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PAPER

A light-emission textile device: conformal spray-sintering of a woven fabric electrode

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Abstract

We report on the realization of an ultra-flexible, light-weight and large-area emissive textile device. The anode and active material of a light-emitting electrochemical cell (LEC) were deposited by conformal spray-coating of a transparent fabric-based electrode, comprising a weave of fine Ag-coated Cu wires and poly(ethylene naphthalene) monofilament fibers embedded in a polyurethane matrix. The yellow-emitting textile featured low turn-on voltage (5 V), high maximum brightness ($>4000 \text{ cd m}^{-2}$), good efficiency (3.4 cd A^{-1}), and reasonable lifetime (180 h at $>100 \text{ cd m}^{-2}$). Uniform emission to the eye was attained from thin and highly flexible textiles featuring a large emission area of 42 cm^2 , without resorting to planarization of the partially wavy-shaped (valley-to-peak height = $2.7 \mu\text{m}$) fabric electrode. The key enabling factors for the functional emissive textile are the characteristic *in situ* electrochemical doping of LEC devices, the 'dry' spray-sintering deposition of the active material, and the attractive mechanical, electronic and optical properties of the fabric-based electrode.

1. Introduction

The burgeoning field of electronic textiles (e-textiles) is about incorporating electronic functionality into fabrics [1], with examples on achieved e-textile functions including sensing [2], power generation [3], and communication [4]. The attractive form factor of textiles, notably their characteristic high flexibility and low weight, distinguishes these e-textiles from other electronic device architectures, and it is anticipated that it will pave the way for a plethora of highly desired wearable applications. This is also a key reason as to why the market for e-textiles is projected to reach \$3 billion in 2026 [5, 6].

With the advent of the organic light-emitting diode (OLED), it became possible to obtain low-voltage and glare-free light emission from flexible and surface-emitting device structures [7–9]. This newfound design freedom does however carry a cost as OLED fabrication involves expensive vacuum technology and relies on the fault-sensitive deposition of thin layers with only nanometer-sized tolerances in thickness [10]. The light-emitting electrochemical cell (LEC) is

an appealing alternative that shares many of the desirable features of the OLED, such as low turn-on voltage and glare-free areal emission [11–17]. However, by virtue of its distinctive *in situ* electrochemical doping operation [18–20], the LEC can be fabricated from solely air-stable and solution-processable materials and tolerate uneven active materials, making it a prime candidate for the realization of low-cost light sources [21–28].

Both LECs and OLEDs are planar devices that rely on a transparent electrode in order to let the generated light escape the device, and the material in vogue as of today is indium-tin oxide (ITO). The drawbacks with ITO are that it is brittle and tends to crack when flexed or bent; and that the scarce indium can be a significant cost driver if large amounts of devices are to be fabricated. Accordingly, there is an ongoing intense activity aimed at evaluating and developing alternative transparent electrodes, which are based on abundant and environmentally sound materials and which are highly flexible [7, 8, 29–38].

It is in this context that we present a highly flexible and light-weight emissive LEC textile device fabricated

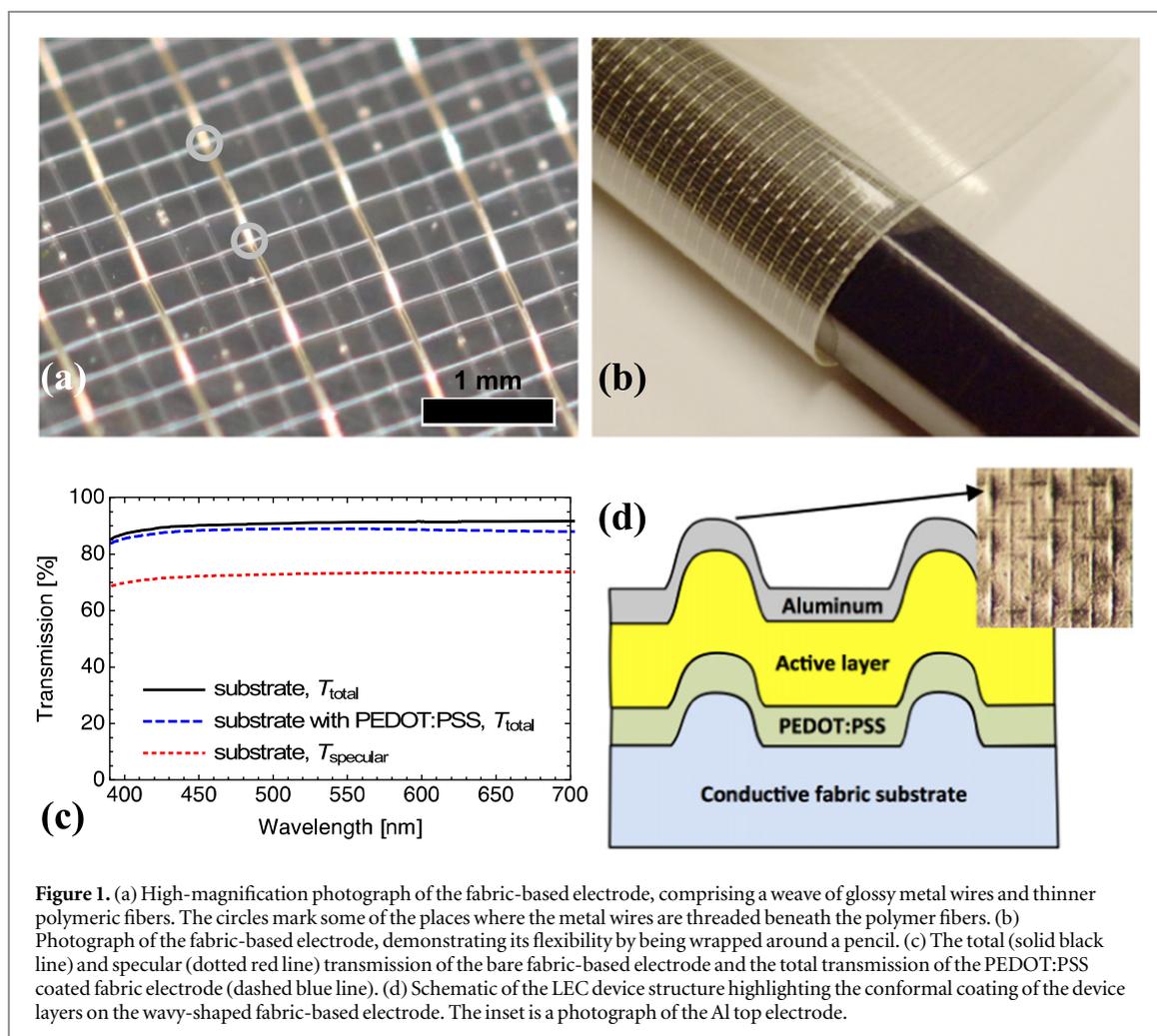


Figure 1. (a) High-magnification photograph of the fabric-based electrode, comprising a weave of glossy metal wires and thinner polymeric fibers. The circles mark some of the places where the metal wires are threaded beneath the polymer fibers. (b) Photograph of the fabric-based electrode, demonstrating its flexibility by being wrapped around a pencil. (c) The total (solid black line) and specular (dotted red line) transmission of the bare fabric-based electrode and the total transmission of the PEDOT:PSS coated fabric electrode (dashed blue line). (d) Schematic of the LEC device structure highlighting the conformal coating of the device layers on the wavy-shaped fabric-based electrode. The inset is a photograph of the Al top electrode.

on a transparent and plastic-supported fabric-based electrode. The embedding of the fabric in a polymer matrix makes it possible to fabricate an area emitter with a high light-emission fill factor. Both, the top-anode and the active-material layers were deposited by conformal solution-based spray-coating under ambient air, and the device delivers bright and efficient electroluminescence at low voltage. Moreover, we demonstrate large-area emissive textile-based sheets that feature uniform light-emission under severe bending and flexing.

2. Experimental details

A close-up photograph of the fabric-based electrode (produced by Sefar AG, Switzerland) is shown in figure 1(a). It displays a weave of polymeric poly(ethylene naphthalene) (PEN) fibers (diameter = 30 μm) and Ag-coated Cu wires (diameter = 40 μm), with the glossy metal wires being oriented in only one direction. The fiber-fiber distance is 280 μm , and the distance between two neighboring metal wires is 850 μm . This mesh is supported on a poly(ethylene terephthalate) barrier foil, and embedded in a polyurethane (PU) matrix.

The thickness of the PU filling matrix is selected so that the metallic wires protrude out of the matrix, forming metallic islands through which the wires can contact upper layers or be contacted by external probes. The height of the metal-protrusion features is 2.7 μm . Height-profile scans over several millimeters that include the metal protrusions yield an average surface roughness of the fabric electrode of 615 nm. The local surface roughness (scan length = 1 μm) between protrusions, however, is approximately 1 nm, as measured by a stylus profilometer (Dektak XT, Bruker). The fabrication of the fabric-based electrode has been described in detail in earlier studies [38, 39].

Figure 1(b) is a photograph of the fabric electrode wrapped around a pencil, demonstrating its excellent flexibility and transparency. The latter was quantified by UV-vis spectroscopy (Lambda 35, PerkinElmer, equipped with an integrating sphere), and figure 1(c) demonstrates that the average total transmission of the fabric electrode is 91% in the wavelength range of 400 to 700 nm, while the average specular transmission is 73%. Accordingly, 18% of the transmitted light results from scattering. Importantly, the transmission coefficient is effectively constant over the entire visible

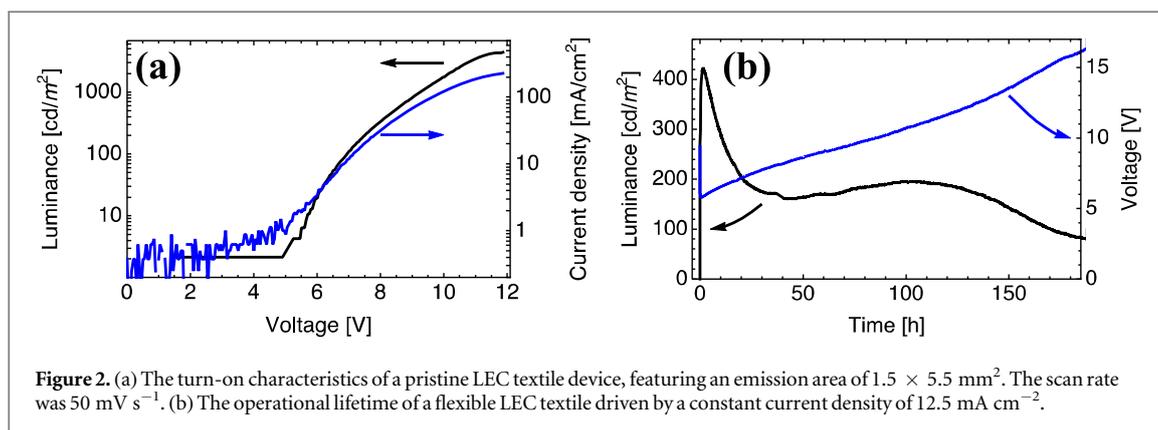


Figure 2. (a) The turn-on characteristics of a pristine LEC textile device, featuring an emission area of $1.5 \times 5.5 \text{ mm}^2$. The scan rate was 50 mV s^{-1} . (b) The operational lifetime of a flexible LEC textile driven by a constant current density of 12.5 mA cm^{-2} .

range, implying that the fabric electrode is fit for broadband white-light applications.

In order to connect the discrete metal protrusions and form a continuous conducting anode surface, a thin layer of poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) was coated onto the fabric electrode. The PEDOT:PSS ink was prepared by diluting 1 ml commercial PEDOT:PSS dispersion (Heraeus Clevis S V3) with 20 ml of ethanol. The ink was shaken vigorously until it appeared homogeneous, sonicated for 30 min, and spray coated onto the fabric electrode, using an in-house developed computer-controlled x - y stage spray coater equipped with a two-fluid nozzle (LI, LunaLEC AB, Sweden) [22]. The atomizing gas was N_2 at 50 psi, and the nozzle was positioned at 10 cm height above the fabric electrode, which was maintained at $50 \text{ }^\circ\text{C}$ by a hot plate. The ink deposition rate was 4.75 ml min^{-1} , and the time to coat a $10 \times 10 \text{ cm}^2$ device was 40 s. The coated PEDOT:PSS film was vacuum dried at $120 \text{ }^\circ\text{C}$ for 12 h before further treatment, and its dry thickness was 50 nm.

The sheet resistance (R_s) of the PEDOT:PSS coated fabric electrode was measured with the 4-point probe technique (Jandel). In the direction parallel to the metal wires we find that $R_s = 348 \text{ } \Omega/\square$, and perpendicular to the metal wires we obtain $R_s = 384 \text{ } \Omega/\square$. We were not able to measure R_s for the non-coated fabric electrode, presumably since we were not able to perfectly align the probe tips in the direction of the metallic wires. We note, however, that the authors of [38] were able to measure R_s of a similar fabric electrode using a modified 4-point probe. The PEDOT:PSS coated fabric electrode displays an essentially intact high transmission of 88% over the entire visible wavelength range, as displayed in figure 1(c).

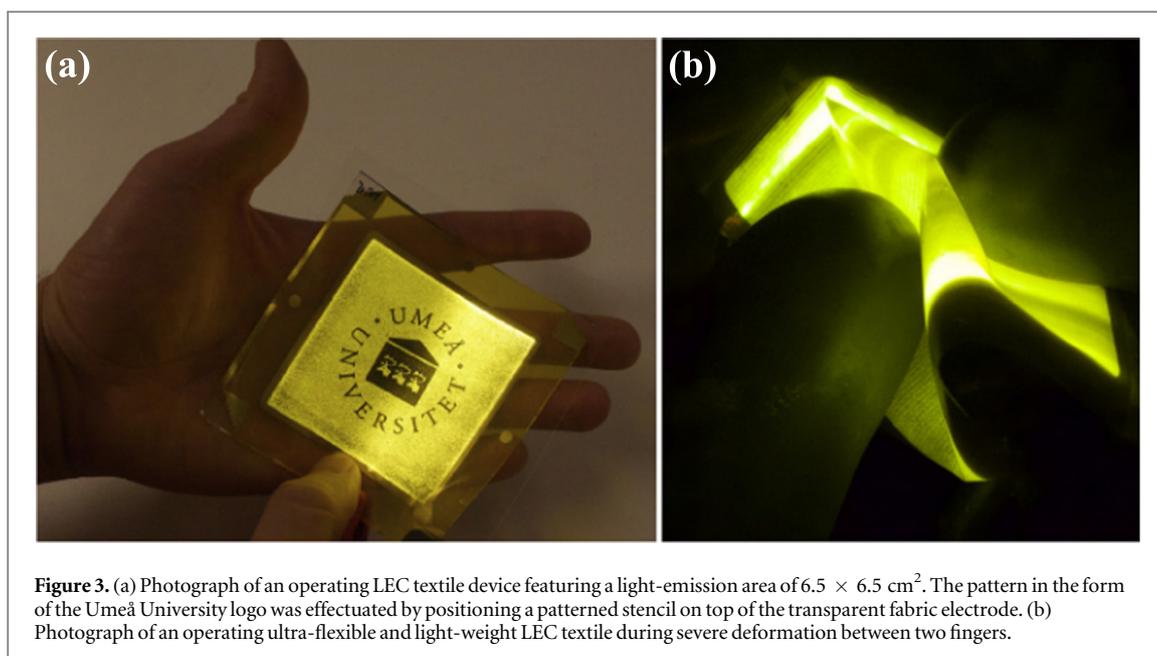
The active layer of the LEC was also deposited by spray-coating under ambient air. To this end, the commercially available conjugated polymer decyloxyphenyl substituted poly(1,4-phenylene vinylene) (Super Yellow, Merck catalog number PDY-132), the ionic solvent hydroxyl-terminated trimethylolpropane ethoxylate (TMPE-OH, Aldrich, $M_w = 450 \text{ g mol}^{-1}$), and the salt KCF_3SO_3 (Aldrich) were each dissolved in

anhydrous cyclohexanone at a concentration of 8 mg ml^{-1} , 25 mg ml^{-1} , and 10 mg ml^{-1} , respectively. These master solutions were mixed together in a solute mass ratio of Super Yellow:TMPE-OH: $\text{KCF}_3\text{SO}_3 = 1:0.15:0.03$ to form the ink used for the active layer in the LECs. Deposition parameters were adjusted to obtain an active layer thickness of 300 nm. A 100 nm thick aluminum cathode was deposited by thermal evaporation onto the active material. A schematic of the complete device stack is shown in figure 1(d).

A visual inspection of the complete device reveals that the characteristic weave pattern of the fabric can be observed at the top surface of the device (i.e. on the surface of the Al cathode), as shown in the photograph in the inset in figure 1(d). The average height of the ‘top-surface protrusions’ is $2.2 \text{ } \mu\text{m}$, and the mean surface roughness of the Al cathode is 660 nm, which is very similar to the values recorded on the bare fabric electrode. Based on these measurements and the visual inspection we conclude that the deposition of the three constituent layers of the LEC on the structured fabric-based electrode is highly conformal, as indicated in figure 1(d).

3. Results and discussion

The device performance of LEC-textile devices with an emission area of $1.5 \times 5.5 \text{ mm}^2$ (as defined by the size of the top cathode) was measured in a N_2 -filled glove box ($[\text{O}_2] < 1 \text{ ppm}$, $[\text{H}_2\text{O}] < 1 \text{ ppm}$), using a computer-controlled source-measure unit and a calibrated photodiode with an eye-response filter (Hamamatsu Photonics). For each presented experiment, we have characterized four different pristine devices, and the presented data are that of a champion device within this group. Figure 2(a) presents the optoelectronic response of the LEC textile when driven by a voltage ramp of 50 mV s^{-1} . The pristine device turns on and begins to emit light at 5 V, and at 12 V the luminance reaches 4370 cd m^{-2} . We tentatively attribute the fact that the device turns on above the band gap of Super Yellow ($E_g = 2.5 \text{ eV}$) to electrochemical side reactions stemming from the inclusion of the PEDOT:PSS



current-spreading layer. Quite interestingly, during this experiment the current conversion efficacy is observed to increase with increasing luminance to reach 1.9 cd A^{-1} at the peak luminance. This behavior is a reflection of an ongoing electrochemical doping process within the active material during the time course of the experiment.

The kinetics and operational stability of the LEC textile were measured at a constant current density of 12.5 mA cm^{-2} . The turn-on time of a pristine device, here defined as the time to reach 100 cd m^{-2} , was found to be a mere 6 s. The long-term behavior of such a galvanostatically driven textile is depicted in figure 2(b), and we observe a peak luminance of 424 cd m^{-2} at 1.5 h. At this time the device displays a current conversion efficacy of 3.4 cd A^{-1} . During this experiment, we consistently observe a second peak in the luminance at around 100 h, and we suspect that this can be attributed to a long-term temporal evolution of the internal doping profile, as discussed by van Reenen and co-workers in a recent publication [18]. Importantly, the operational stability of the LEC textile is good, as evidenced by that we could operate the device in figure 2(b) for more than 180 h at a luminance exceeding 100 cd m^{-2} . It is also plausible that a further improvement in device performance could be attained with well-established procedures such as optimization of the electrolyte concentration [40] and employment of pulsed driving [41].

Inspired by the good performance of the small-area LEC textile devices, we opted to fabricate significantly larger sized devices with the same method. Figure 3(a) presents a photograph of an LEC textile featuring a large emission area of 42 cm^2 , and with a patterned stencil positioned on top of the transparent fabric electrode. The latter resulted in the patterned

emission, but we call particular attention to the observed uniformity of the emission to the eye over the entire area. We attribute this achievement to the high conductivity of the fabric electrode, the current-spreading capability of the intermediate PEDOT-PSS layer, and the elimination of deposition defects by the spray-sintering technique. The latter property is discussed in detail in [22]. We mention that the non-encapsulated large-area devices were operating under ambient air, and that no signs of significant degradation were observed during the optical interrogation period of approximately 30 min. Nevertheless, it is in general recommended to protect LEC devices from ambient oxygen and water during operation, and information on appropriate flexible encapsulation structures can be found in the scientific literature [42, 43]. In order to scale-up our device design to even larger emission areas we are currently evaluating alternative and more appropriate methods for the fabrication of the top contact, such as lamination [23] or spray-coating [22].

However, the most attractive feature of the LEC textile device is most plausibly its unique form factor. The photograph in figure 3(b) displays a flexible, large-area LEC being severely pinched between two fingers during light emission, and as indicated by the photograph the entire device remains emissive despite this harsh treatment. In fact, the device could be repeatedly pinched and released without sustaining folding marks or defects. The light-emission textile device also features some stretchability, and at 5% elongation it remains functional. A further advantage of the LEC textile device is its light weight, and while a standard LEC device fabricated on a 1 mm thick glass substrate carries a weight of 265 mg cm^{-2} [44], the flexible LEC weighs in at only 14 mg cm^{-2} . With a

proper encapsulation, we envisage potential for our device architecture in flexible electronics applications like architecture, signage, backlighting, and wearables.

4. Conclusions

To summarize, we report on the realization of light-weight and ultra-flexible emissive textile devices displaying low turn-on voltage for light-emission, high maximum brightness, and good efficiency and stability. We also show that such textile devices can deliver uniform and fault-tolerant light emission from large emissive surfaces, and attribute these achievements to the high-performance fabric-based electrode, the electrochemical doping of the LEC device, and the fault-tolerant spray-sintering of the active material.

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