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**PAPER**

# Screen printing and *in situ* electropolymerization as a simple manufacturing method to produce long cycling lifetime electrochromic indicators

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**Keywords:** *In situ* electropolymerization, 3,4-ethylenedioxythiophene, printed electronics, printed electrolyte, electrochromic display, smart label

Supplementary material for this article is available online

## Abstract

Successful commercialization of electrochromic technologies into smart labels necessitates that they can be produced in high-volumes and at a low cost. Herein we present a simple method to produce poly(3,4-ethylenedioxythiophene) electrochromic indicators via screen printing and charge controlled *in situ* electropolymerization. This method reduces the number of printed layers, thus lowering the overall time and cost of production. The optical contrast and visual homogeneity of the indicators can be tuned by modifying the electropolymerization voltage and limiting the charge. Due to the opaque nature of the printed electrolyte, the switching speed can be increased, and the operating voltage can be lowered by electropolymerizing an electrochromic film onto the counter electrode to act as a charge storage material. The best performing indicators exhibit a contrast of 31.1%R (600 nm), with a 1 s switching speed. The CIELAB color co-ordinates of the indicators were measured over 100 k cycles. During this period, the indicators showed an overall increase in contrast. The long cycling lifetime of the indicators makes them promising candidates for applications where frequent switching is required.

## 1. Introduction

Electronic smart labels use embedded technologies to capture and communicate real-time information about items. These technologies can include sensors, indicators, displays, and data transmitters. Key markets for smart label technologies are logistics, healthcare [1], anti-counterfeiting [2], and food packaging [3] where real-time access to information and data is critical. Electrochromic indicators and displays have emerged as a promising candidate to serve as visual interfaces for smart label technologies due to their low-power consumption, and flexible form factor [4–10]. However, broad commercialization of electrochromic devices (ECDs) in smart labels is limited by their high cost compared to non-visual data transmitters such as radio-frequency identification, and near-field communication [11]. Thus, further developments are necessary to lower the manufacturing cost of electrochromic indicators without compromising key operational parameters such as contrast, switching speed, and cycling lifetime.

To this end, *in situ* electropolymerization has been proposed as a simple method to produce electrochromic displays. *In situ* electropolymerization refers to the process of electrochemically depositing a polymer inside a solid state device [12, 13]. The concept was introduced by the group of Sotzing, and they have reported using a range of different monomer precursors to form the active electrochromic layers in both glass and polyethylene terephthalate (PET) based devices [12, 14–18]. They achieve a maximum photopic contrast of 50% for glass-indium tin oxide (ITO) devices with 2,2-Dimethyl-3,4-propylenedioxythiophene as the monomer precursor [16]. However, a key limitation for *in situ* polymerized ECDs is their low cycling lifetime due to the continued deposition of unreacted monomer on the electrodes. In one report, this was improved by modifying a 3,4-propylenedioxythiophene monomer with an acrylate group [14]. The unreacted monomers were UV-crosslinked to the gel electrolyte after the electrochromic film was formed to prevent ongoing electrodeposition on the counter electrode during use. This yielded a lifetime of 10 k cycles with a loss of photopic contrast of only 3%.

Another approach to lower the manufacturing cost of ECDs is implementing high-volume industrial manufacturing processes, such as screen printing [5, 9, 10]. To the best of our knowledge, there are no reports of using screen printing in conjunction with *in situ* electropolymerization. Combining these two techniques would be beneficial since it can reduce the number of layers that need to be printed during the device assembly process directly cutting down the production time. Figure 1(a) outlines the manufacturing steps for an ECD using only printing. In this example, there are seven printed layers: silver conductors on both electrodes, an electrolyte layer, a spacer or dielectric separator, an electrochromic layer, and a charge storage layer (in this case, a second electrochromic layer). Figure 1(b) outlines the manufacturing steps for the same ECD using both printing and *in situ* electropolymerization. In this case, two fewer layers need to be printed.

In this work, we employ screen printing in conjunction with *in situ* electropolymerization to optimize the manufacturing of electrochromic indicators. 3,4-ethylenedioxythiophene (EDOT) is used as the precursor monomer to form electrochromic poly(3,4-ethylenedioxythiophene) (PEDOT) films. The thickness of the films is controlled by limiting the charge during the reaction. We use a diffusely reflective, opaque electrolyte ink, which is a variation of the electrolyte ink previously reported by Leite, Maggini *et al* [19, 20]. One of the key advantages of using an opaque electrolyte, is that an additional electrochromic layer can be deposited on the counter electrode of the device to act as a charge storage material without compromising the overall device contrast (as would be the case with a transparent electrolyte). In this study, we manufacture several variations of electrochromic indicator with different thicknesses of electrochromic film, with and without a charge storage layer. Then, the performance of the indicators is evaluated based on their optical contrast, switching speed, and cycling lifetime.

## 2. Materials and methods

### 2.1. Electrolyte composition and processing

The electrolyte used is a modified version of what was reported in [18]. The printable electrolyte (pQSPEv2) was formulated by adding 47.08 wt% titanium dioxide ($TiO_2$;∼200 nm, rutile pigment with 2.30% $Al_2O_3$ surface treatment), to a mixing solution of propylene carbonate (PC):ethylene carbonate (EC):lithium perchlorate ($LiClO_4$) (0.57:0.26:0.18) with 2.02 wt% of a hyperbranched polyester nanoparticle stabilization agent. The slurry is mixed at high shear rate (1000 rpm) using an overhead stirrer with a dispersing blade for 10 min, after which 11.98 wt.% glycerol propoxylate-(1PO/OH)-triacrylate (GPTA) was added. After 10 min of mixing, 18.26 wt.% polyvinylidene fluoride (PVDF, MW ∼ 534 000 by gel permeation chromatography) was slowly added to the slurry while continuing stirring the formulation at 1000 rpm. High shear rate mixing was maintained for 24 h. Finally, 0.50 wt% Irgacure 819 photoinitiator and 2.00 wt% EDOT monomer were added and mixed for 2 h.

### 2.2. Indicator printing and assembly

The indicators were printed on two separate ITO coated PET substrates and then laminated. The silver, dielectric, and adhesive layers of the indicator were prepared in an industrial setting (Invisible S.A.) according to Invisible industrial standards and protocols using a THIEME LAB1000 sheet to sheet screen printer. The first substrate was prepared by printing silver paths followed by a layer of adhesive. The second substrate was prepared by printing silver paths, followed by a layer of dielectric ink.

The electrolyte layer was added to the second substrate using a benchtop A4 screen printer with a 100/40 screen. Then, the two substrates were laminated using a weighted roller, and the electrolyte was UV-cured (LOCTITE® UVALOC 97 035 with a 1000 W Mercury doped lamp, 1600 mJ $s^{-1}$). The individual indicators were separated from the sheets using a cutting machine. An image of the screen printing environment, the electrolyte ink after printing, and the configuration of the stack are shown in figure S1 of the supplementary information.

### 2.3. Electrodeposition of 3,4-ethylenedioxythiophene

*In situ* electropolymerization was performed using a Metrohm Autolab PGSTAT100 potentiostat by controlling the charge consumed while monitoring film formation on the working electrode at 600 nm on a Varian Cary 300 UV–Vis spectrophotometer. Each indicator was preconditioned by cycling with ±1.0 V in 1 s pulses for 8 min before any further tests.

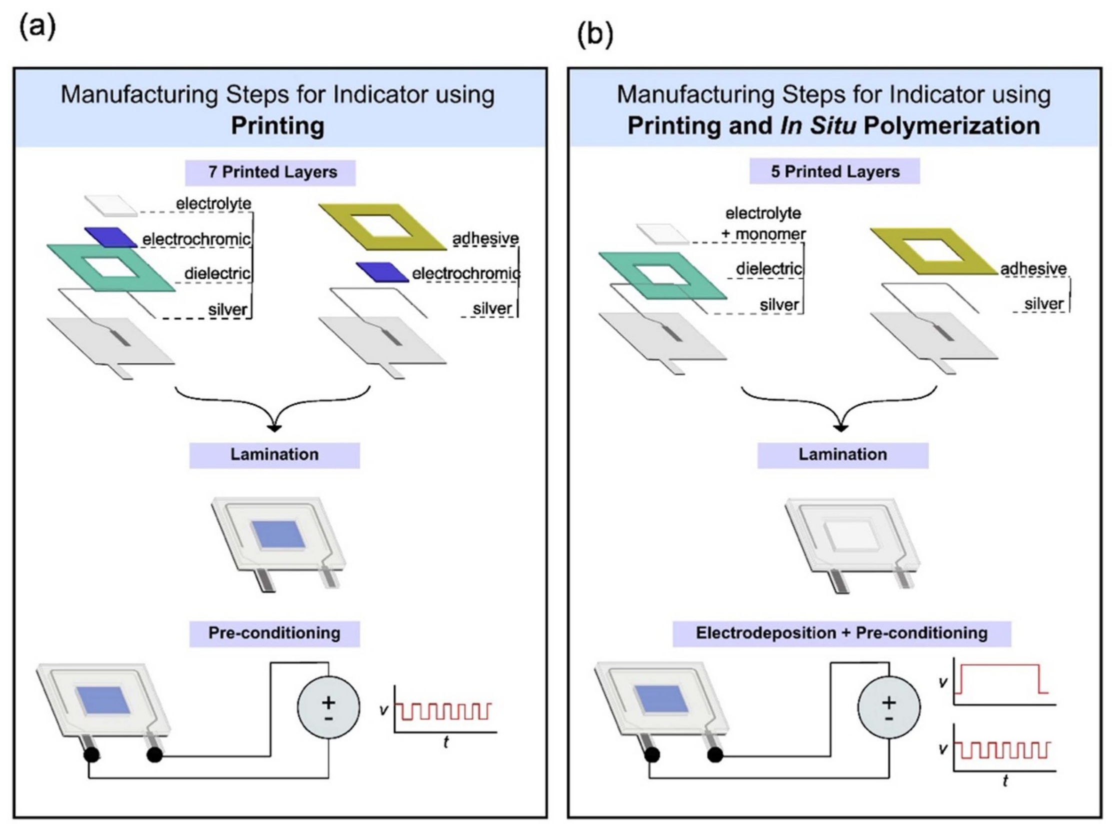

**Figure 1.** Comparison of manufacturing steps for producing an electrochromic indicator using (a) printing, and (b) printing and *in situ* electropolymerization.

### 2.4. Switching time analysis

The switching time of the indicators was measured using an adaptation of the electrical pulse method proposed by Hassab *et al* [21]. The switching time was evaluated at 1.5 V, and 1.2 V using and electrical protocol containing successive cathodic and anodic pulse with increasing time periods. Pulse lengths of 0.5 s, 1 s, 2 s, and 3 s were repeated 10 times. Pulse lengths of 5 s and 10 s were repeated five times. The final pulse length of 20 s was repeated twice. See supplementary figure S2 for a visual representation of the protocol.

### 2.5. Cycling analysis

Indicator cycling lifetimes were evaluated by monitoring the color states of the indicators over 100 k cycles. Cycling was performed with both ±1.2 V and ±1.5 V (1 s pulse on each electrode). Digital images were captured inside a custom-made cycling chamber with an IDS Imaging UI-3590CP-C-HQ Camera and then color corrected using an x-rite color checker in the frame of each image to output (see supplementary figure S3). The colors were converted from RGB to the CIE $L^*a^*b^*$ color space (D65 illuminant) and then the contrast $\Delta E$ was calculated according to equation (1).

$$\Delta E = \left[\left(\left(l^*{}_{(\max)} - l^*{}_{(\min)}\right)\right)^2 + \left(\left(a^*{}_{(\max)} - a^*{}_{(\min)}\right)\right)^2 + \left(\left(b^*{}_{(\max)} - b^*{}_{(\min)}\right)\right)^2\right]^{(1/2)}. \quad (1)$$

## 3. Results and discussion

### 3.1. *In situ* electropolymerization

The term ‘activation’ is used here to describe the *in situ* electropolymerization process used to form the electrochromic layers. Two variations of indicator were manufactured for this study: asymmetrical indicators, and symmetrical indicators. For the asymmetrical indicators, a PEDOT layer is only formed on the working electrode. For the symmetrical indicators, an additional electropolymerization step with a reversed polarity was added so that PEDOT is formed on both the working electrode and the counter electrode. The difference between the two variations of indicator is highlighted in figure 2(a).

It was more challenging to obtain a homogenous polymer film on the counter electrode than on the working electrode. We tested multiple voltages and found that the most uniform counter electrode films were formed using—4.0 V (see supplementary figure S4). Thus, the indicators are activated with

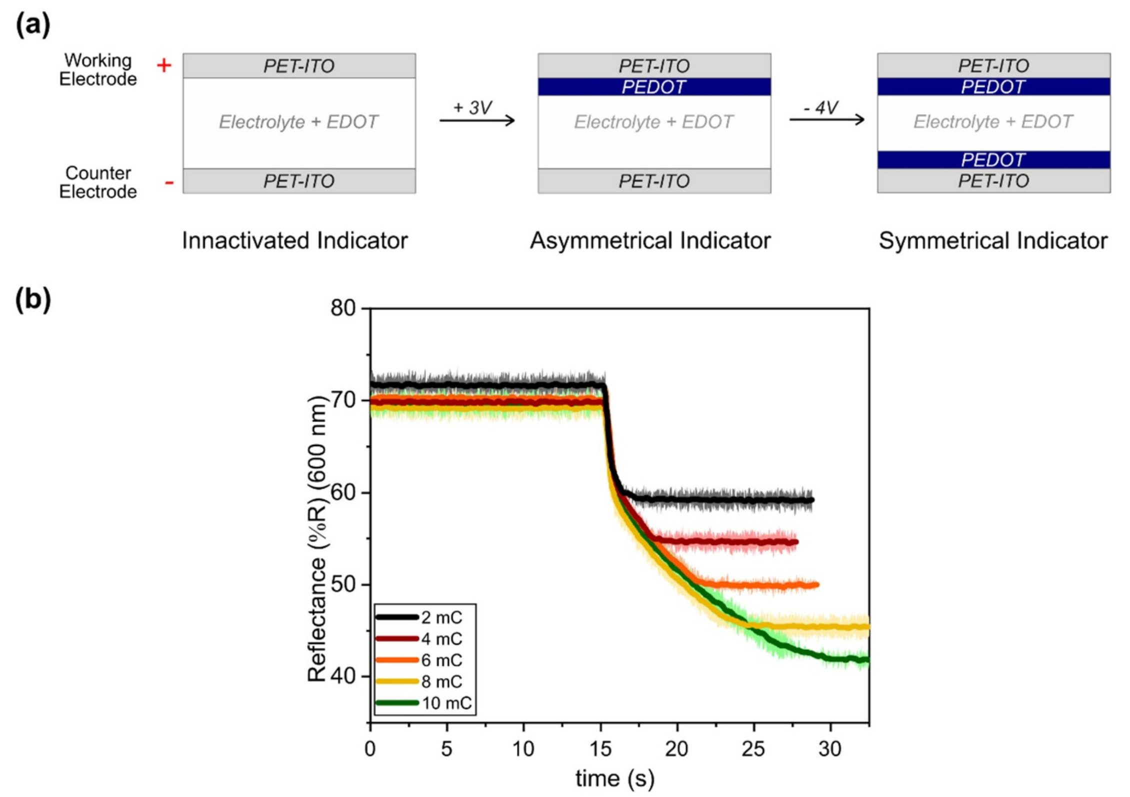

**Figure 2.** (a) Schematic illustrating the difference between the symmetrical and asymmetrical indicators, and (b) reflectance of working electrode of asymmetrical indicators during activation with 3 V. Data is shown for indicators activated until they reach 2, 4, 6, 8, and 10 mC.

3.0 V on the working electrode, and then—4.0 V on the counter electrode (for symmetrical indicators). The indicators are always viewed and evaluated from the side of the working electrode.

The change in reflectance for asymmetrical indicators during activation is shown in figure 2(b). As the charge applied increases, a lower reflectance is reached, indicating that more PEDOT was deposited. The time to form the film ranges from less than 2 s for the 2 mC film to around 15 s for the 10 mC film.

### 3.2. Contrast and switching time of indicators

After activation, the contrast and switching time of the indicators were measured using the protocol described in the materials and methods section. Figure 3(a) shows the contrast of the asymmetrical indicators. The results are an average of three indicators, and the voltage used for the switching time protocol was ±1.5 V. As the amount of charge used during the activation increases, there is a decrease in both the minimum and maximum reflectance values. The highest contrast is achieved in indicators formed with 6 mC of charge. Figure 3(b) shows the contrast of the symmetrical indicators. They show a similar trend to the asymmetrical indicators with a maximum contrast occurring at 6 mC. However, at higher activation charges (8 and 10 mC) some yellowing/browning is observed on the working electrode after activation. This suggests there is a limit to how much PEDOT we can form on the counter electrode without damaging the ITO\PEDOT on the working electrode.

The switching speed for the 6 mC symmetrical and 6 mC asymmetrical indicators are shown in figure 4 for driving voltages of ± 1.2 V and ± 1.5 V. Three key differences in their switching time and contrast are observed:

- The symmetrical indicators have a lower overall contrast (34.7 Δ%Reflectance for symmetrical indicators for 20 s at ± 1.5 V, compared to 36.4 Δ%R for asymmetrical indicators for 20 s at ± 1.5 V). While not visible to the human eye, the electrolyte is not completely opaque (see supplementary figure S5). Thus, for the symmetrical indicators, there is a slight contribution of the counter electrode PEDOT film to the color of the indicator as observed from the working electrode. To increase the electrolyte reflectiveness, more $TiO_2$ could be added to the formulation, but this would change the rheology and reduce the printability.
- The symmetrical indicators exhibit faster switching at both ±1.2 and ±1.5 V. With a 0.5 s pulse length, the symmetrical indicators have a 13.2%

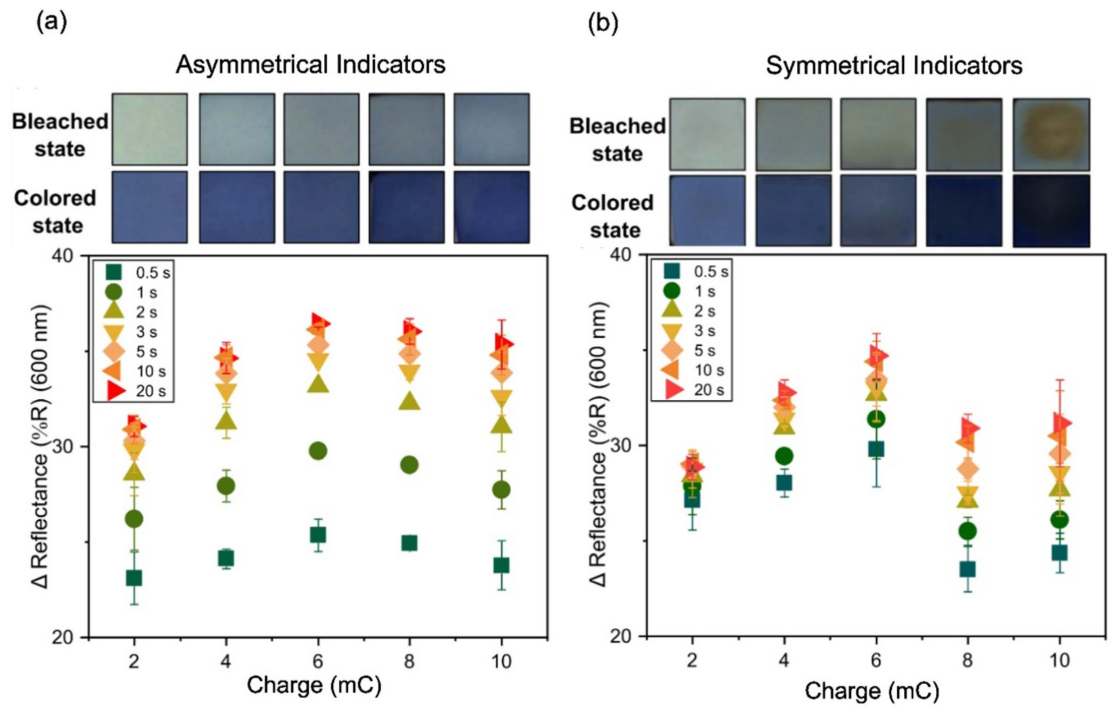

**Figure 3.** (a) Contrast, as measured in Δ reflectance at 600 nm, for asymmetrical indicators activated with 2, 4, 6, 8 and 10 mC. Color corrected digital images of their bleached and colored states are above the plot. (b) Contrast, as measured in Δ reflectance at 600 nm, for symmetrical indicators activated with 2, 4, 6, 8 and 10 mC. Color corrected digital images of their bleached and colored states are above the plot. Indicators switched with ±1.5 V and data is shown for 0.5, 1, 2, 3, 5, 10, and 20 s pulses.

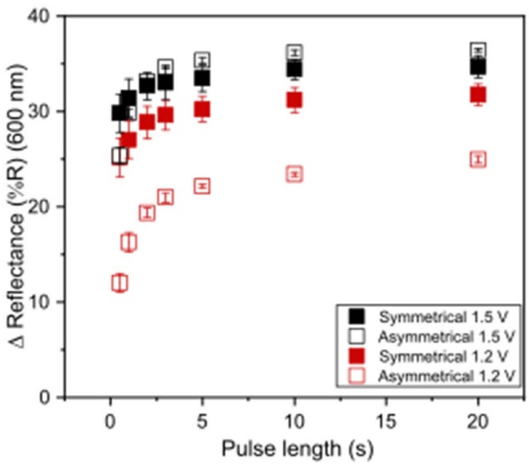

**Figure 4.** Δ reflectance at different pulse lengths for 6 mC asymmetrical and symmetrical indicators switched with ± 1.2 V and ± 1.5 V.

higher contrast at ±1.2 V, and 4.4% higher contrast at ±1.5 V.

- At ±1.2 V, the symmetrical indicators show significantly higher contrast across all pulse lengths.

The difference in the switching times and contrast between the symmetrical and asymmetrical indicators originates from the nature of the charge storage material used. The charge storage material is critical to determining the overall performance of an ECD [22, 23]. As a redox reaction takes place to change the oxidation state of the electrochromic film on the working electrode, a simultaneous complementary redox reaction must take place on the counter electrode.

For the symmetrical indicators a second PEDOT films acts as the charge storage material, whereas for the asymmetrical indicators ITO acts as the charge storage material. Electropolymerized PEDOT films can exhibit coloration efficiencies up to 180–190 cm$^2$ °C$^{-1}$ [24, 25], which gives rise to redox capacities in the range of 1–10 mC cm$^{-2}$ depending on the film thickness and contrast. In studies of ITO in PC solutions, Bressers and Meulenkamp report an initial charge density for ITO films during cycling of 4 mC cm$^{-2}$, however this rapidly decreases over 2 k cycles to around 1 mC cm$^{-2}$ [26].

Thus, for the asymmetrical indicators the device is likely to have an unbalanced configuration, where the overall charge passing through the device will be limited by the redox capacity of the ITO. Unbalanced configurations where there is excess charge capacity on the electrochromic layer can result in higher switching voltages and can induce additional reactions at the electrode or in the electrolyte [23, 27]. In ITO, this is often characterized by yellowing or browning and a loss in conductivity due to irreversible reduction [6, 26–29]. So, while ITO may be a suitable charge storage material for proof-of-concept devices, or when a long cycling lifetime is not required, it is not ideal for applications where long-term stability and performance are criterion.

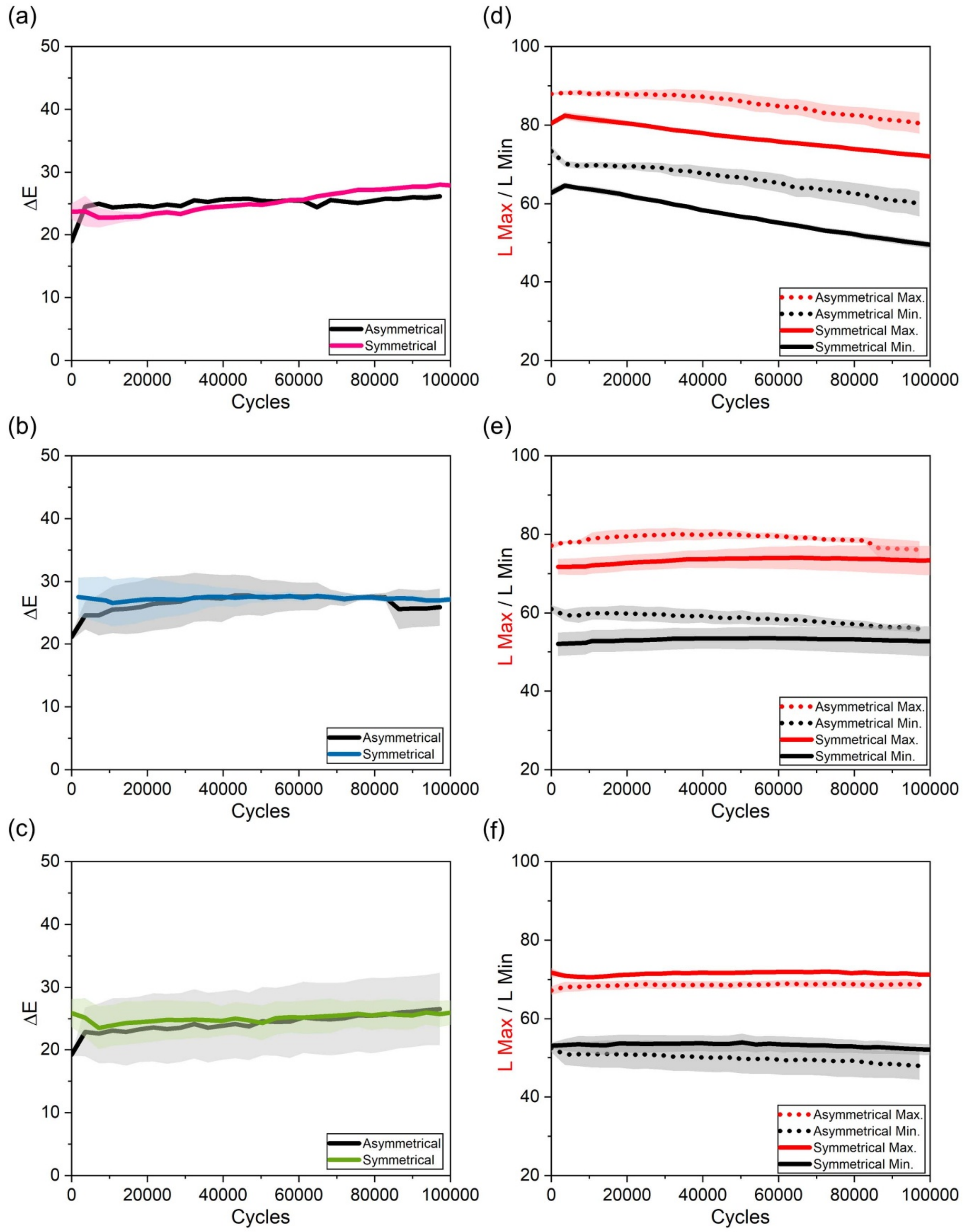

**Figure 5.** ΔE for triplicates of asymmetrical and symmetrical indicators activated with (a) 2 mC, (b) 4 mC, and (c) 6 mC over 100 k cycles of ± 1.2 V applied in 1 s pulses. Minimum and maximum L* values for the same triplicates of asymmetrical and symmetrical indicators activated with (d) 2 mC, (e) 4 mC and (f) 6 mC over 100 k cycles of ± 1.2 V applied in 1 s pulses.

### 3.3. Lifetime cycling of indicators

The previous sections in this study have focused on the activation and then the performance of the indicators after only a brief pre-cycling protocol. However, commercial applications can require that indicators undergo tens of thousands to hundreds of thousands of cycles. Thus, it is critical to understand how the performance of the indicators evolve during use.

Symmetrical and asymmetrical indicators activated with 2, 4 and 6 mC were cycled for 100 k. The driving protocol consists of ± 1.2 V pulses with 1 s at each polarity. The contrast (measured in $\Delta E$), the minimum $L^*$ value, and the maximum $L^*$ value during this cycling period is shown in figure 5. Almost all the indicators exhibit an overall increase contrast over 100 k cycles. For example, the 2 mC symmetrical and 2 mC asymmetrical indicators have an increase in contrast of 5.2 and 6.5 $\Delta E$ respectively. The increase in contrast is attributed to continued electrodeposition of PEDOT on the working electrode during the

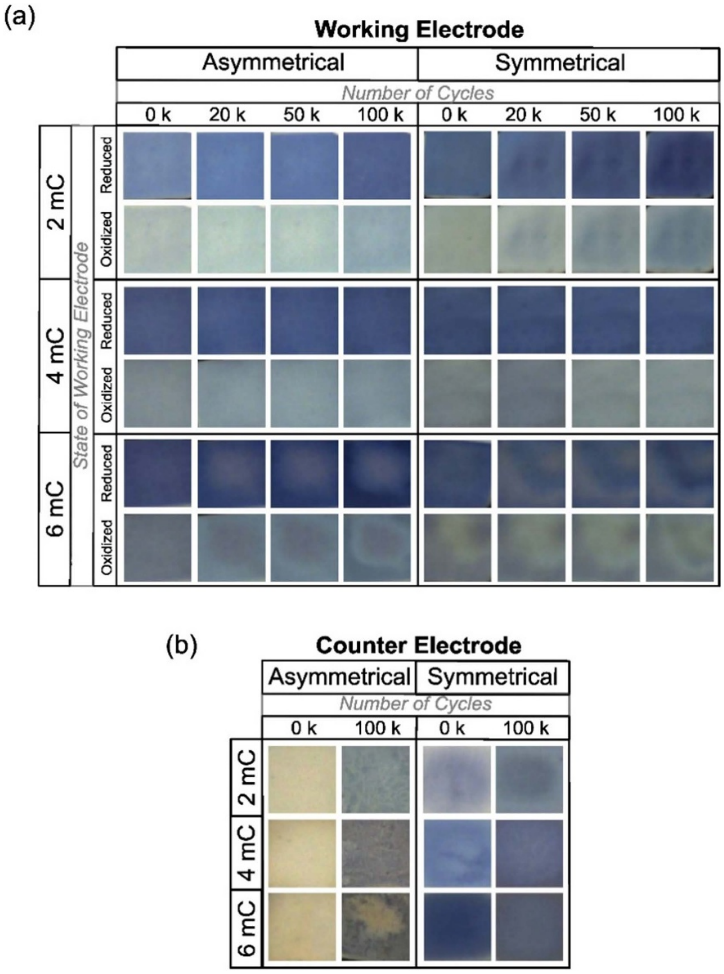

**Figure 6.** Digital images of the (a) working electrode and the (b) counter electrode sides of the indicators over 100 k cycles. Cycling performed at ± 1.2 V in 1 s pulses.

cycling process. This can be observed via the decrease in the minimum and maximum $L^*$ values (which indicates a darkening of both the colored and the bleached states over time) and in the color corrected digital images of the indicators shown in figure 6(a).

The extent to which the color states drift over time is dependent in the initial thickness of the PEDOT film. For example, the 2 mC symmetrical and asymmetrical indicators show a decrease in their maximum $L^*$ values of 9.01 and 8.20 respectively, whereas the 6 mC symmetrical and asymmetrical indicators only decrease by 0.38 and 4.67 respectively. Another key difference between the indicators based on their initial film thickness is the reproducibility between the triplicates. For the 2 mC indicators, the triplicates have a standard deviation of 0.52 $\Delta E$ for the symmetrical variation, and 0.31 $\Delta E$ for the asymmetrical variation. These values are significantly higher for the 6 mC indicators, which have a standard deviation of 2.15 $\Delta E$ for the symmetrical variation, and 5.27 $\Delta E$ for the asymmetrical variation. Thus, regarding the starting PEDOT thickness, there is a trade-off between the stability of the color states, and the reproducibility of the indicators during cycling. The 2 mC indicators show good reproducibility but have a visible drift in their color states. On the other hand, the 6 mC indicators have a negligible drift in their color state but have poor reproducibility.

In addition to the ongoing deposition of PEDOT on the working electrode, there is also ongoing deposition of PEDOT on the counter electrode during cycling. This can be seen in the digital images of the counter electrode, in figure 6(b), before and after the 100 k cycling period. The result of this, is that over time the asymmetrical indicators (which begin with only ITO on the counter electrode) turn into symmetrical indicators (which have PEDOT on the counter electrode). This effect is most apparent in the 2 mC asymmetrical indicators where there is an increase in contrast of 5.14 $\Delta E$ between the first time point measured at less than 1 k cycles, to the second time point measured at 3.6 k cycles. Using symmetrical indicators from the start avoids this abrupt change in their performance during the early stages of use.

Cycling results for the same set of indicators over 100 k cycles with a driving protocol of ± 1.5 V with 1 s at each polarity are in the supplementary

**Table 1.** Performance comparison of indicators in this work to prior works.

| | [19] | [17] | This work |
|---|---|---|---|
| Printed device | Yes | No | Yes |
| Deposition of electrochromic layer | Screen printing | *In situ* electropolymerization | *In situ* electropolymerization |
| Switching time (s) | $t_{90} = 5.30$ $t_{75} = 1.70$ | $t_{100} = 2.00$ | $t_{90} = 1.00$ $t_{75} = 0.50$ |
| Contrast (%) | $\Delta R_{Max} = 35$ | Photopic contrast = 45% | $\Delta R_{Max} = 35$ |
| Cycling | 10 000 | 10 000 | 100 000 |
| Voltage | ± 1.5 V | ± 2.0 V | ± 1.2 V |

**Figure 7.** Digital images of 2 mC symmetrical indicators with smart label covers demonstrating use cases for impact, high temperature and freeze warning labels. Indicators are switching with ± 1.2 V in 0.5 s pulses.

information, see figure S6. As expected, with a higher driving voltage all the indicators have a higher $\Delta E$, but the minimum and the maximum $L^*$ values drift more significantly over time as the ongoing polymerization reaction is more severe at ± 1.5 V. These results highlight the trade-offs between contrast, switching speed, and cycling lifetime when selecting an appropriate driving protocol for electrochromic indicators.

Table 1 summarizes the key performance attributes of the devices in this study compared to prior studies of printed and *in situ* polymerized electrochromic indicators. Overall, we observe a 10 times improvement in cycling lifetime and an improvement in switching time using this novel manufacturing technique compared to devices using screen printed PEDOT with a previous iteration of the pQSPEv2 electrolyte [19]. We also observe an improvement from 10 k to 100 k in cycling lifetime in comparison with previous studies of *in situ* electropolymerized devices [17].

### 3.4. Use-case prototype of indicators

Graphical overlays printed by inkjet were designed to highlight envisioned use-case scenarios for these indicators (figure 7). Due to their low-cost manufacturing method, slim profile, fast switching, and long cycling lifetime they are well suited for integration into visual smart labels. In this case, the indicators are used in smart warning labels, where they could flash on and off to alert the user if there is a problem.

## 4. Conclusions

We have successfully reported fully printed electrochromic indicators incorporating an *in situ* electropolymerization technique. To the best of our knowledge, this is the first report on these two techniques being used in conjunction. The proposed method of manufacturing is simple and cost efficient—only requiring five printed layers—and the final properties of the indicator can be optimized by modifying the voltage and charge used to electrodeposit the electrochromic films. We demonstrate the production of two variations of indicator: symmetrical and asymmetrical. The key difference between them is that symmetrical indicators have PEDOT films electropolymerized on both the working electrode and the counter electrode, whereas the asymmetrical indicators only have a PEDOT film electrodeposited on the working electrode. The use of PEDOT as a charge storage material in the symmetrical indicators lowers the operational voltage and increases the switching speed compared to asymmetrical indicators: further highlighting the importance of balanced electrode configurations in ECDs. In this case, is only possible to produce symmetrical indicators due to the diffusely reflective and opaque nature of the printable electrolyte, which blocks the complementary color transitions of the PEDOT film on the counter electrode from the view of the user.

Compared to previous reports of *in situ* electropolymerized devices, we were able to achieve a significantly longer cycling lifetime, up to 100 k cycles. This is again attributed to the opaque nature of the electrolyte. Here continued electrodeposition of the unreacted EDOT monomers during cycling results in an increase in contrast rather than a decrease in contrast as previously reported for *in situ* electropolymerized devices [14]. The long cycling lifetime, and low-cost proposition of the manufactured indicators make them a promising candidate for use in smart label applications. However, a present limitation for this method of manufacturing is that they have only one pixel. To create displays with more segments, one would either need to pattern the conductive substrate into multiple electrically isolated regions that can be electropolymerized separately or use an array with several indicators.

## Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

## Acknowledgment

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## Conflict of interest

The authors declare no conflict of interest.

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