Towards woven logic from organic electronic fibres

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Published online: 4 April 2007; doi:10.1038/nmat1884

The use of organic polymers for electronic functions is mainly motivated by the low-end applications, where low cost rather than advanced performance is a driving force. Materials and processing methods must allow for cheap production. Printing of electronics using inkjets[1] or classical printing methods has considerable potential to deliver this. Another technology that has been around for millennia is weaving using fibres. Integration of electronic functions within fabrics, with production methods fully compatible with textiles, is therefore of current interest, to enhance performance and extend functions of textiles[2]. Standard polymer field-effect transistors require well defined insulator thickness and high voltage[3], so they have limited suitability for electronic textiles. Here we report a novel approach through the construction of wire electrochemical transistor (WECT) devices, and show that textile monofilaments with 10–100 μm diameters can be coated with continuous thin films of the conducting polythiophene poly(3,4-ethylenedioxythiophene), and used to create micro-scale WECTs on single fibres. We also demonstrate inverters and multiplexers for digital logic. This opens an avenue for three-dimensional polymer micro-electronics, where large-scale circuits can be designed and integrated directly into the three-dimensional structure of woven fibres.

The industrial revolution was spun out of textile production and a wealth of knowledge is integrated in this mature production technology. The number of potential applications for multifunctional fabrics or electronic textiles is tremendous[2], and could open new avenues of development in many areas. Even though most of the demonstrated prototypes of functional electronic textiles are today based on the approach of integration of conventional electric components into clothes[4,5], true integration of electronic materials into textiles is a necessary route for the realization of future e-textiles. Some of the advantages of organic electronic materials, and especially conjugated/conducting polymers as compared to their inorganic counterparts, include high elasticity, mechanical flexibility and an unlimited number of chemical-synthesis and processing possibilities, which allows for a natural integration of conjugated/conducting polymers into fabrics[6,7]. These materials are therefore a most promising class of materials for future e-textile applications[8]. One of the main challenges of all-organic e-textiles is the realization of textile fibres endowed with electronic functions. The realization of polymer-based single-fibre electronic devices opens the way for truly low-cost, large-area e-textiles.

The most important electrical component in fibre electronics (and in conventional electronics) is the transistor. Present day organic electronics mainly uses the field-effect transistor and both this and bipolar transistors have been implemented in planar technology. The difficulties of moving the conventional field-effect device onto a textile fibre are very large, if not insurmountable. Conventional organic field-effect transistors (OFETs) suffer from a number of serious drawbacks, including the sensitivity to the gate insulator thickness, the large operating voltages, which prevent them from safely being incorporated into e-textile, the need for using metals as the fibre core and the need for micro-patterning along each individual fibre, through masking and evaporation steps. Conventional OFETs that are directly patterned on fibres are not believed to ever rival planar devices in performance[3,9]. Alternative devices are therefore needed to create organic fibre electronics.

Here we show that a class of all-organic transistors called electrochemical transistors (ECTs) is an attractive solution for the realization of active transistor components on individual fibres. The operation of an ECT is on the basis of a reversible process of doping/de-doping of electronic polymers. One of the most promising materials so far for ECTs is the low-bandgap conducting polymer poly(3,4-ethylenedioxythiophene) (PEDOT), owing to its high conductivity, water dispersibility, for example in the form of poly(3,4-ethylenedioxythiophene)/poly(styrene sulfonic acid) (PEDOT/PSS), and environmental stability.

The operation of PEDOT ECTs relies on switching the conductivity of a transistor channel consisting of PEDOT/PSS, through a reversible redox process in thin films of PEDOT that are in contact with a common electrolyte. Doping and de-doping of PEDOT can result in conductivity changes of up to five orders of magnitude[10]. Figure 1 demonstrates an ECT on a planar substrate where ECTs have previously been demonstrated on flat surfaces[11,12] (Fig. 1a), such as glass, plastics and paper, through patterning of the conducting polymer film followed by patterning of a second layer of electrolyte. Large-scale digital logic devices[13] and displays[14] have also been demonstrated by patterning a number of ECTs on flat surfaces. ECTs can however also be realized in non-planar geometries. By using cylindrical PEDOT films it is possible to construct ECTs on wires. We call such transistors wire ECTs or WECTs. One attractive design of a WECT is shown in Fig. 1b, where two fibres act as substrates for cylindrical thin PEDOT/PSS films, with an electrolyte contact at the junction of the fibres. The realization of these WECTs on textile fibres requires monofilaments of textile fibres that are coated with PEDOT films. PEDOT has previously been coated on textile from an aqueous solution of poly(3,4-ethylenedioxythiophene) doped with poly(styrenesulfonate) (PEDOT/PSS)[15], and *in situ* polymerization

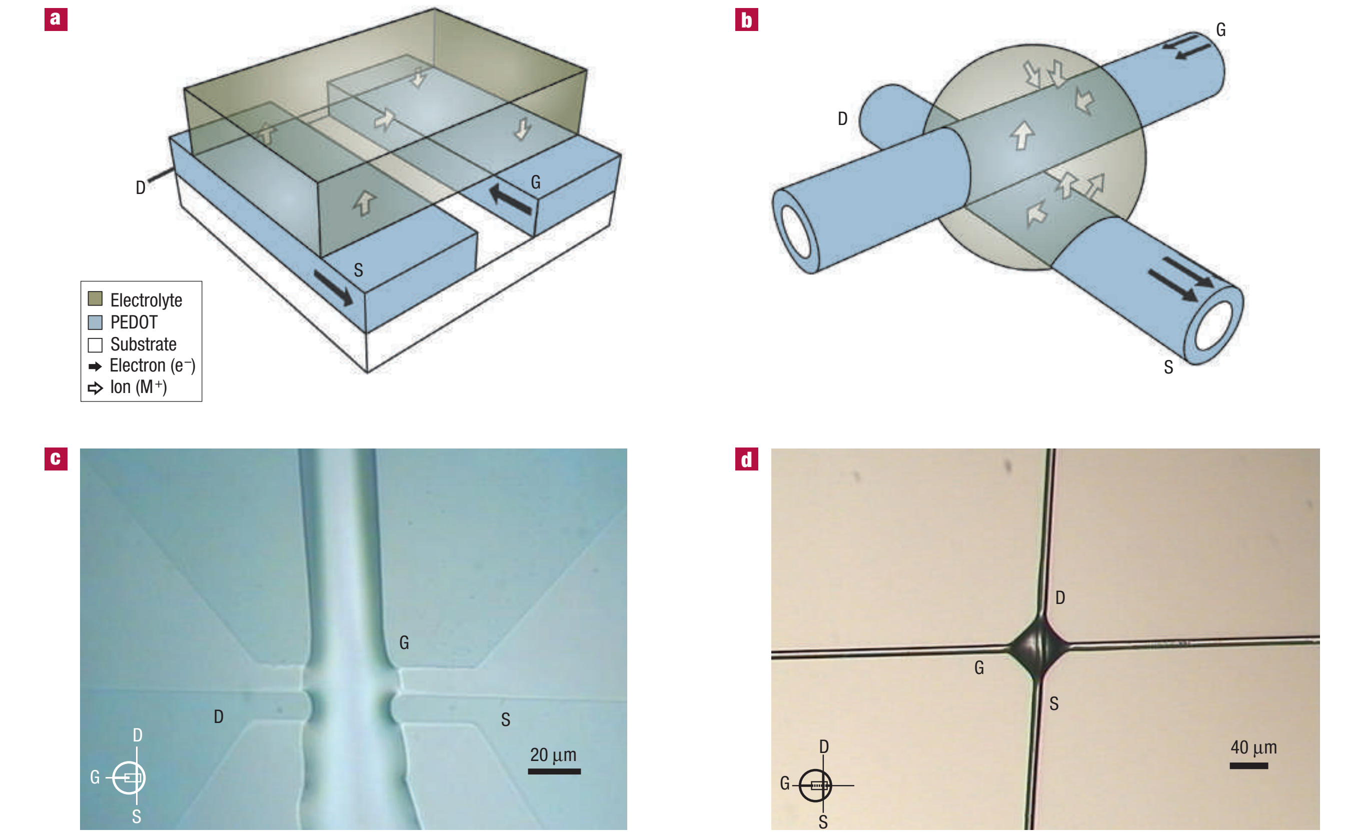

Figure 1 Pictures of planar and cylindrical electrochemical transistors (ECTs). a, Schematic picture of a planar ECT (S, source; D, drain; G, gate). **b**, Schematic picture of a wire ECT (WECT). **c**, Optical micrograph of a planar micro-ECT (channel width = 15 μm). The dark-blue structures are PEDOT/PSS micro-patterns that were created on plane glass substrate through subtractive photo-lithography. The vertical line crossing the PEDOT/PSS structure is a solid polymer electrolyte structure that was patterned from solution using the soft-lithography technique of micro-moulding in capillaries. **d**, Optical micrograph of a WECT constructed at the junction of two PEDOT/PSS-coated polyamide monofilaments with 10 μm diameter. Electrical circuit symbols for the ECTs are also shown in **c** and **d**.

of PEDOT on nylon-6 and polyethylene terephthalate[7]. We have shown that textile can also be created by weaving coated monofilaments instead of coating the already woven textile. We demonstrated this by using a high-conductivity[16] formula of PEDOT/PSS for solution coating of monofilaments of a number of common textile materials including nylon (polyamide). The PEDOT/PSS film showed very strong adhesion and uniformity along the monofilaments (see Supplementary Information, Figs S1,S2). The fibre coating can withstand mechanical stretch and scratch using standard textile processes such as weaving and knitting, allowing for the construction of electronic structures. Construction of fabric from coated monofilaments has many advantages, such as the possibility of fibres with continuous and uniform coatings (see Supplementary Information, Fig S2) and the possibility of different fibres in the same fabric.

Single WECTs were realized by suspending two PEDOT/PSS-coated fibres in a cross geometry and creating an ionic contact by adding a solid polymer electrolyte from solution, at the junction of fibres (Fig. 1d). Surface energy helps direct the fluid carrying the solid polymer electrolyte to the crossing, where it covers both fibres entirely and dries. Electrical characteristics of two WECTs are shown in Fig. 2b,d. The WECTs show current saturation[17] with increasing drain voltage. The transistor is in the on state at gate voltage 0 V; as the gate voltage is increased, the transistor channel is depleted and the transistor is turned off, with on/off ratios greater than 1,000 for very low gate voltages between 0 and 1.5 V (Fig. 2a). The *I*–*V* character of the ECT is similar to the solid-state p-type depletion metal–oxide–semiconductor field-effect transistor. For comparison between the WECTs and planar ECTs, a planar ECT with micrometre channel dimensions was constructed using a combination of photo-lithography and soft-lithography patterning (Fig. 1c). The *I*–*V* characteristics of the planar micro-ECT (Fig. 2c) show current saturation characters and on/off ratios similar to that of the WECT, even though the WECTs possess a completely different geometry. This similarity is due to the fact that the operation of the electrolyte-gated transistor is dominated by the interface between the electrolyte and the conducting polymer film, where the main potential difference is located. Owing to the interface-dominated operation, the active part of the transistor channel comprises the entire cylindrical film which is in contact with the electrolyte. Furthermore, the operating voltage is insensitive to the distance between the gate and the channel, and the WECT always operates at voltages between 0 and 1.5 V, which is the electrochemical potential window of the PEDOT/PSS.

The redox conversion using an electrolyte contact thus solves all the major drawbacks of conventional field-effect device operation, such as sensitivity to gate distance, which make impossible the switching operation of an entire cylindrical film at micro-dimensions using another fibre as gate, and the need for

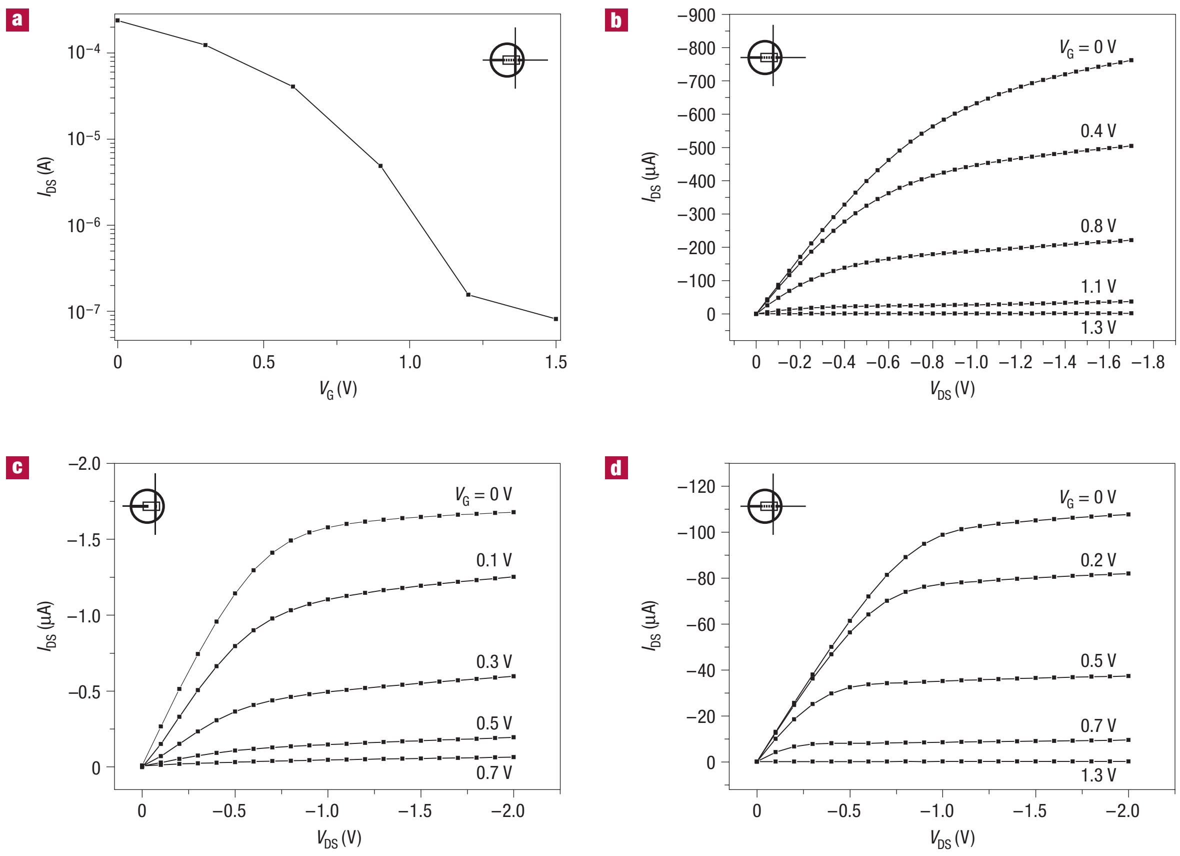

Figure 2 Electrical characteristics of electrochemical transistors. a, Transfer characteristics of a WECT with 100 μm fibres for both channel and gate. **b**, Output characteristics of a WECT with 100 μm fibres. **c**, Output characteristics of the planar ECT depicted in Fig. 1c. **d**, Output characteristics of the WECT in Fig. 1d, with 10 μm fibres for both channel and gate.

high gate voltages. Furthermore, the WECT operation is insensitive to vertical displacement between fibres, owing to the interface operation of the device, and it is also insensitive to horizontal displacement along fibres because source and drain contacts consist of the same material as the channel and the gate. The WECT operation is also insensitive to the shape or amount of the electrolyte. In summary, the local geometry of the fibres or electrolyte patterns does not have a major impact on device function, and the need for precise positioning and stability of geometry is gone. This matches the production of fabrics, and the need to make these take on many geometries and drapes. WECTs can therefore be easily constructed across any micro-fibre junction in a three-dimensional weave. The fabrication of WECTs also eliminates the need for lithography patterning steps, which are only two dimensional and hardly compatible or cost efficient for e-textiles.

The WECTs not only possess interesting properties for electronic fabrics, but they also have characteristics that are beneficial for large-scale electronic design on fabrics. WECTs are symmetrical, meaning that principally any of the four connection points to the transistor can be chosen as a gate and any of the corresponding two connections on the other fibre can then be chosen as source and drain. It is also possible to place many transistors along one single fibre because the channel consists of the same conducting material as the rest of the fibre. The WECT component can also easily be completed with ohmic connectors, for example by placing fluids of conductive polymers at fibre junctions. Furthermore, the coated monofilaments have a constant resistance/length (see Supplementary Information, Fig. S2), which makes possible the realization of resistors of various sizes, by using varying fibre lengths and also by using different formulas of PEDOT/PSS with different conductivity values. The symmetry and ease of construction of WECTs together with the passive components and the unlimited possibilities of different three-dimensional fabric topologies and material mixtures can be used for the realization of large-scale integration of general micro-electronics directly on fabrics. We have demonstrated that two transistors at a distance of $2d$ (d = fibre diameter) on a fibre can operate without any crosstalk. The demonstration of WECTs (Fig. 1d) on 10 μm fibres therefore indicates that it would be theoretically possible to construct ~100,000 transistors cm^{-2} using 10 μm fibres and a two-dimensional mesh. Transistor packing densities would increase by taking advantage of the third dimension. The architectural design of any general fabric electronic circuit, for example digital circuits, is now just a question of design, where the combination of three-dimensional fabric design followed by textile patterning can be used to construct a complete device, as schematically shown in Fig. 3c.

We have designed electric circuits by using a fabric circuit diagram (FCD) containing proposed symbols for wires/fibres, WECTs and ohmic connections (Fig. 3a). Conventional circuit diagrams can be translated into FCDs as shown in Fig. 3a. Fabric circuits can then be created as a direct replica of the FCD, for example by weaving a mesh similar to the FCD wires and then patterning WECTs and ohmic connections according to the FCD pattern at the fibre junctions.

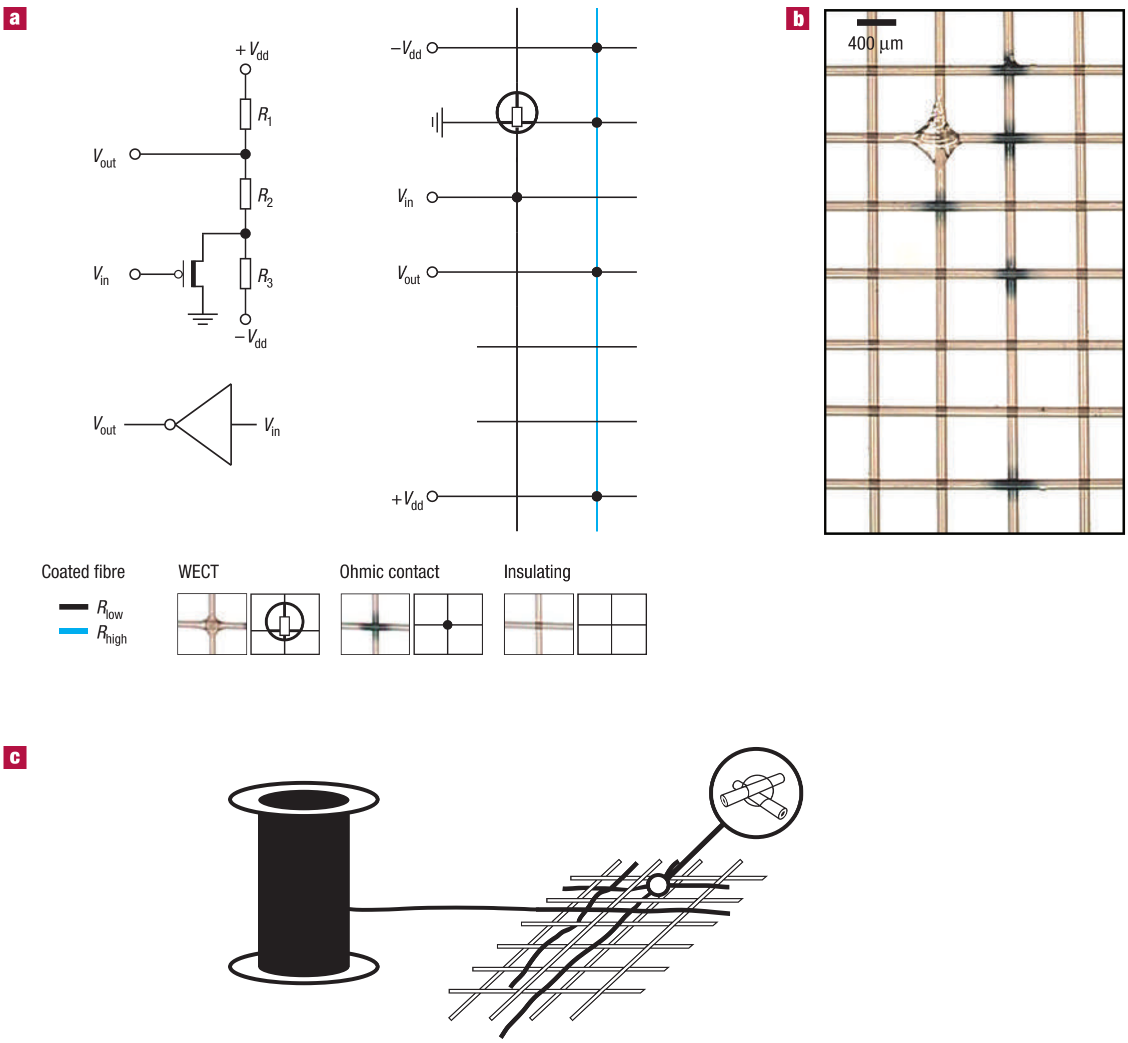

Figure 3 Design and construction of logic circuits. a, A schematic picture showing on the left a classical circuit diagram design of an inverter, based on a p-type depletion FET, and three resistors, where $R_2 + R_3 = R_1$. The corresponding fabric circuit diagram (FCD) of the inverter is shown on the right, containing symbols for the WECT, and for ohmic connection between wires. The blue and black wires represent coated fibres with low- and high-resistance coatings. **b**, Optical micrograph of a fabric/mesh constructed from the FCD design of **a**. The fibres are coated polyamide monofilaments with 100 µm diameter. The low-resistance wires were coated with Baytron P, 5 wt% DEG: 100 S cm^{-1}, and the high-resistance wires were coated with Baytron P, 1.6% DEG: 10 S cm^{-1}. The mesh was constructed by hand knitting of coated monofilaments. PSS electrolyte was used for WECTs and PEDOT/PSS was used for ohmic connections, and these were placed and self-assembled at fibre junctions from solution form using micro-nozzles. **c**, A schematic picture demonstrating the creation of organic electronic textiles by weaving coated monofilaments.

We have demonstrated universal logic operations by constructing an inverter on the basis of a voltage shifting logic design, comprising three resistors and a depletion p-type transistor. A possible translation of the inverter circuit diagram into an FCD is shown in Fig. 3a. This FCD was then constructed on fibres (Fig. 3b) by manually creating a fibre crossbar from coated monofilaments and placing electrolyte and PEDOT at the crossbar junctions. The dynamic switching characteristic of the inverter in Fig. 3b is shown in Fig. 4d. The inverter operates at 1–2 V, which is more than an order of magnitude lower than logic on the basis of conventional OFETs.

A more complex fundamental digital addressing device or multiplexer was also realized on fibres. The multiplexer enables encoding of information from a large number of data sources into a single channel, and this device can be designed on a fibre crossbar, by placing WECTs in a pattern that represents a binary tree multiplexer structure, where N channel lines are addressed using $2\log_2 N$ address wires. This design takes advantage of the three dimensionality of the fabric, where one fibre is used as the gate for several transistors. Figure 4a shows a circuit diagram of a multiplexer with four channel lines, and Fig. 4b shows the realization of the binary tree portion of the multiplexer on a fibre mesh. Dynamic operation characteristics of the multiplexer is shown in Fig. 4c, where each of the four channel lines is uniquely addressed using the four possible binary combinations as inputs to the two address lines.

To isolate and insulate the individual fibre to electrical and chemical disturbance from the environment, a coating with an insulating polymer from solution or dispersion is envisaged. This coating would be done after formation of a system on fibres. The porous mesh should allow for effective coating.

The realization of fabrics from electrically active monofilaments together with the construction of WECTs constitute a new step towards the realization of analogue and digital micro-electronics

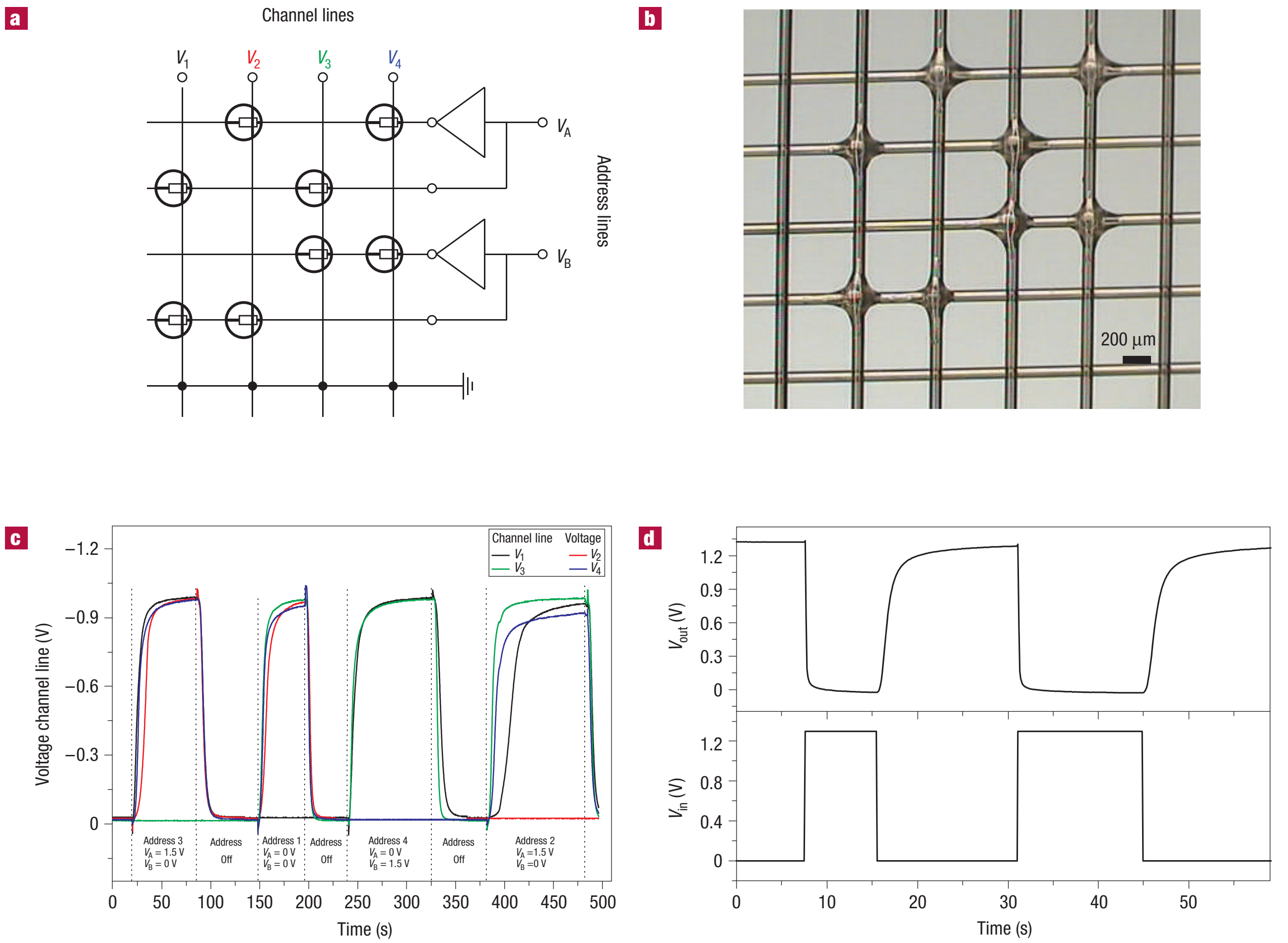

Figure 4 Design and electrical characterization of logic circuits on fibres. a, Circuit diagram of a binary tree multiplexer with two address lines and four channel lines. **b**, Optical micrograph of a binary tree multiplexer on a fabric mesh. The fabric was constructed using the same method and material as for the inverter in Fig. 3b. **c**, Dynamic electrical characteristics of the multiplexer in **a**. **d**, Dynamic switch characteristics of the inverter shown in Fig. 3a.

directly into textile, and open many new possibilities for integration of electronic function on new carriers. These may be relevant as addressable neural implants with biocompatible electronics, as the electrode material PEDOT has been demonstrated to work well as a neural electrode[18]. They could be used as computational wear for sensors and actuators including electrochromic displays and chemical sensors, and the inspiration for building three-dimensional micro-electronic systems using production methods of textile technology. The concept of electrolyte-gated ECTs on fibre can probably be extended to other all-organic electrolyte-gated components, such as electrolyte-gated OFETs or light-emitting electrochemical cells. Some of these devices also have the potential of being realized on submicrometre wire dimensions, using coated nano-wires or assembled nano-wires incorporating electronic polymers[19].

METHODS

COATING OF MONOFILAMENTS AND ELECTRICAL CHARACTERIZATION

We used a high-conductivity formula of PEDOT/PSS containing 95% by weight of PEDOT/PSS (Baytron P from Bayer, Krefeld, Germany), 5% of the secondary dopant diethylene glycol and 0.1 wt% of the surfactant Zonyl FS-300 (Chemika/Fluka) for coating the monofilaments. The coating was carried out using a vertical flow of the PEDOT/PSS fluid in the form of drops, under the influence of gravity, down the exterior of the suspended monofilaments. This method was proven to be versatile and reproducible, and most of the common textile fibre materials, such as polyamide, Kevlar and polyester, have been successfully coated up to several metres, and with fibre diameters varying from a few hundred micrometres down to 5 μm.

Resistance was measured as a function of applied force along coated polyamide monofilaments, using a four-probe set-up and a Keithley 4200 parameter analyser, where the fibre was mechanically probed using gold-coated micro-probes. The resistance of coated monofilaments with 100 μm diameters (Wonder thread, Shakespeare, UK) remained fairly unchanged under loads that correspond to the loads used in processing the fibres in processes such as weaving or knitting.

Resistance measurements were also carried out over longer distances along coated fibres (see the Supplementary Information), using a two-probe measurement set-up. These measurements showed that uniform coating of fibres is also achieved at lengths greater than 1 m. Using the measured resistance and assuming uniform coating, we estimate the thickness of the PEDOT/PSS layer to be ≈300 nm.

FABRICATION AND ELECTRICAL CHARACTERIZATION OF CIRCUITS ON FIBRES

Polyamide monofilaments were coated with PEDOT/PSS containing 5 wt% diethylene glycol and 0.1 wt% Zonyl. The coated fibres were manually attached to anchor points to create a crossbar geometry. Each wire electrochemical transistor (WECT) was constructed by placing a drop of solid polymer electrolyte containing 33 wt% PSS, 12wt% glycol, 8 wt% sorbitol, water and 0.1 molar $NaClO_4$ at a fibre junction. Ohmic connections were constructed by placing drops of PEDOT/PSS (Baytron P from Bayer, Krefeld, Germany) at fibre junctions. The solutions were placed manually using micro-syringes under an optical stereo microscope. The fibres were connected to metallic wires using silver paint. The metallic wires were connected to a data acquisition device (USB1208-LS from Measurement Computing). Data were collected from the device using LabVIEW (National Instruments).

DEVICE FABRICATION OF PLANAR ELECTROCHEMICAL MICRO-TRANSISTORS

The planar ECTs (Fig. 1c) were constructed in two steps, where in the first step a micro-pattern of PEDOT/PSS was created and in a second step a micro-pattern of a solid polymer electrolyte was patterned on top.

The PEDOT/PSS micro-pattern was constructed in the following steps. (1) PEDOT/PSS containing 10 wt% glycol was spin coated on a glass substrate at 1,500 r.p.m. (2) The coated glass substrate was heated to 100 °C for 10 min. (3) A layer of positive photo-resist (Shipley 1818) was spin coated on the glass substrate, and a micro-pattern was constructed using a standard photo-lithography procedure. (4) The substrate was plasma etched in an RF oxygen plasma at 200 W for 10 min, in order to remove naked PEDOT/PSS structures. (5) The resist was finally stripped using acetone.

The electrolyte pattern was constructed in the following steps. (1) A replica of a micro-channel with 3 μm height and 15 μm width was replicated in the elastomeric material (Sylgard-184 poly(dimethyl siloxane) (PDMS) from Dow Corning). (2) The elastomeric stamp was aligned and placed on top of the PEDOT/PSS micro-structure. (3) A solid polymer electrolyte containing (33 wt% PSS with a molecular weight of 70,000, 12 wt% glycol, and 8% sorbitol, 0.1 molar $NaClO_4$, and water) was placed at the channel opening. (4) The electrolyte entered the channel by capillary forces, and dried. (5) The stamp was removed.

Electrical measurements were made by first painting silver paint on the source, drain and gate pads and contacting the silver paint using microprobes. A Keithley 4200 parameter analyser was used for measurement and data collection.

Received 15 January 2007; accepted 28 February 2007; published 4 April 2007.

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Acknowledgements

These investigations were financially supported by the Centre of Organic Electronics (COE) at Linköping University, Sweden, financed by the Strategic Research Foundation SSF. We thank W.-Y. Lin for electrical measurements on single WECTs and monofilaments, M. Asplund for discussions and K. Hamedi for graphic design. Textile fibres were kindly donated by Shakespeare, UK, and IFP Research, Mölndal, Sweden.
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Supplementary Information accompanies this paper on www.nature.com/naturematerials.

Author contributions

M.H. carried out experiments. R.F. contributed to development of the logic design. O.I and M.H. wrote the manuscript. M.H., O.I. and R.F contributed to project planning.

Competing financial interests

The authors declare competing financial interests: details accompany the full-text HTML version of the paper at www.nature.com/naturematerials/.