Flexible Active Matrix Addressed Displays Manufactured by Screen Printing

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Screen printed monolithically integrated active matrix addressed displays are demonstrated. An organic electrochemical transistor controls the color of its corresponding electrochromic pixel, thereby preventing cross-talk in neighboring pixels in active matrix addressed displays. The electrolytic interfaces of both devices enable low voltage operation, both devices also rely on the same materials; the latter allows for a simplified screen printing process.
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Abstract
A flexible, electrochromic, active matrix addressed display (AMAD) is demonstrated. The monolithically integrated AMAD, which contains a 3×3 array of organic electrochromic smart pixels (OESP), is manufactured on a plastic substrate solely using screen printing. Each OESP is based on the combination of one organic electrochromic display (OEDC) and one organic electrochemical transistor (OECT), where both devices are screen printed into multilayered vertical architectures. The conduction state of the OECT enables control of the color state of its corresponding OEDC, thereby circumventing cross-talk effects in the resulting AMAD device. The manufacturing approach also involves electrical wires, which connect each OEDC with its corresponding OECT and also serve as the addressing lines of the resulting AMAD device, that are formed by screen printing of an ink based on either silver or nanocopper.
1. Introduction

Printed electronics is a rapidly growing research field, and a large number of research articles describing different kinds of electronic devices and electronic systems, as well as their utilization in a vast array of applications, are currently being published. Printing of electrically conducting materials is not novel in itself; materials such as carbon and silver have been deposited by printing techniques for many years, aiming at, for example, flexible electrical wires, membrane switches and electrodes in sensor applications.[1] The major reason for the large efforts that currently are being spent within the field of printed electronics is related to the combination of such relatively simple printed electrical wires and electrodes with the novel electronic functionalities that often originate in devices based on organic (semi)conducting materials, where one primary goal is to obtain complete electronic systems by only using printing techniques in the manufacturing process. (Semi)conducting polymers are one class of materials that have been thoroughly explored, mainly due to the possibility to control the conduction state of the materials, since this allows device architectures such as diodes and transistors.[2,3] Another focus area lies within electroluminescence, where the ability of exciton formation within the semiconducting polymer enables organic light emitting diodes.[4] Many of the most common electronic devices that are being developed are relying on field-effect operation,[5] which typically results in appealing device performances along with a relatively complex manufacturing approach often involving several vacuum and photolithographic processing steps to achieve the required dimensions of the deposited materials.

Electrochemical switching is yet another type of operation mode that can be obtained in devices based on (semi)conducting polymers. In the case of electrochemical switching, the dielectric layer is simply replaced by an electrolyte capable of providing the ions necessary for the electrochemical reaction. Devices operated through an electrolytic interface show much more relaxed requirements in terms of device dimensions, as compared to field-effect-
driven devices, which in turn allows for utilization of printing techniques in the manufacturing process. In an organic electrochemical transistor (OECT), the electrodes that are bridged by the electrolyte are denoted gate electrode and transistor channel, and the source and drain electrodes are extending from the respective side of the transistor channel. The conduction state of the transistor channel, and hence the current throughput between the source and drain electrodes, is controlled by the amplitude of the gate voltage, which is applied in relation to the source electrode serving as the reference electric potential. Poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) is one of the most well-studied conducting polymers.[6] It is an intrinsically conducting polymer, wherein the positively charged bipolarons along the polymer chains are compensated by the negatively charged PSS chains. PEDOT:PSS is one example of a conducting polymer that strongly responds to electrochemical switching. The oxidized state corresponds to the conducting state, and by adding electrons along the PEDOT chains upon electrochemical reduction, the conductivity drops by several orders of magnitude. This implies the possibility to use the material in OECTs,[7] and the resulting devices exhibit high current levels in their ON state due to conduction throughout the entire bulk of the PEDOT:PSS-based channel. It can also be noted that, owing to the electrolytic property of the PSS that is mixed with PEDOT to form PEDOT:PSS, the material can be utilized in various kinds of bioelectronic applications, for example ion pumps.[8]

In addition to this, PEDOT:PSS also exhibits an electrochromic property. The conducting oxidized state appears transparent to the human eye, or light blue in case of thicker films, while the semiconducting reduced state of PEDOT:PSS appears dark blue. This feature can be utilized in organic electrochromic displays (OECD), either in transmission mode,[9] or in reflection mode.[10] An organic electrochromic smart pixel (OESP) is obtained by combining one OECD with one OECT, in which the conduction state of the transistor enables control of the color state of the display. Arranging several OESPs into a cross-point
matrix results in an active matrix addressed display (AMAD), in which the OECTs and the OECDs typically are controlled row by row and column by column, respectively. Several reports on the topic of PEDOT:PSS-based AMADs have been published previously, however, they were completed by a combination of different manufacturing techniques; subtractive patterning of a uniform pre-coated layer of PEDOT:PSS followed by lamination to complete the devices, resulting in AMADs configured on either a paper substrate or a plastic substrate,[11] or by a combination of printing and coating techniques to implement vertically configured OESPs in which the OECTs and OECDs were located on different sides of a plastic substrate and connected by electronic vias.[12] In addition to this, Cao et al. reported on an electrochromic AMAD manufactured by using screen printing as the deposition method, in which the OECDs were based on PEDOT:PSS while carbon nanotubes served as the active material in the printed organic field-effect transistors (OFET). Their AMAD device was, indeed, all-printed, but the immediate disadvantages of using OFETs are elevated drive voltages (~10 V for OFETs vs. ~1 V for OECTs) and low current throughput (~1 µA for OFETs vs. ~1 mA for OECTs), despite using a channel width as wide as 1000 µm.[13]

Here, we are reporting all-printed electrochromic AMADs manufactured solely by screen printing, wherein both the OECT and the OECD rely on electrochemical switching of an organic material; the electrically conducting polymer PEDOT:PSS. The electrical wires, which connect each OECD with its corresponding OECT and serve as the addressing lines facing an external electronic circuit, were also deposited by screen printing. Different screen printing inks were used to create two different sets of AMADs; one commercially available silver-based ink and one novel ink based on nano-copper. The nano-copper ink was developed by Intrinsiq Materials as one of the objectives in the PLASMAS project funded by the European Commission.[14] The multilayered architectures of the OECT and the OECD, with their corresponding functionalities, and the architectures and functionalities of the resulting OESPs and AMAD devices are described in the following sections.
2. Results and Discussion

2.1. Organic Electrochromic Displays (OECD)

The electrochromic property of PEDOT:PSS makes it possible to use the material as the color changing electrode in OECDs. Furthermore, attributable to the processability of PEDOT:PSS from a water-based emulsion, the display devices can be manufactured via common printing techniques. Yet another advantage of using PEDOT:PSS is that, due to its inherent electronic conductive property, transparent conductive oxide (TCO) materials can be omitted. In general, electrochromic displays can be structured either laterally or vertically, and they can be operated either in transmission or reflection mode. The lateral display architecture is the most simple version and consists of two adjacent PEDOT:PSS electrodes bridged by an electrolyte layer. However, the lateral structure also entails an immediate drawback, namely that the adjacently located counter electrode requires a certain area and therefore significantly lowers the fill factor of the resulting display. Displays operating in transmission mode are appealing, especially since the oxidized state of PEDOT:PSS is close to transparent. However, the major disadvantage of this structure is that a complementary counter electrode is required to obtain an acceptable color contrast, which typically implies the necessity of a TCO material to support the conductivity of the material serving as the complementary electrode with respect to the PEDOT:PSS electrode. Hence, in this work, we are focusing on a vertical OECD operating in reflection mode. A schematic of this display architecture is shown in Figure 1a. The OECDs are manufactured by using screen printing in every processing step; a technique that has been matured at RISE (formerly Acreo) during recent years.[10]

A standard PET foil is used as the carrier, which also adds mechanical protection since the color change of the display is observed through this plastic substrate. The PEDOT:PSS color changing electrode is screen printed directly onto the PET foil, followed by thermal curing to allow water to evaporate from the ink. The display segments are then created by printing an electrolyte layer according to the desired shape on the PEDOT:PSS layer,
followed by UV light exposure to cure the electrolyte layer. In this case, to obtain a display that is operated in reflection mode, the electrolyte contains pigment to make it white and opaque. Optionally, the display segment pattern can also be created via the deposition of an insulating layer, in which holes correspond to the desired shapes of the segments; the electrolyte layer is then instead printed into the holes of the insulating layer. A counter electrode layer is subsequently printed on top of the electrolyte layer. The counter electrode can be selected from a variety of different materials, mainly due to the opaqueness of the electrolyte. PEDOT:PSS is used as both color changing electrode and counter electrode in this particular work. The OECD is completed once the solvent of the counter electrode has been evaporated. Optionally, a highly conducting metal ink may be printed to lower the overall electrical resistance of the resulting device. In addition to this, a mechanical protection layer may also be screen printed on top of the device.

The resulting OECD responds to an electric potential difference applied across the electrodes, and the color state of the display depends on the amplitude and polarity of this voltage. A negative voltage on the counter electrode results in oxidation of the PEDOT:PSS in the color changing electrode into its transparent state, which appears white due to the color of the background electrolyte. A positive voltage on the counter electrode results in reduction of the PEDOT:PSS in the color changing electrode into its dark, blue-colored state. The printed electrochromic display is operated at low voltages, and exhibits very low power consumption. On average, a display segment with an area of 1 cm$^2$ requires approximately 500 µC for each switching event, which corresponds to about $10^5$ switch cycles when supplying the display with a printed battery having a capacity of approximately 10-15 mAh. Figure 1b shows the switching characteristics of an OECD (pixel current (I$P$) vs. time during reduction and oxidation) when applying ±1.5 V for 1 s.
2.2. Organic Electrochemical Transistors (OECT)

The ability to modulate the electronic conductivity of PEDOT:PSS through electrochemical switching is utilized in the OECTs, instead of taking advantage of the electrochromic property of the polymer to enable the color change that is used in the OECD. The active part of the OECT device is relying on the same structure as an OECD; two PEDOT:PSS electrodes bridged by an electrolyte. The electrodes of the OECT can, similarly to the OECD, be arranged either laterally or vertically. The lateral OECT device is manufactured by using a minimum set of processing steps, but it also results in larger footprint and longer switching time,\cite{15} as compared to OECT devices that are processed according to the vertical architecture;\cite{16} the vertical OECT architecture is used throughout this report. In the OECT device the PEDOT:PSS electrodes are denoted gate electrode and transistor channel, and the transistor channel extends outside the area covered by the electrolyte to form the source and drain electrodes. Applying a constant voltage between the source and drain electrodes results in a certain current level through the transistor channel, and by adjusting the gate voltage the current throughput can be modulated by several orders of magnitude; the ON/OFF ratio depends on many parameters and typically varies in the range $10^{3}$-$10^{6}$ in fully screen printed OECT devices. The PEDOT:PSS-based OECT is operated in depletion mode, i.e. the conducting ON state is obtained by applying $\sim$0 V at the gate electrode, while a positive gate voltage switches the OECT channel to its OFF state.

OECTs feature two distinct advantages when compared to OFETs; high current throughput and low voltage operation. The high current throughput, which is advantageous in e.g. AMAD and supercapacitor balancing circuit applications,\cite{11,12,17} is explained by that the complete bulk of the OECT semiconductor is used for charge conduction through the channel, in contrast to the very thin layer of charges that are accumulated in the channel of an OFET. The low voltage operation is enabled by the electrical double layers formed at the electrolytic interfaces, which concentrates the electric field that is applied at the gate electrode. OECDs
and OECTs rely on the same switching mechanism and both devices therefore exhibit low voltage operation; a feature that makes the devices compatible with each other since they can be powered from the same energy source, e.g. a printed 1.5 V battery. Additionally, from a manufacturing point of view, it is of course an advantage that the active parts of both OECDs and OECTs are based on the very same conducting polymer and electrolyte materials, since this significantly reduces the total amount of materials and printing steps in monolithically integrated circuits.\textsuperscript{[18]} The transfer and dynamic switching characteristics of an OECT are shown in Figure 2, along with a schematic of the all-printed vertically configured transistor device.

**2.3. Organic Electrochromic Smart Pixels (OESP)**

An OESP is obtained through the combination of one OECT and one OECD, where the conduction state of the OECT channel controls the color state of the OECD. Figure 3 shows the schematic layout of an OESP and a photo of a screen printed OESP.

Both the OECT and the OECD rely on the same kind of electrochemical switching; oxidation and reduction of PEDOT:PSS. This not only reduces the number of materials in the manufacturing process, it also simplifies the drive protocol since both devices are operated at low voltages. Figure 4 shows the functionality of an OESP in operation. The first part of the measurement cycle, until \(~13\) s, shows that the OECT prevents unintentional coloration of the OECD, while the second part of the measurement demonstrates that the OECT maintains the charge of an already colored OECD. In other words, the former property is used in AMADs to prevent cross-talk effects in neighboring display pixels, while the latter is used in AMADs to increase the retention time of charged pixels, which, in turn, lowers both the refresh pulse frequency and the overall power consumption.
2.4. Active Matrix Addressed Displays (AMAD)

Even though only one OESP is described in Figure 3 and 4, it can be imagined that the concept would work well upon arranging many OESPs into a monolithically integrated AMAD device. The fact that the non-conducting transistor channel withstands the applied pixel voltage indicates that it should be possible to update an AMAD, consisting of many OESPs, without cross-talk. AMADs are typically operated row by row; this is obtained by connecting the gate electrodes of all OECTs along one row by a common row line, and each specific row of the AMAD is uniquely addressed in a subsequent manner through *e.g.*, a decoder circuit. Once a specific row has been selected, the OECDs are updated through the column drive lines. An arbitrary number of pixels in the selected row can be updated simultaneously since the columns are updated in parallel, *e.g.*, by a shift register circuit.

The OECTs, OECDs, OESPs and AMADs were all manufactured solely using screen printing. The pixel size is $5 \times 5 \, \text{mm}^2$ in the characterized OESP and AMAD devices. $3 \times 3$ AMADs were manufactured simply by creating a layout with three rows and three columns, where each intersection contains one OESP device according to the design shown in the photograph in Figure 3. Electrical wires, based on either silver or nano-copper ink, were screen printed onto the flexible foil, and these wires established the electronic connection between the OECD pixels along the columns as well as between the OECTs along the rows, see *Figure 5*a and 5b. The design was targeting $200 \, \mu\text{m}$ line widths and $150 \, \mu\text{m}$ separations, and both inks could be printed at dimensions relatively close to the targeted dimensions. The silver-based ink resulted in line widths of 220-230 $\mu\text{m}$, and therefore a separation of 120-130 $\mu\text{m}$, while the nano-copper-based ink resulted in line widths of 240-250 $\mu\text{m}$ and a separation of 100-110 $\mu\text{m}$. The conductivity of the silver-based electrical wires showed relatively consistent sheet resistance values in the range 50-75 m$\Omega$/□. The sheet resistances of the nano-copper-based electrical wires showed larger fluctuations, and the lowest value observed
was ~100 mΩ/□. However, despite the fluctuating sheet resistance values of the nanocopper-based ink, both inks resulted in electrical wires with sufficiently high conductivity since both carbon and PEDOT:PSS exhibit lower conductivities and therefore limit the current throughput in the AMAD devices.

The AMADs were operated by selecting the rows one by one; a $V_G$ of 0 V was applied to the selected row, while non-addressed rows were biased at a $V_G$ of 1.6 V. Once a certain row had been selected, the desired OECD pixels were colored by applying a sufficiently high $V_P$ of -2.5 V to the color changing electrode(s) of the corresponding column(s). The remaining rows of the AMAD were then selected one by one, until reaching the final row, which implies that an arbitrary image can be displayed with this technique. Decoloration of the OECD pixels was obtained by applying $V_G$=0 V to at least one row of gate electrodes, followed by the application of $V_P$=2 V to the color changing electrode(s) of the corresponding column(s). This report demonstrates the concept of active matrix addressing in AMAD devices having only three rows and three columns, i.e. nine OESPs, which of course is too few to provide any useful images or messages in the resulting display. Nevertheless, besides successful operation by using an active matrix addressing protocol, it also demonstrates that AMADs can be manufactured by only using screen printing, wherein the active OECT and OECD components all are arranged according to the same kind of multilayered vertical architecture, rely on the same types of active materials and are operated by the same kind of low voltage switching mechanism. Figure 5c and Video 1 and 2 in the Supporting Information show the concept of screen printed AMADs. Figure 5d shows an area of a plastic substrate containing screen printed devices: standalone OECTs, 3×2 OESPs and 3×3 AMADs.
3. Conclusions

The reported work demonstrates that it is possible to manufacture monolithically integrated active matrix addressed displays solely using screen printing. The organic electrochemical transistors and the organic electrochromic pixels of the active matrix addressed display device are based on the same active polymer material, PEDOT:PSS, and the combination of PEDOT:PSS and an electrolyte enables current modulation and color switching in transistors and displays, respectively. Addressing lines and low-ohmic connections between electrochemical transistors and electrochromic displays were relying on a screen printed layer of an ink consisting of either silver or nano-copper. Despite the limited number of organic electrochromic smart pixels in the resulting active matrix addressed displays presented herein, the concept is nevertheless demonstrated and the possibility to utilize the technology for displaying an arbitrary message in fully screen printed electronic smart labels in future Internet of Things applications is enabled.

4. Experimental Section

Device Manufacturing: All materials were deposited by flatbed, sheet-fed, screen printing equipment (DEK Horizon 03iX) on top of plastic substrates based on polyethylene terephthalate (PET), for example Polifoil Bias purchased from Policrom Screen. The screen printing tools were based on standard polyester mesh. Electrical wires and contact pads for probing purposes were first screen printed on top of the PET foil, these patterns were based on either silver or nano-copper ink. The silver ink was purchased from DuPont (Ag 5000) and the nano-copper-based ink was provided by Intrinsiq Materials. PEDOT:PSS, Clevios SV3 purchased from Heraeus, which serves as the electrochemically active transistor channel and the electrochromic display electrode material, was then deposited, followed by the addition of carbon source and drain electrodes (7102 conducting screen printing paste purchased from DuPont) on top of the previously deposited PEDOT:PSS transistor channels. Silver,
PEDOT:PSS and carbon inks were dried at 120 °C for 5 min in a hot-air conveyor belt oven, while the nano-copper ink was sintered in a separate process by Intrinsiq Materials. An insulating layer (e.g. 5018 purchased from DuPont) was deposited and UV-cured in the following process step. The purpose of the insulating layer is to define the active areas of the PEDOT:PSS-based transistor channels (W×L≈200×250 µm²) and electrochromic displays (5×5 mm²). This is obtained by openings in the insulating layer and an electrolyte layer subsequently printed into these openings, which provides the ions required to achieve the transistor and display functionalities. The electrolyte (AFI VV009 provided by RISE) was cured by UV light exposure. Finally, another layer of PEDOT:PSS was printed on top of the patterned electrolyte. This layer serves as the counter electrode and the gate electrode in the display and the transistor, respectively. This implies that both transistors and displays are fully screen printed in a vertical arrangement, and that the current throughput and the color contrast is modulated in the respective component upon applying an electric potential difference between the corresponding electrodes. The AMAD requires electronic vias due to the row and column lines that are crossing each other, this was obtained by printing an additional conductor on top of a previously printed insulating layer in these junctions.

Device Characterization: All measurements were performed at a temperature of ~20 °C and at a relative humidity of ~45 %RH. Characterization of individual OECDs and OECTs was performed by using a semiconductor parameter analyzer (HP/Agilent 4155B) and a waveform generator (Agilent 33120 A). Individual OESP s and 3×3 AMADs were characterized by using a data acquisition card (DAQCard-6036E from National Instruments). The DAQ card supplied the required voltages to the OECD and the OECT of the OESP, or the rows and columns of the AMAD, respectively.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.
Acknowledgements

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References


**Figure 1.** a) Schematic architecture of a vertically configured OECD device. The color change of the display electrode is observed through the plastic substrate, and this architecture is operated in reflection mode due to a white, opaque electrolyte layer. The electrical wires are screen printed from an ink based on either silver or nano-copper. b) The switching characteristics of a standalone 5×5 mm² OECD, i.e. the current through the display device is not governed by the OECT, upon applying ±1.5 V as the switching voltages. The current vs. time behavior is relatively similar when comparing reduction and oxidation, note that the absolute values of the oxidation data are plotted. The charges injected/extracted during 1 s of reduction/oxidation amount to approximately 150 µC for the respective switching event.
Figure 2. Typical transfer and dynamic characteristics of an all-printed OECT device are shown in a) and b), respectively. A maximum ON/OFF ratio of $\sim 1.6 \times 10^4$ is obtained in the transfer sweep, while an ON/OFF ratio of $\sim 2 \times 10^3$ is obtained in the dynamic measurement. c) The schematic illustrates the architecture of a vertically configured OECT device.
Figure 3. a) Illustration of an OESP; the amplitude of the gate voltage ($V_G$) controls the conduction state of the OECT channel, and thereby also the color state of the OECD independent of the applied pixel voltage ($V_P$). When the OECT is switched to its conducting state ($V_G=0$ V), the amplitude and polarity of $V_P$ can be used to set the OECD to a specific color state. b) Microscope image showing the OECT and its connection with one OECD, together they form a fully screen printed OESP device. Note that only the OECT and a small part of the $5\times5$ mm$^2$ OECD are shown in the image.
Figure 4. The OESP functionality is demonstrated by this graph showing the pixel current ($I_P$, black line) vs. time. The OECD is initially in its white OFF state. After ~4 s, the gate voltage ($V_G$, red line) is applied and the channel is thereby switched to its non-conducting OFF state. After ~6 s, a pixel voltage ($V_P$, blue line) of -1.5 V is applied. However, no OECD coloration current peak can be observed due to the non-conducting OECT channel. Hence, the first part of the test demonstrates that the OESP withstands the voltage supplied by $V_P$. After ~12 s, $V_G$ is changed to 0 V, which switches the OECT channel to its ON state. $V_P$ is also changed to 0 V at the same time, followed by a short (2 s) $V_P$ oxidation pulse at 0.5 V after ~16 s, the purpose of this voltage pulse is to set the OECD into a well-defined color state before starting the second part of the test. After ~18 s, the OECD is reduced by -1.5 V to its blue colored ON state; a switching event requiring ~93 µC. At ~22 s the OECT channel is switched to its OFF state, and it is kept in this state during 24 s. $I_P$ is maintained at a very low level during this sequence, despite that a positive $V_P$ of 1.5 V (from 46 s to 54 s) is being used in an attempt to force decoloration of the OECD. Finally, the OECT channel is switched to its ON state by changing $V_G$ to 0 V, which results in decoloration of the OECD into the previously obtained well-defined color state by that $V_P=0.5$ V concurrently is applied. ~87 µC is released during the OECD decoloration switching event. Hence, the second part of the test demonstrates that
the OECT can be used to retain charges in a colored OECD and thereby prolong the retention time, since \(~94\%\) of the coloration charge is maintained within the OECD during the measurement cycle.
Figure 5. The a) and b) image shows screen printed electrical wires based on silver and nanocopper, respectively. c) The image shows the concept of active matrix addressing, i.e. an electronic display device in which the pixels are colored according to a certain pattern without cross-talk. The colored diagonal line is only obtainable by alternating the gate voltages to activate the rows one by one, so that each transistor controls the color state of its corresponding pixel. Without the alternating gate voltages, either all nine, or none, of the pixels would become colored, hence, unique pixel addressability would be disabled. d) The image shows an area of a sheet containing screen printed organic electronic devices: standalone OECTs (lower section of the image), 3×2 OESP arrays (upper right) and 3×3 AMADs (upper left).
Supporting Information

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The video clips illustrate unique addressability of organic electrochromic displays upon combining them with organic electrochemical transistors into organic electrochromic smart pixels. The color state of each display pixel is controlled via its corresponding transistor, and the resulting active matrix addressed displays are manufactured solely using screen printing. Low voltages are used during the pixel update; the gate voltage is switched between 0 V and 1.6 V and the pixel voltage is switched between -2 V and 2.5 V. One video clip (Video 1) demonstrates an update of an active matrix addressed display forming a diagonal line, while a cross is demonstrated in the other video clip (Video 2); two pixel patterns that only are accessible through the combination of addressing transistors and an active matrix drive protocol.