical cells (LECs). LECs are solution processed and molecule based light-emitting devices similar to OLEDs, but they make use of ions to overcome electronic injection barriers at the electrodes. The presence of the ions allows the use of air-stable electrodes and makes the devices less sensitive to thickness variations.

In the course of the project it was shown that ionic iridium complexes are ideal candidates to fulfill all functions of the LECs, greatly simplifying the architecture of the device. Approximately 50 new complexes were developed, synthesized, characterized and evaluated in LEC architectures leading to the identification of some very efficient and stable candidates which have been produced on the tens of gram scale at high purity.

Significant improvements in understanding the physical phenomena governing the operation of the LECs have been obtained which have been used to optimize the device layout and the electrical driving conditions. This has led to LECs with sub-second turn-on in combination with several thousands of hours of lifetime at high initial luminances (> 1000 cd/m^2), which is a major improvement compared with pre-project state of the art performances.

Demonstrators were successfully prepared on large and small area substrates. High efficiencies and stabilities reaching 17 lum/Watt and a lifetime in excess of 6000 hours (at an initial luminance of 1000 cd/m²) have been demonstrated.

Using embedded grid lines, large areas (210 cm²) flexible LECs were prepared on a roll-to-roll (R2R) coating line. It was also shown that grid lines can be prepared using printing techniques compatible with R2R. Small area demonstrators were prepared on these printed grid lines using a multi-layer PEDOT: PSS stack as an interlayer, showing the potential of this approach.

The LECs can be encapsulated by laminating commercially available barrier foils on the substrate and on top of the device. This leads to lifetimes of several hundred hours in ambient conditions. These simple encapsulated LECs are significantly more stable than similarly encapsulated OLEDs showing the benefits of using ion assisted electronic charge injection in combination with air-stable electrodes.

Based on the processes developed and the demonstrator evaluation, a feasibility study was performed. This study showed that due to the robustness of the LEC architecture R2R processing equipment can be implemented at low investment costs. This makes the production of LECs profitable at much lower production volumes (when compared to OLED and LED production) allowing for the targeting of smaller markets and reducing economic risks. Hence, manufacturing sites are possible in Europe and, as a consequence, the technology development can keep pace in Europe in the long term.

As a result of the project two patent applications were filed. Additionally, in the course of the project 49 papers in high impact scientific journals were published. A total of 27 oral presentations at national and international conferences have been given by the partners. CELLO started on 1-1-2010 and ended on 31-12-2012. More details can be found on the website: www.cello-project.eu.



3. Background.

LECs and organic light emitting diodes (OLEDs) (Figure 1) are both thin film electroluminescent devices.

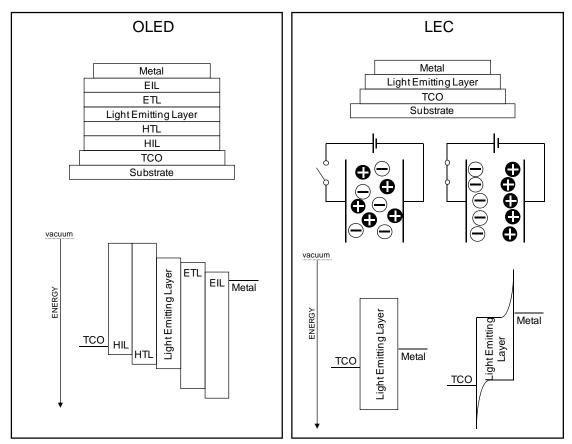


Figure 1. Schematic structure of an OLED (left) and a LEC (right). TCO=transparent conductor, HIL=hole injection layer, HTL=hole transporting layer, ETL=electron transporting layer, EIL=electron injection layer. An **OLED** is prepared by thermal vacuum evaporation and consists of several layers, each with a specific function. The injection of electrons in OLEDs is achieved by the use of i) a low work function metal or ii) an electronically doped electron injection layer, both are unstable in air and require rigorous encapsulation. A **LEC** is a single-, or at most, a double-layer device and consists of positive and negative ions (both typically optoelectronically active) that are displaced when an external bias is applied. Upon displacement an interfacial field is generated that allows for efficient hole and electron injection from air-stable metals.



4. Most significant results.

4.1 Materials.

Typical ionic transition metal complexes developed and upscaled during the project:

Figure 1. Representative ionic transition metal complexes developed and upscaled in CELLO.

4.2 Devices.

A lot of effort has been devoted to the identification of the operation mechanism of LECs using ionic transition metal complexes. Our findings indicate that, when a bias is applied to LECs, doped layers are formed dynamically at both electrode interfaces, which gradually move towards the center of the devices (as schematically depicted in the figure below).



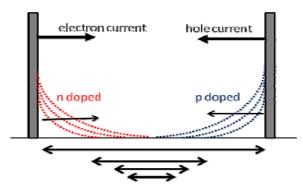


Figure 2. Illustration of the growth of p and n doped regions over time that decrease the effective thickness of the device.

Significant improvements in device performance were obtained during CELLO, the most stable LECs have a lifetime > 6000 hours at an initial luminance > 1000 cd/m² (see Figures below).

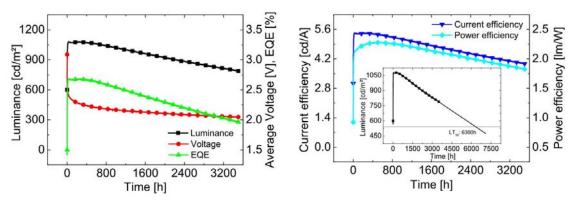
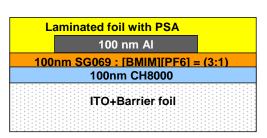


Figure 3. Device performance for SG069 under optimized conditions: ITO / 60nm Al4083/ 10 nm IL / 100nm SG069:[BMIM][BF4] = 3:1 / 150 nm Al; Pulsed current operation: 0V / 66.6 mA cm^{-2} ; 1kHz; 30% duty cycle; avg. 20 mA cm^{-2} .

In the course of the project it was confirmed that LECs are significantly less sensitive to ambient conditions when compared with OLEDs. This allows for less demanding encapsulation options (see Figure below).





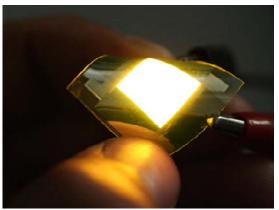


Figure 4. Device structure of fully flexible LEC devices (left). Photo of flexible LEC with an active area of $^{\sim}$ 1.5 cm². Acronyms used are the following: ITO stands for Indium Tin Oxide (transparent conductor), CH8000 is the conductive polymer formulation (PEDOT:PSS), SG069 is the ionic iridium complex, BMIM:PF $_6$ is the ionic liquid, Al is the top aluminum electrode, PSA stands for pressure sensitive adhesive.

4.3 Large area Processing.

In CELLO a lot of infrastructure for large area solution based processing was available and has been developed. See figures below:



Figure 5. ROKO flexo print pilot line at VTT.





Figure 6. Image of part of the R2R coating line at OSRAM (inset shows a 14 by 15 cm prototype LEC produced on the line).

To enable large area devices on ITO coated PET, additional grid lines need to be present. R2R inkjet and flexography printed gridlines were developed in CELLO (see Figure below).

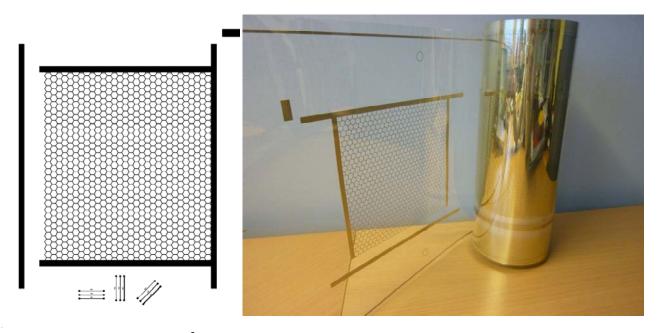


Figure 7. Layout of $15x15 \text{ cm}^2$ honeycomb grids (left) and picture of a flexo printed grid foil (right).



Grid line containing substrates were also used to prepare large are LECs on the R2R coating line. An images of this large area demonstrator (210 cm²) can be seen below.



Figure 8. Photograph of a R2R coated large area (210 cm²) prototype LEC.