



# Large area self-powered semitransparent trifunctional device combining photovoltaic energy production, lighting and dynamic shading control

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## ABSTRACT

We present a large area trifunctional glass prototype combining a photo-electrochromic (PEC) device and an organic light-emitting diode (OLED), interfaced through a properly designed electronic control system. A  $12 \times 17 \text{ cm}^2$  PEC glass-on-glass module was realized, containing four dyesensitized solar cells (DSSCs) and a central electrochromic (EC) section deposited on the same glass panel. All PEC layers are screen-printed, including the mesoporous electrochromic layer, obtained from a custommade tungsten paste. DSSCs show an efficiency of 2.4%, while the coloration efficiency of the EC section reaches a value of  $40 \text{ cm}^2 \text{ C}^{-1}$  at 700 nm. A  $10 \times 8 \text{ cm}^2$  transparent white OLED was also realized, designed and tailored in order to unbalance the emission of light, i.e. maximizing the bottom emission. The efficiency of large area OLED section reaches 8 cd A<sup>-1</sup> in the operative conditions and without light outcoupling enhancement systems. The OLED device is clamped on the back of the PEC module and all sections are electrically connected to an external electronic control system. The energy collected by the DSSCs is stored in supercapacitors and used when requested, either applied to the EC section to produce a light shading effect in the daytime, or to the OLED for illumination at night.

## 1. Introduction

In less than 30 years after their first practical demonstration [1], OLED (Organic Light-Emitting Diode) technology has attracted high relevance from both research and commercial point of view. Currently, the general aim goes towards the realization of more efficient devices with specific structural features: large emission area, flexibility, transparency and longer lifetime at high initial luminance. Different approaches are possible to fabricate white OLEDs for the lighting sector such as using three emitting materials (one for each color, red, green and blue) within a single emissive layer [2], or using different emitting layers for each color [3]. To achieve high luminance and long lifetime "stacked" OLEDs (or tandem OLED) were proposed, consisting of two or more organic stacks in series, interfaced by a charge generation layer [4]. The state of the art in OLED display currently shows high luminous efficiency [5], and small area OLEDs ( $6.7 \text{ mm}^2$ ,

not transparent) with power efficiency close to  $90 \text{ lm W}^{-1}$ , including optimized light outcoupling systems, have been fabricated [6]. Still, the requirements for lighting applications are even more stringent and upscaling the process towards large emitting area introduces significant issues mainly related to the high sheet-resistance of transparent anode, with a negative impact on device performance, both in terms of efficiency and emission uniformity [7]. Innovative devices that combine OLED functionality with photovoltaic (PV) response are of big interest, as they can generate electricity in the daytime and provide illumination at night. However, the conflictual requirements on separation/recombination of electrons and holes for OLEDs and PV make dual-function structures very challenging, therefore hybrid integration in stacked and separated geometries has been proposed for high efficiencies [8,9].

Furthermore, in the last decade industrial research focused its attention on "smart" advanced architectural glazing based on EC (electrochromic) devices [10] that could improve energy efficiency in

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buildings, with modulated transmittance of light upon externally applied potential and dynamic solar control [11,12]. EC glass windows can provide control of solar light glare and a variable visual connection with the external world [13,14], enhancing the human comfort of the internal environment, and - at the same time - regulating the solar energy transfer through the window, reducing the expense for building heating and cooling. Also in this case, an interesting evolution of this technology is represented by an integrated photovoltaic/electrochromic (PV/EC) device [15]. In fact, a photo-electrochromic (PEC) window could function both as solar cell module and as self-powered electrochromic smart window, able to provide a shading control, thus creating new market and business opportunities in the field of green energy applications in buildings.

According to the aforementioned considerations and stand to our knowledge, the integration of two technologies, OLED/PV or PV/EC has already been proposed, while no study has been published on the development of a trifunctional device involving “smart” panels in which energy production (PV), lighting (OLEDs) and solar control/sun screening (EC) are combined.

The work reported in this paper concerns the construction and characterization of a glass-frit sealed  $12 \times 17 \text{ cm}^2$  semitransparent photo-electrochromic device, combined with a transparent white OLED panel in a stacked configuration. In particular, a transparent large active area ( $10 \times 8 \text{ cm}^2$ ) white OLED was realized and was stacked on a PEC module that combines four DSSC (dye-sensitized solar cells,  $0.75 \times 10 \text{ cm}^2$  each) and an electrochromic section ( $10 \times 10 \text{ cm}^2$ ) on the same glass panel. A dedicated external electronic circuit was developed for energy management, based on three different microcontrollers that work independently from each other to maximize the functional flexibility of the prototype. The main target is to demonstrate the technical feasibility of an innovative trifunctional module that combines devices of different nature, thus optimization of each subcomponent was beyond the scope of this work.

## 2. Material and methods

### 2.1. PEC assembly

Fluorinated Tin Oxide (FTO) conductive glass substrates (Pilkington TEC15, 4 mm,  $12 \times 17 \text{ cm}^2$ ) were cleaned by dipping in a 2% Hellmanex solution in ultrapure water for 5 min and rinsing by ethanol and acetone. Two substrates per device were used, previously laser-scribed to provide the correct electrical isolation. A pair of holes (1 mm diam. approx.) was drilled on the cathodic back glass substrate for each DSSC/EC section, to be used later for filling the sections with the dye solution or the liquid electrolyte. The cathodic catalytic layers were obtained by screen-printing a commercial nano-platinum paste (PT-105030611 3DNano). Ag fingers were printed (Ferro GSSP SP 1963 Ag Paste) between adjacent sections in order to reduce the charge transport resistance of each cell. The electroactive layers of the DSSC sections were printed using a  $\text{TiO}_2$  commercial paste (18NRT Dyesol). An electrochromic  $\text{WO}_x$  printable paste was prepared by dissolving 0.5 g of  $\text{WCl}_6$  ( $\text{WCl}_6$ , > 99.9%, Sigma-Aldrich) in 20 ml of ethanol (Absolute Ethanol, 99.8%, Sigma-Aldrich) solution at 50 °C for 24 h, containing a  $\beta$ -diketone complexing agent such as acetylacetonate (0.4 ml) [16,17]. Then the resulting suspension was concentrated and mixed with 8 g of terpineol (anhydrous, mixture of isomers, Sigma-Aldrich) and an ethanol solution of ethyl cellulose (Sigma-Aldrich, 46 cp in 20/80 toluene/ethanol) in ethanol (1.6 g in 16 ml). The suspension was alternatively sonicated and stirred for several hours. Then, after removing ethanol under reduced pressure the paste was obtained, having 3–5% w/w  $\text{WO}_x$  content. A screen-printable glass frit (GF) paste [18] was used as sealant on all sections of the device. A high-accuracy Screen Printer (Aurel Automation 900) was used to print Pt, Ag,  $\text{TiO}_2$ ,  $\text{WO}_3$  and GF layers on the FTO-coated side of the substrates. All deposits were left at rest for 30 min at room temperature, then dried

between 50 and 100 °C (FOC 70 C FOCUS). Both the anodic and cathodic substrates were subjected to a thermal treatment in air, during which the temperature was linearly raised to temperatures from 450 to 500 °C. All layers were sintered at the same temperature in a single firing step. During the first part of the thermal step, the organic components of the screen-printed inks were carbonized and volatilized, then sintering of the Pt, Ag,  $\text{TiO}_2$  and  $\text{WO}_3$  layers occurred and the GF underwent partial fusion. A slow cooling to room temperature was performed. After overlaying the cathode and the anode glass substrates, so that the two GF tracks match and the  $\text{WO}_3/\text{Pt}$  and  $\text{TiO}_2/\text{Pt}$  electrodes faced each other, a bonding thermal step was performed at 500–550 °C to promote GF softening and fusing. PEC devices were transferred into an oxygen- and humidity-free glove box (MBrawn) filled with nitrogen ( $\text{O}_2$  and  $\text{H}_2\text{O}$  concentrations < 0.1 ppm). The holes on the back glass were used as input and output ports for colouring and electrolyte insertion. Dyeing of the DSSC sections was made inside the glove box by flushing 4 h a solution of G3 dye [19] in tetrahydrofuran (THF, Sigma Aldrich) using a syringe pump (NE-4000). DSSC electrolyte was a sulfonate and ionic liquid-based electrolyte [20,21] containing the redox couple  $\text{I}^-/\text{I}_3^-$ . As electrolyte on the EC section we used a ionic liquid-based electrolyte having the following composition: 0.05 M HMIImI (1,3-hexyl-methyl imidazolium iodide), 2 M LiTFSI (lithium trifluoromethanesulfonimide) in EMImTFSI (1,3-ethyl-methyl trifluoromethanesulfonimide). Both electrolytes were manually injected in the cells using a plastic syringe. A two-component commercial epoxy resin (Torreseal) was used to seal the holes. After resin curing (> 8 h), all cells were cleaned, inspected and made available for electrochemical and optical characterization.

### 2.2. OLED assembly

The OLED devices have been fully fabricated by high vacuum thermal evaporation in a Kurt J. Lesker multiple chamber system with a base pressure around  $10^{-7}$  mbar, without breaking the vacuum and employing suitable shadow masks, that define the device active area. Two commercial phosphorescent Iridium complexes have been employed in each electroluminescence unit: iridium(III)[bis(4,6-difluorophenyl)pyridinato-N,C2]-picolinate (FIrpic) and Bis(2-methylidibenzo [*f,h*] quinoxaline)(acetylacetonate)-iridium(III) ( $\text{Ir}(\text{MDQ})_2(\text{acac})$ ) for blue and red-orange emitting layers (EL), respectively. FIrpic and  $\text{Ir}(\text{MDQ})_2(\text{acac})$  have been dispersed into two different host material: 2,2',2'' (1,3,5-benzenetriyl)tris(1-phenyl-1H-benzimidazole) (TPBi) for blue emitter and 4,4',4''-tri(N-carbazolyl) triphenylamine (TCTA) for the red one. In order to confine the excitons recombination within the emitting region, two thin layers of TPBi and N,N'-diphenyl-N,N'-bis(1-naphthylphenyl)-1,10-biphenyl-4,4'-diamine (NPB) have been used as hole and electron blocking layers (HBL and EBL), respectively. Finally, a film of N,N,N',N'-Tetrakis(4-methoxyphenyl)benzidine (MeoTPD) doped with 2,3,5,6-Tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F4TCNQ) acts as hole transport layer (HTL) and a layer of Bathophenanthroline (BPhen) doped with Cesium (Cs) acts as Electron transport layer.

### 2.3. Measurements

All sintered PEC layers were measured by a contact profilometer (Veeco DEKTAK 150).  $\text{WO}_3$  layers were characterized by XRD analysis (Bruker AXS, Rietveld method using TOPAS v4), and field emission scanning electron microscopy FE-SEM (S4500 Hitachi, Working Distance = 10 mm@20 kV, magnification 1000–200000X). I-V curves on DSSC sections were taken using a solar simulator (Oriel SOL 3A™ Newport system) set to 1.5 AM and 1 sun illumination. During measurements, DSSCs were kept at 25 °C by clamping them onto the plate of a Peltier cooling system. IV curve measurement was performed at a potential scan rate of  $2 \text{ mVs}^{-1}$ , starting from open circuit potential going towards 0 V. Electrochemical impedance spectroscopy (EIS),

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