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ELECTRONIC SUPPLEMENTARY INFORMATION (ESI)

Fully Integrated Electrochromic-OLED Devices for Highly Transparent Smart Glasses

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We took particular care to address the relevant problems associated with the employing of unreliable and unstable liquid or semisolid electrolytes and to ensure that the engineering design of the device architecture was carefully assessed preventing the formation of undesirable short-circuits paths. In particular, the processing of materials constituting the EC and OLED systems were fine-tuned to guarantee the best device performances as well as environmental and operational stability. High-resolution SEM images of EC and OLED cells (Fig. S1) clearly show the monolithic system of the integrated device, highlighting the features of the nanostructured organic light emitting diode layers (Fig. S1 (d)), and electrochromic WO_3 film deposited on polymer Nafion film (Fig. S1 (c)). In Fig. S1 (b) and (d), is also well noticeable the intermediate ITO film (325 nm thick) deposited on top of polymer electrolyte and acting as starting layer for OLED growth.

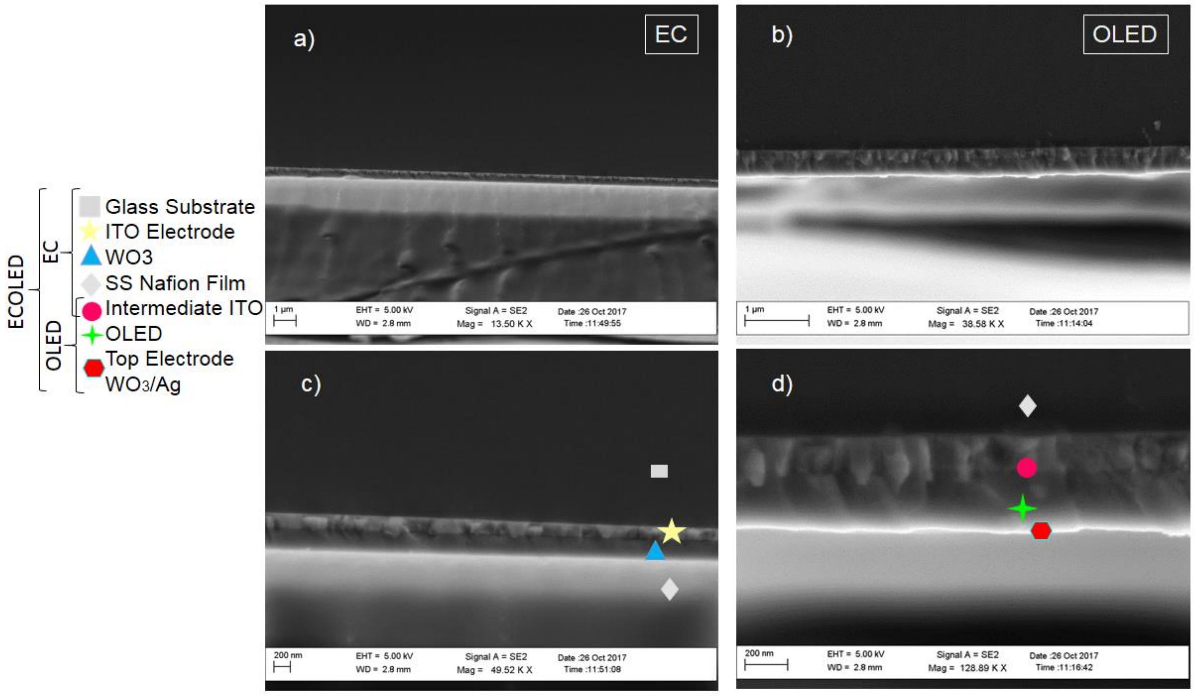

Fig. S1. SEM images at different magnifications of (a, c) EC and (b, d) OLED structures for integrated ECOLED device.

In figure S2, we show the scheme of the OLED device architectures fabricated for the optical modeling analysis. We perform the simulations on TC electrode by simultaneously tuning the thickness of W_b (WO_3 bottom), W_t (WO_3 top) and Ag layers, keeping the total thickness of the whole underlying organic stack to 115 nm (50 nm for HTL, 15 nm for EML, 50 nm for ETL) and ITO to 75 nm.

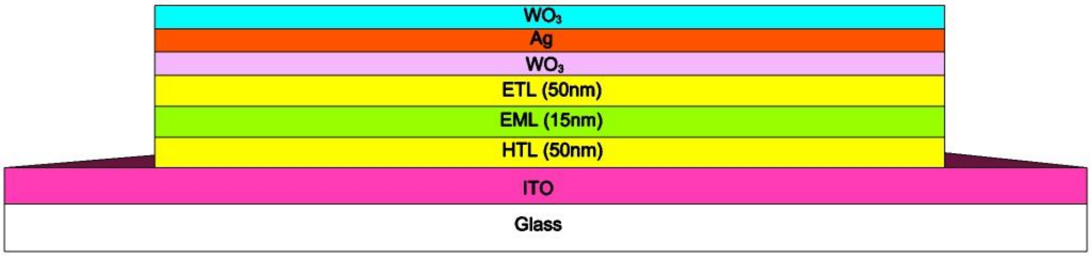

Fig. S2. Schematic view of the OLED device layout architectures fabricated for optical modeling analysis.

As reported in Table S1, the values of main electrical parameters obtained for RF-sputtered ITO films with thicknesses of 300 nm, 150 nm and 75 nm were compared with those obtained for commercial ITO glass having a 150 nm thick ITO layer (Visiontek). Thinner RF sputtered ITO films (75 and 150 nm) were prepared using the same process conditions described in the Experimental section for 325 nm thick ITO films. Unfortunately, 75nm and 150 nm RF sputtered ITO films showed higher sheet resistance, making them unsuitable candidates as intermediate electrode layer of EC cell in ECOLED device. Furthermore, as reported in XRD analysis of our previous work [36], contrary to ITO film with thickness of 300 nm, ITO film with lower thickness (150 nm) showed broad diffraction pattern with no characteristic peaks clearly highlighting its amorphous nature.

Tab. S1 Sheet resistance (Rs) values and transmittance value (% T @ 550 nm) of commercial ITO, 75 nm, 150 nm and 300 nm thick RT sputtered ITO films.

	Thickness [nm]	Sheet Resistance [ohm/square]	Transmittance [% T @ 550 ITO]
Commercial ITO	150	12	97
RF sputtered ITO	75	55	75
RF sputtered ITO	150	26	76
RF sputtered ITO	325	13.6	77

In Fig. S3a we show the transmittance of OLED A (green curve), which is in line with the theoretical calculation. It is worth to note that the transmittance overcomes the 75% from 410 nm up to 610 nm, with a peak of 87% at 450 nm. In Fig. S3b, three pictures in OFF and in ON states under different applied voltage (4V, 6V), are reported. At intermediate voltages (i.e. 4V) the background is still visible through the emissive area, whereas saturation occurs at 6V. The opto-electronic characterizations of the OLED A device are reported in Fig. S3c. Luminance above the minimum required for display application (300 cd/m^2) and lighting (above 800 cd/m^2) are obtained for OLED A, at 9 mA/cm^2 and 27 mA/cm^2, respectively. The current efficiency starts from a value of 6 cd/A, rolling off to 2.5 cd/A at high luminance, probably due to triplet-triplet annihilation phenomena.

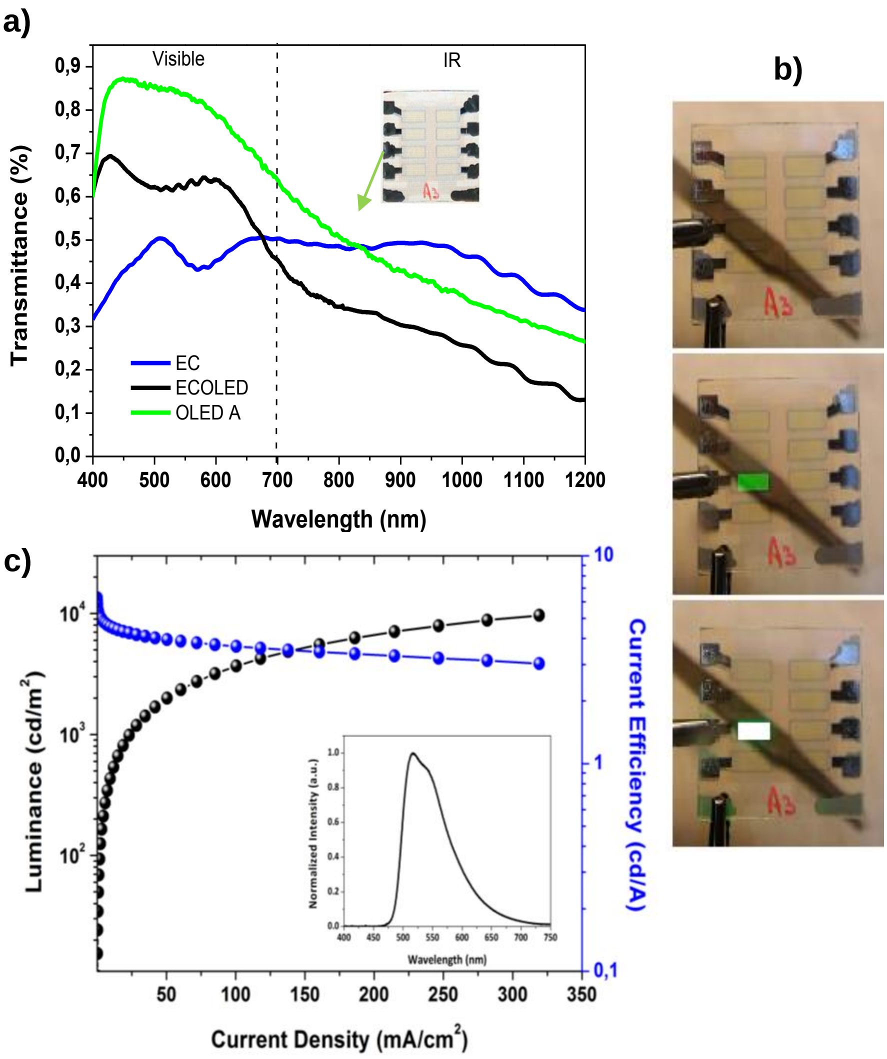

Fig. S3 (a) Transmittance spectra of the OLED A and white ECOLED in OFF state, illuminated from the backside by white light, compared with that of SS-EC device. (b) Picture of OLED A, fabricated on glass substrates with the architecture reported in the simulation section, in OFF and ON states under different applied voltage (0V, 4V, 6V). (c) Opto-electronic characterization of OLED A.

To obtain a more immediate view of the optical modulation in the visible region and provide insight into the optical property of the integrated ECOLED device, each wavelength was mapped with respect to potentials applied for colouration and bleaching processes. The resulting optical variations were compared with those of EC transmittance modulation mapping data, as shown in Fig. S3. ECOLED device exhibited higher bleached/coloured transmission modulation of c.a. 10% in the visible region (400 nm -800 nm) than EC device with a maximum increment of about 20 % in the range between 450 nm -500 nm and from 550 nm to 600 nm.

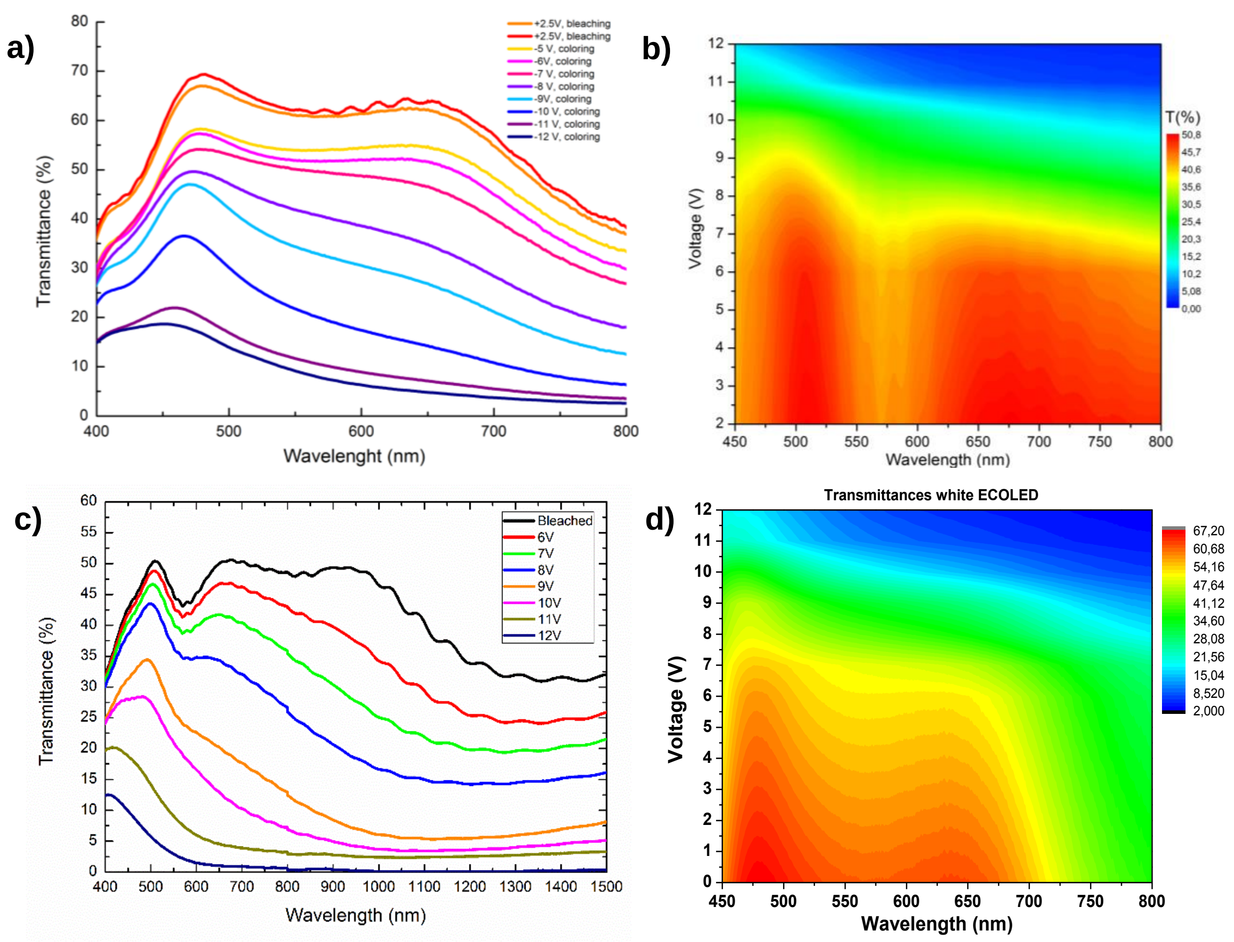

Fig. S4. (a) Optical transmittance spectra of white ECOLED device in the range of wavelengths between 400 nm and 800 nm, with applied potentials in a range between 6 V and 12 V and -2.5 V for bleaching and coloration states, respectively. (b) Intensity color map (wavelength vs applied voltage) of white ECOLED device. (c) Optical transmittance spectra of SS-EC device in the range of wavelengths between 400 nm and 1500 nm. (d) Intensity color map of SS-EC device (wavelength vs applied voltage).

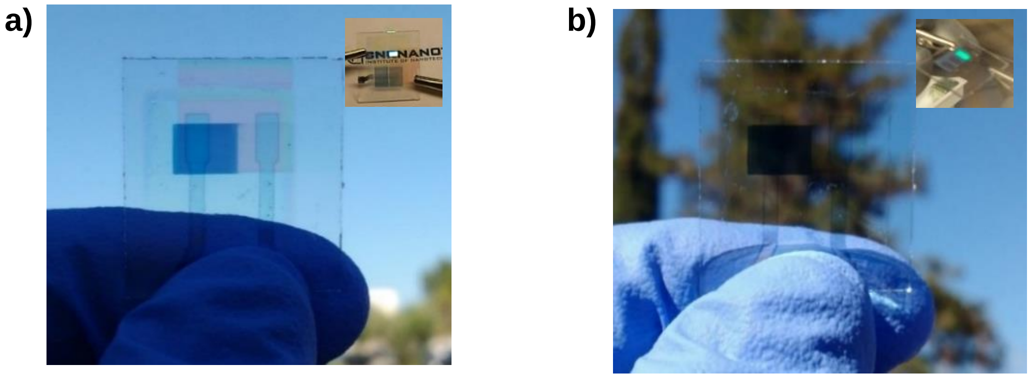

Fig. S5. (a) Pictures of integrated white (left side) and (b) green (right side) EC-OLED devices in coloured and bleached states.

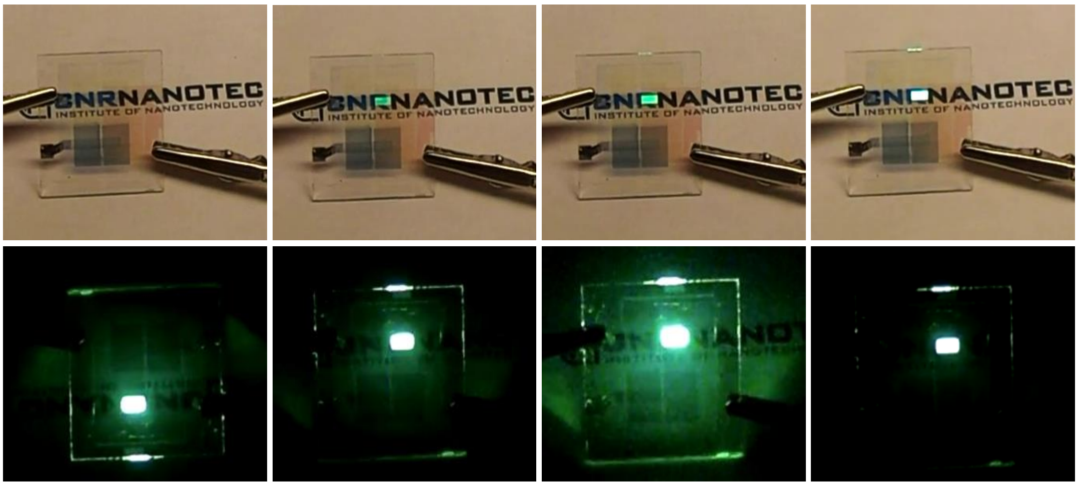

Fig. S6. Photographs of white ECOLED devices in ON state (OLED function) under beached and colored states (EC function) for light and dark conditions.

The opto-electronic characterizations of the white ECOLED device are reported in Figure S7. A maximum luminance value of 350 cd/m2 at the voltage of 10 V. The turn-ON voltage was about 3 V, in line with the highest gap of the used materials.

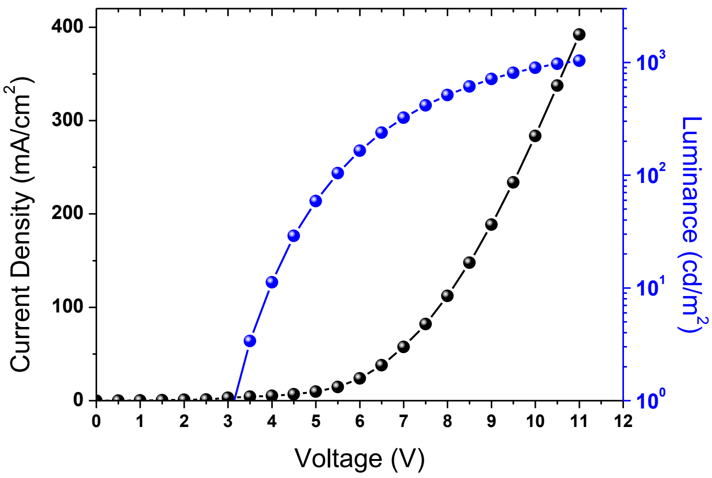

Fig. S7 Electro-optical characteristics of white ECOLED: current density and luminance versus applied voltage.

Fig. S8 reports the characteristics of a multifunctional ECOLED device operating in reflection mode usable for a wide range of electronic systems such as self-dimming mirrors and low-power reflective displays. Taking into account the layout architecture of Fig.1, the reflective ECOLED device was obtained by replacing the top OLED electrode with a thin Aluminum (Al) layer (100 nm) evaporated at RT conditions (pressure: mbar; deposition rate was about Å/s; power was about 9%). Contrary to the conventional EC mirrors, so far reported in literature, in which the reflective state is given by one of the two electrodes of the EC device, in our case this was provided by the cathode of the OLED superimposed over the fully transparent device.

Fig. S8a shows the reflectance spectrum of the integrated device reporting a reflectance of about 7% at 650 nm in the reflective state and a maximum reflectance modulation (ΔR) of 17.5 % at 650 nm and of approximately 30% at 460 nm. These ΔR values were respectively obtained by applying a potential of -2.5 V and +12 V, and a reasonable modulation was also observed at lower potentials (e.g. ΔR of about 10% under an applied potential of 6 V). In addition, the analysis of the chronoamperometric measurements (Fig. 8b) exhibited similar electrical features with that obtained for the full transparent white and green ECOLED devices: low charge/discharge absorptions, fast kinetics and reproducible colouration and bleaching processes. Coloration switching response times are obtained by applying different biases, where a driving force of at least 8V was required to activate 90% of the maximum ΔR, as shown in Fig. 8c. Full coloration states are achieved in a time lapse of 90-120 s, whereas lower switching times in a range of 30s and 60 sec are required to pass in reflective (bleached) states by applying a bias of -2.5 V.

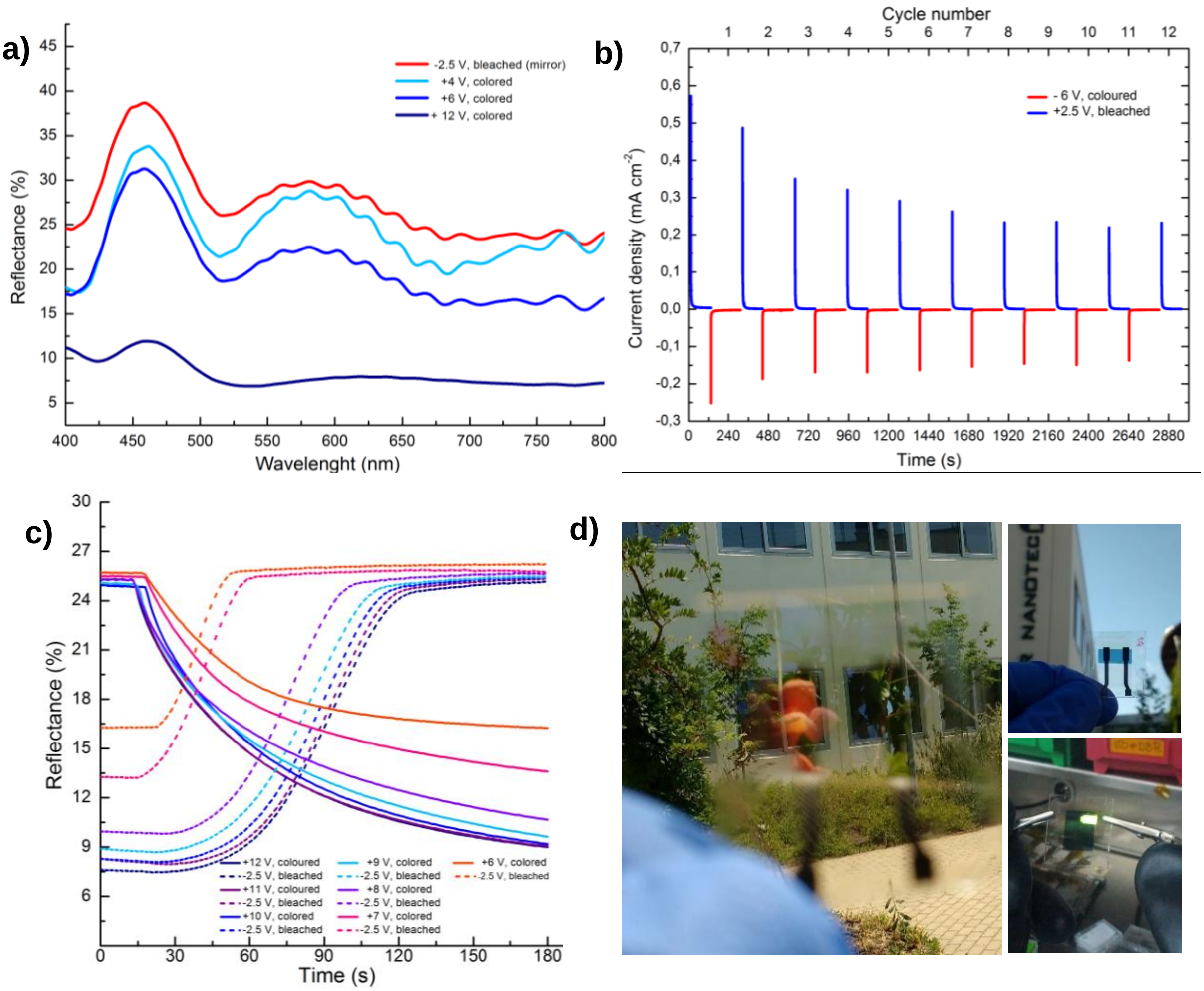

Fig. S8 (a) Optical reflectance spectra under colored and reflective states in the visible region (400 nm – 800 nm) and (b) chronoamperometric curves of fully integrated switchable mirror and lighting ECOLED device. (c) Color switching time measured at a wavelength of 650 nm with applied potentials in a range of 6 V- 12 V and of -2.5 V for colouring and bleaching processes. (d) Pictures of the integrated reflecting ECOLED devices in reflective (bleached), colored states and in ON (OLED) state under colored condition.