

Flexible organic transistors and circuits with extreme bending stability

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Flexible electronic circuits are an essential prerequisite for the development of rollable displays, conformable sensors, biodegradable electronics and other applications with unconventional form factors. The smallest radius into which a circuit can be bent is typically several millimetres, limited by strain-induced damage to the active circuit elements. Bending-induced damage can be avoided by placing the circuit elements on rigid islands connected by stretchable wires, but the presence of rigid areas within the substrate plane limits the bending radius. Here we demonstrate organic transistors and complementary circuits that continue to operate without degradation while being folded into a radius of 100 μm . This enormous flexibility and bending stability is enabled by a very thin plastic substrate (12.5 μm), an atomically smooth planarization coating and a hybrid encapsulation stack that places the transistors in the neutral strain position. We demonstrate a potential application as a catheter with a sheet of transistors and sensors wrapped around it that enables the spatially resolved measurement of physical or chemical properties inside long, narrow tubes.

An important trend in large-area electronics for display and sensor applications is mechanical flexibility^{1–22}. The ultimate goal is electronic systems that cannot only be flexed (into a bending radius of a few centimetres or perhaps a few millimetres), but can also be tightly rolled, bent around sharp edges or repeatedly creased without degradation of the electronic functionality. This requires the development of devices that can withstand extremely small bending radii without suffering damage. Although there are many reports of flexible organic and inorganic transistors^{6–12}, in all previous reports the transistors were degraded or destroyed when being bent into a radius smaller than a few millimetres, owing to damage by the bending-induced mechanical strain. There is one report by the Princeton University group⁹ of transistors that survive bending to a radius of 500 μm , but these transistors require relatively high operating voltages (15 V), they were not tested during bending (only before and after bending), and integrated circuits were not demonstrated. Although bending radii of 500 μm to a few millimetres are sufficient for displays or sensors that can be wrapped around a macroscopic object^{1,2}, much smaller bending radii are required if the devices are expected to survive repeated crumpling, creasing or sharp folding. In addition to large bending stability, the transistors and circuits should also have good electrical performance characteristics, such as large carrier field-effect mobility and low operating voltage, and they should not exhibit bending-stress-induced changes or degradation. As Table 1 illustrates, combining extreme bending stability, large field-effect mobility and low operating voltage has been rather challenging. Here we demonstrate organic transistors and circuits with mobilities of 0.5 $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ that operate with supply voltages of only 3 V and while being folded into a bending radius as small as 100 μm . To demonstrate a potential electronic application that requires high-performance circuits that operate while being folded into extremely small bending radii, we have manufactured a very thin catheter that measures the spatial distribution of pressure by using a foldable transistor and sensor matrix wrapped around its surface in a helical structure.

The first prerequisite for the fabrication of transistors that can be bent into a small radius is the use of a very thin substrate. Organic thin-film transistors (TFTs) are often fabricated on sheets of polyimide, polyethylene naphthalate or polyethylene terephthalate that are typically around 100 μm thick, with a few reports of organic TFTs on substrates as thin as 25 μm (ref. 1). To allow bending into radii well below 1 mm, we use a polyimide substrate that has a thickness of only 12.5 μm (UPILEX-12.5S, Ube Chemical). These substrates have a surface roughness of about 2.5 nm r.m.s. (see Supplementary Fig. S1b), much rougher than silicon substrates (see Supplementary Fig. S1a) and too rough to permit the formation of well-ordered organic semiconductor films with large field-effect mobility²³. To reduce the surface roughness, our polyimide substrates are coated with a 500-nm-thick polyimide planarization layer (KEMITITE CT4112, Kyocera Chemical), which is deposited by spin-coating and cured at a temperature of 180 °C. The surface roughness of the planarization layer is 0.3 nm r.m.s., which is similar to the surface roughness of silicon dioxide layers thermally grown on carefully polished single-crystalline silicon wafers (see Supplementary Fig. S1c).

On this very thin, almost atomically smooth surface we have fabricated organic TFTs and organic complementary circuits with excellent static and dynamic performance. As Fig. 1a shows, the thin polyimide foil is indeed extremely flexible. The schematic structure of the organic TFTs is shown in Fig. 1b. Twenty-nanometre-thick aluminium (Al) gate electrodes, a 6-nm-thick hybrid aluminium oxide (AlO_x)/organic self-assembled monolayer (SAM) gate dielectric, 30-nm-thick semiconductor layers and 50-nm-thick Au source/drain contacts were sequentially formed on the planarization layer, so the total thickness of the TFTs is only 106 nm. The organic semiconductor is either pentacene (for the p-channel TFTs) or hexadecafluorocopperphthalocyanine (F_{16}CuPc , for the n-channel TFTs). Al gates, organic semiconductors and Au source/drain contacts were all patterned using shadow masks, so that no resist baking is required and the maximum process temperature is less than 100 °C. Creating both p-channel and n-channel

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Table 1 | Comparison with literature data.

	Polycrystalline silicon TFTs (ref. 6)	Metal oxide TFTs (ref. 7)	Amorphous silicon TFTs (refs 8,9)	Organic TFTs (ref. 10)	Organic TFTs (ref. 12)	Organic TFTs (this work)
Field-effect mobility	$>10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	$7 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	$0.5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	$0.5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	$0.1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$	$0.5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$
Operating voltage	4 V	10 V	15 V	40 V	2.5 V	2 V
Minimum bending radius	10 mm	30 mm	0.5 mm	0.5 mm	2.5 mm	0.1 mm

Carrier field-effect mobility, operating voltage and bending stability reported for inorganic and organic TFTs.

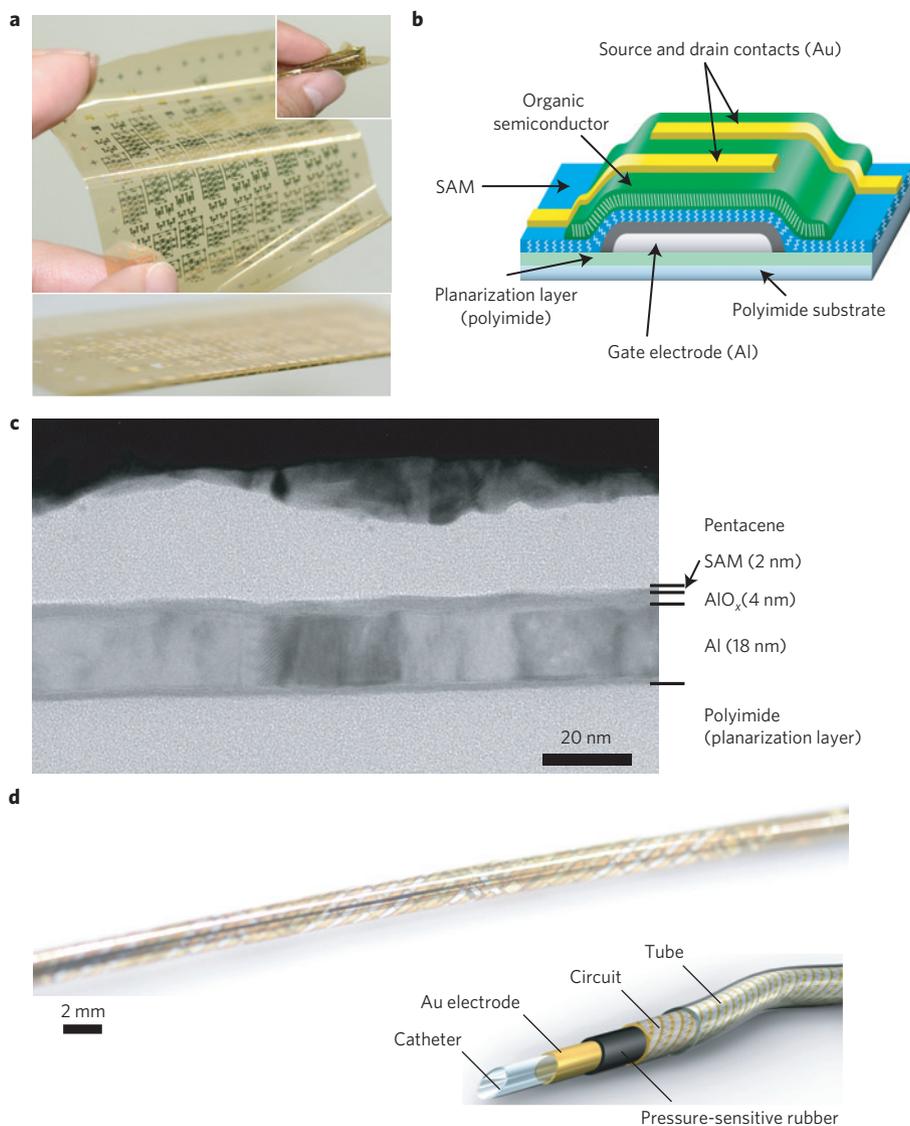


Figure 1 | Ultraflexible integrated circuits. **a**, Photographs of a 12.5- μm -thick polyimide substrate with functional organic TFTs and organic complementary circuits. The array has an area of $75 \times 75 \text{ mm}^2$. **b**, Schematic cross-section of the TFTs. **c**, High-resolution cross-sectional electron microscopy image of a flexible TFT. The specimen was prepared using a focused ion beam and imaged by transmission electron microscopy (300 kV). **d**, Photograph of a thin catheter that measures the spatial distribution of pressure along its length and circumference by means of an active-matrix sensor helix. A cross-sectional illustration of the development view is also shown.

TFTs allows for the design of complementary circuits, which have substantially smaller power consumption, larger gain and greater noise immunity than unipolar circuits^{24–26}.

The final process step is the deposition of a hybrid encapsulation stack that serves two purposes. As the encapsulation stack is composed of a metal layer (Au, 200 nm thick) sandwiched between two polymer layers (parlylene, 300 nm and 12.5 μm thick), it is

an excellent barrier against the diffusion of oxygen and humidity from the ambient air into the organic semiconductors, and hence greatly increases the air stability of the devices and circuits²⁷. Moreover, as the encapsulation stack has the same thickness as the polyimide substrate (that is, 13 μm), the TFTs and circuits are now located in the neutral strain position (where bending-induced compressive and tensile strain cancel each other), which

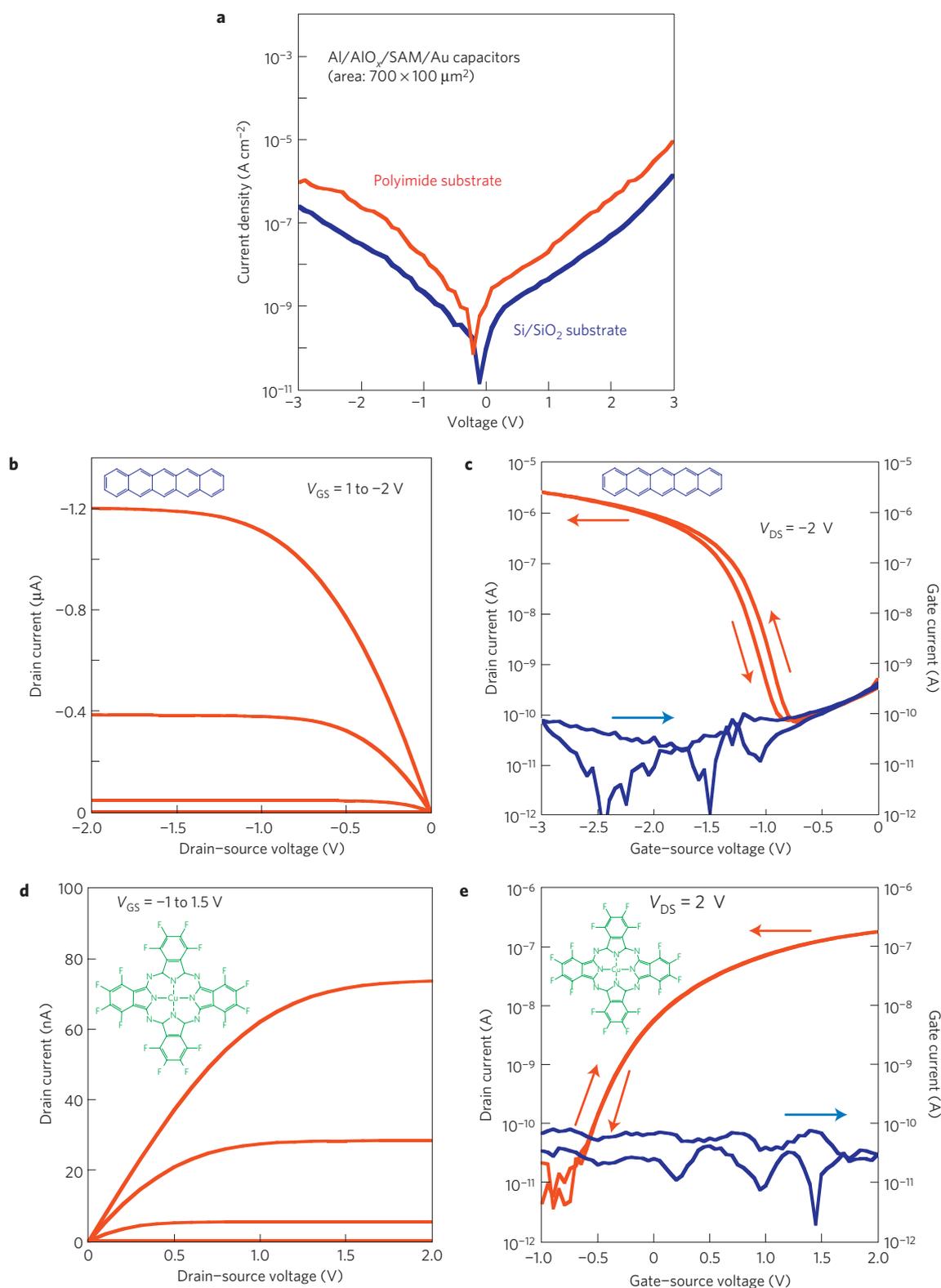


Figure 2 | Performance of organic TFTs on plastic substrates. **a**, Leakage current through the 6-nm-thick AlO_x/SAM gate dielectric as a function of applied voltage. Measurements were carried out on Al/AIO_x/SAM/Au capacitors on two different substrates: a 12.5-μm-thick polyimide substrate with a 500-nm-thick polyimide planarization layer (red curve), and a thermally oxidized, single-crystalline silicon wafer (blue curve). **b**, Drain current as a function of drain-source voltage for a pentacene p-channel TFT (gate-source voltage (V_{GS}) in steps of 0.5 V). **c**, Drain current and gate current as a function of gate-source voltage for a pentacene p-channel TFT (drain-source voltage (V_{DS}): -2 V). Owing to the small thickness of the gate dielectric (6 nm), the operation voltage is about 2 V. The field-effect mobility extracted from the transfer characteristics is 0.5 cm² V⁻¹ s⁻¹. **d**, Drain current as a function of drain-source voltage for a F₁₆CuPc n-channel TFT (gate-source voltage in steps of 0.5 V). **e**, Drain current and gate current as a function of gate-source voltage for a F₁₆CuPc n-channel TFT (drain-source voltage: 2 V). The mobility is 0.01 cm² V⁻¹ s⁻¹. Hysteresis and gate leakage are very small for both types of TFT.

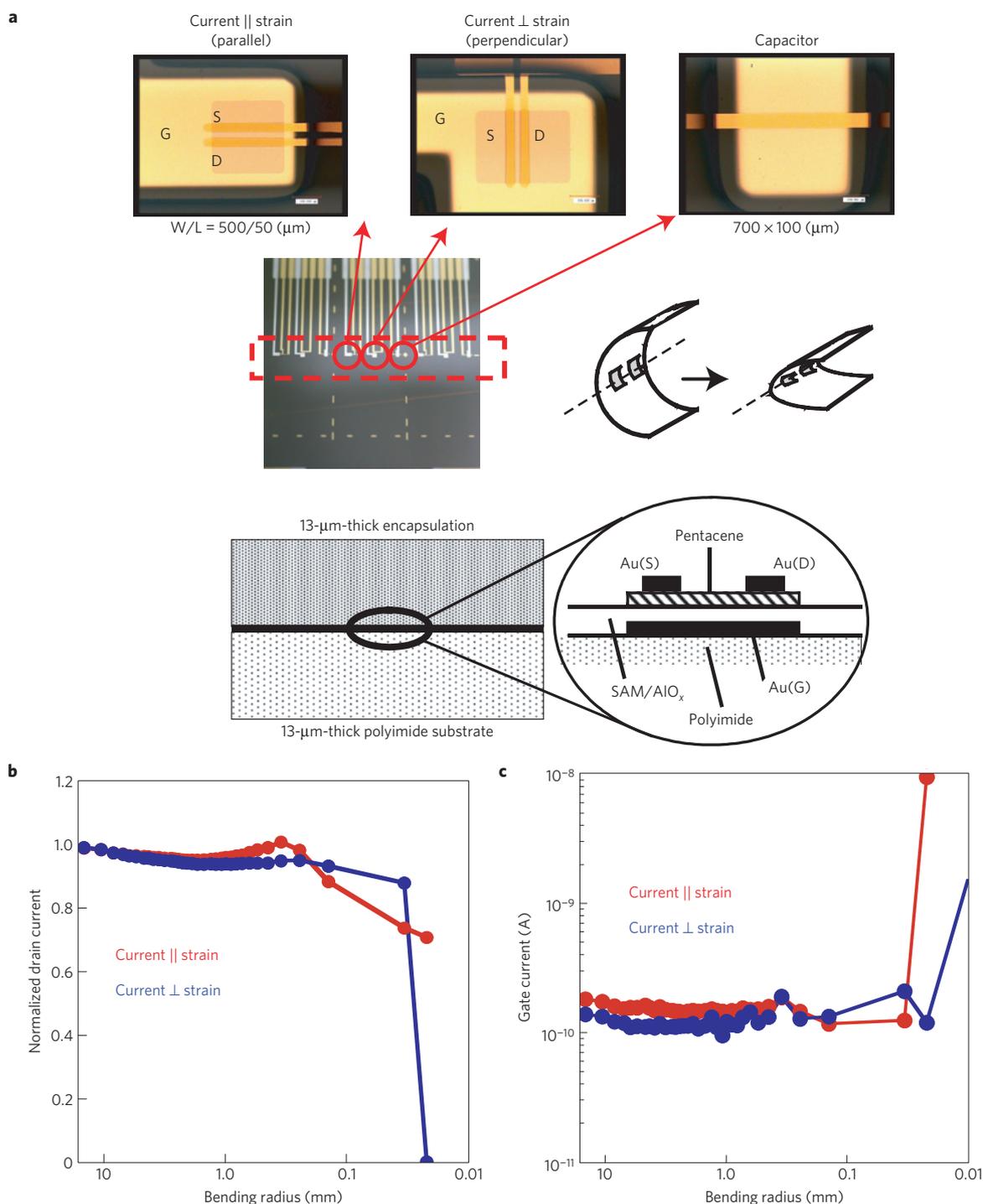


Figure 3 | Bending tests on ultraflexible organic TFTs. **a**, Photographs of a TFT in which the drain current flows parallel to the bending-induced strain, of a TFT in which the current flows perpendicularly to the strain and of an Al/AIO_x/SAM/Au capacitor that allows measurement of the gate dielectric capacitance during bending. The TFTs have a channel length of 50 μm and a channel width of 500 μm. The capacitor has an area of 700 × 100 μm². Bending was carried out using a custom-built precision bending apparatus. **b**, Measured drain currents of two pentacene TFTs (red: strain parallel to the drain current; blue: strain perpendicular to the drain current) as a function of bending radius during inward bending, normalized to the initial drain current measured in the flat state. **c**, Gate currents of the same TFTs measured during inward bending. Drain-source voltage: −2 V; gate-source voltage: −2.5 V.

means they are not subjected to strain and thus are highly stable during sharp bending^{9,10}.

Figure 1c shows a high-resolution cross-sectional electron microscopy image of a completed transistor. The specimen was prepared using a focused ion beam (FB-2100, Hitachi High-Technologies) and imaged by transmission electron microscopy (HF-3300 Cold-FE TEM, 300 kV, Hitachi High-Technologies). The

Al gate as well as the 4-nm-thick AIO_x layer and the 2-nm-thick SAM of the hybrid gate dielectric is clearly resolved in the transmission electron micrograph. Owing to the small surface roughness provided by the 500-nm-thick polyimide planarization layer, the individual TFT layers are very smooth. When the planarization layer is omitted, the TFT layers are much rougher (see Supplementary Fig. S2b).

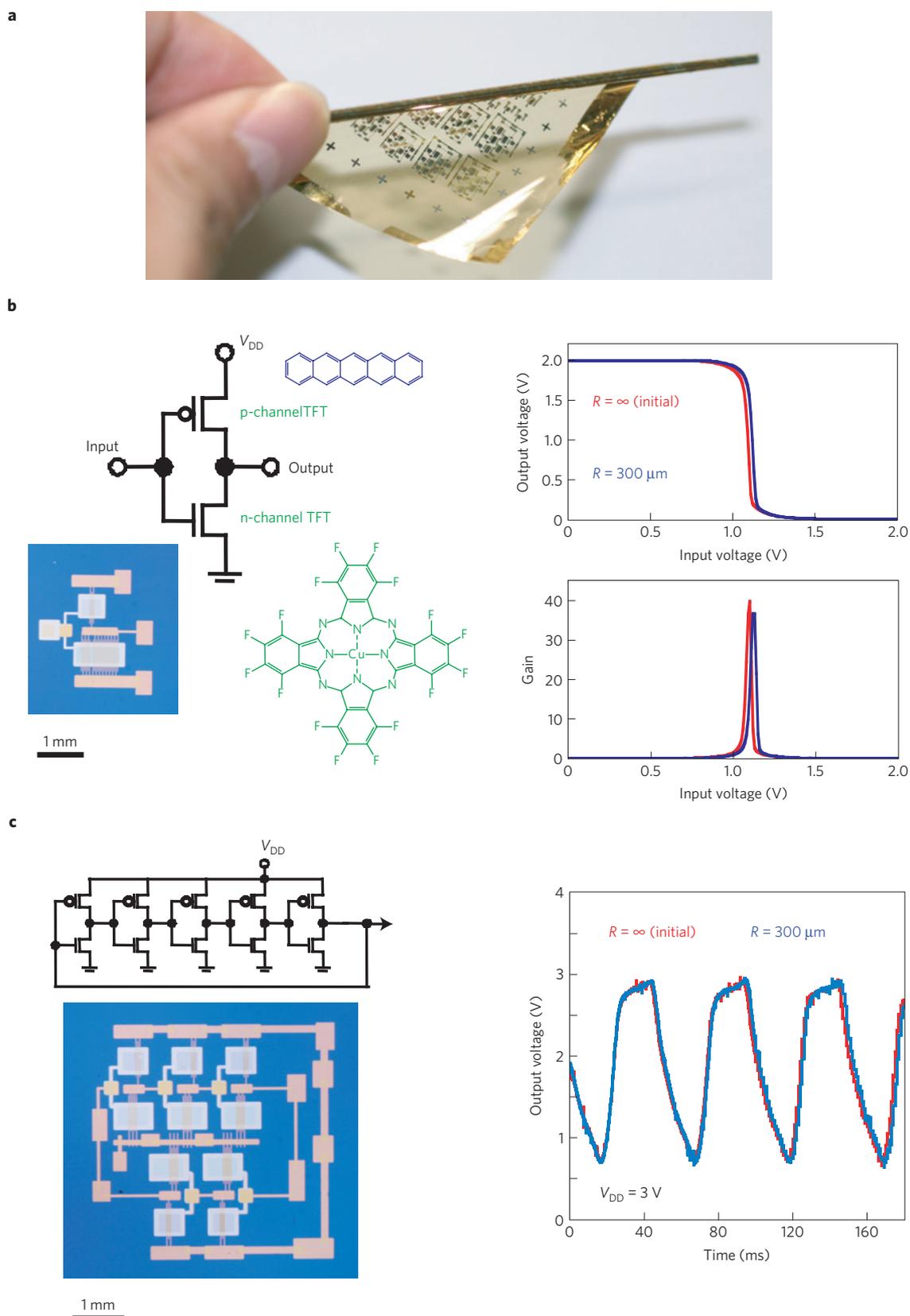


Figure 4 | Ultraflexible organic circuits. **a**, Photograph of a polyimide substrate with functional organic TFTs and circuits wrapped around a cylinder with a radius of 300 μm . **b**, Circuit diagram, photograph and electrical transfer characteristics of a complementary inverter (composed of a pentacene p-channel TFT and a F_{16}CuPc n-channel TFT) operated with a supply voltage (V_{DD}) of 2 V. The TFTs have a channel length of 20 μm . The transfer characteristics were measured in the flat state and while the inverter was bent into a radius of 300 μm . There is no discernible change in characteristics during bending. **c**, Circuit diagram, photograph and output signals of a five-stage complementary ring oscillator operated with a supply voltage (V_{DD}) of 3 V in the flat state and while bent into a radius of 300 μm . The circuit oscillates with a signal delay per stage of 4.5 ms, both in the flat state and during tight bending.

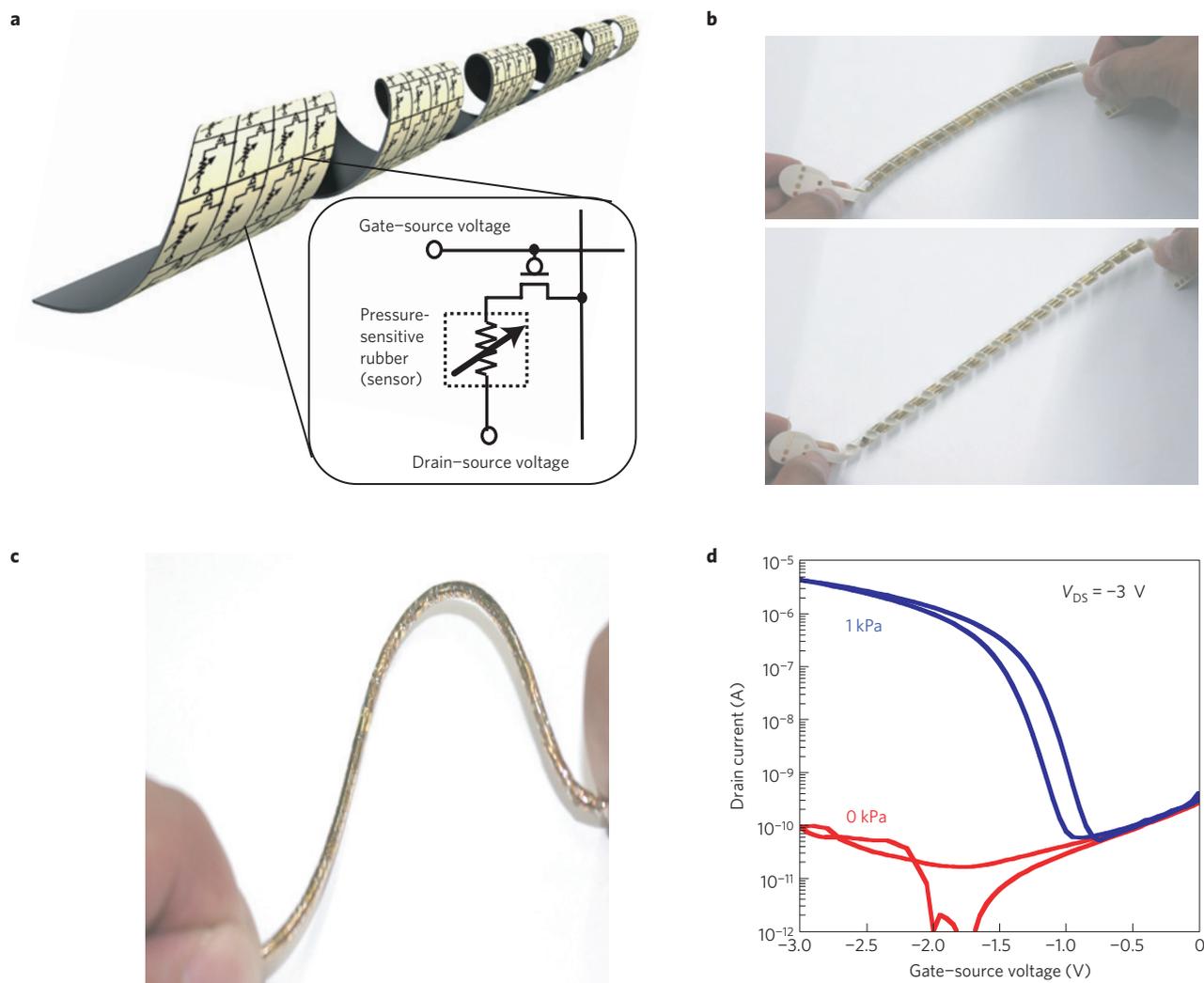


Figure 5 | Tightly wound pressure-sensor helix. **a**, Circuit diagram of an ultraflexible active-matrix pressure-sensor array in the shape of a tightly wound helix. **b**, Photographs of a TFT array fabricated on a shape-memory polymer film (NIPPON MEKTRON, Japan) and permanently transformed into a helix. Even when the helix is stretched by 50% the TFTs remain functional. **c**, Photograph of a tightly wound active-matrix sensor array for a catheter that measures the spatial distribution of pressure. The array was fabricated by laminating three sheets: (1) a foldable 4×36 array of pentacene TFTs, (2) a pressure-sensitive rubber sheet and (3) a $12.5\text{-}\mu\text{m}$ -thick polyimide sheet with a Au counter electrode. **d**, Transfer characteristics of an individual sensor cell measured at two different pressures. (Note that applying pressure to the pressure-sensitive rubber sheet creates a conducting path between the source of the TFT and the counter electrode. Thus, the potential of -3 V is present on the source of the TFT only when pressure is applied, allowing the array to measure the spatial distribution of pressure.)

Despite the small gate dielectric thickness (6 nm), the leakage currents through the dielectric are below 10^{-5} A cm^{-2} at an applied voltage of 3 V (which corresponds to an electric field of 5 MV cm^{-1} ; see Fig. 2a). The current–voltage characteristics of a pentacene p-channel and a F_{16}CuPc n-channel TFT fabricated on a substrate with a 500-nm -thick polyimide planarization layer are shown in Fig. 2b–e. The mobilities ($0.5\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ for pentacene, $0.01\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ for F_{16}CuPc), subthreshold swings and on/off ratios are essentially identical to those of TFTs on glass²⁵, and the hysteresis in the transfer curves (Fig. 2c,e) is negligible. The electrical characteristics are stable during exposure to air for six months, which is a direct result of the excellent gas barrier characteristics of the $13\text{-}\mu\text{m}$ -thick parylene/Au/parylene passivation stack²⁷.

To confirm the critical role of the 500-nm -thick polyimide planarization layer for achieving good device performance, we have also fabricated TFTs on a polyimide substrate without a planarization layer. The lack of a planarization layer leads to a much greater surface roughness of the Al gate electrodes and the AlO_x/SAM gate dielectric (see Supplementary Figs S1 and S2b), so

the pentacene films in these transistors are characterized by much smaller field-effect mobility ($0.01\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$) and substantial hysteresis (see Supplementary Fig. S3).

The bending stability of the transistors was evaluated by bending the substrate into a radius as small as $100\text{ }\mu\text{m}$ along an axis running exactly through the TFTs, with the direction of the bending-induced strain aligned either parallel or perpendicularly to the direction of the current flow from drain to source (see Fig. 3a). Bending was carried out using a custom-built bending apparatus, and the electrical characteristics were measured while the devices were being bent (see Supplementary Fig. S4). Figure 3b shows the measured drain currents of two pentacene TFTs (one bent in the parallel direction and one bent in the perpendicular direction) as a function of bending radius, normalized to the drain current measured in the flat state. Figure 3c shows the gate currents of the same TFTs for each bending radius. Supplementary Fig. S5 shows the capacitance of an $\text{Al}/\text{AlO}_x/\text{SAM}/\text{Au}$ capacitor as a function of bending radius. As can be seen, the electrical characteristics of the TFTs are stable for bending radii down to $100\text{ }\mu\text{m}$, which is by far the smallest bending

radius reported for an organic (or inorganic) field-effect transistor. Comparing Fig. 3b and c suggests that the damage that occurs when the bending radius is decreased below 100 μm originates in the gate dielectric of the transistors.

For comparison, we have also carried out bending tests on pentacene TFTs fabricated on thicker polyimide substrates (thickness 75 μm instead of 12.5 μm) and fabricated without the 13- μm -thick parylene/Au/parylene encapsulation stack. In this case, the TFTs are not located in the neutral strain position and thus are fully exposed to the bending-induced mechanical strain. The measurement results, shown in Supplementary Fig. S6, indicate that in these TFTs damage occurs at a much larger bending radius (~ 3.5 mm), which confirms the benefit of employing an encapsulation stack that places the TFTs in the neutral strain position.

We have also evaluated the bending stability of organic complementary inverters and ring oscillators fabricated along with the pentacene p-channel and F_{16}CuPc n-channel TFTs on the flexible polyimide substrates. There are only very few reports of flexible organic complementary ring oscillators, and in all of them the circuits either required large operating voltages^{28,29} or had to be operated in an inert environment owing to the use of semiconductors with air degradation³⁰. Figure 4a shows a photograph of a 12.5- μm -thick polyimide substrate (complete with a 500-nm-thick planarization layer, organic devices and circuits, and a 13- μm -thick encapsulation stack) tightly wrapped around a cylinder with a radius of 300 μm . Figure 4b shows the circuit diagram, a photograph and the static transfer characteristics of a complementary inverter (composed of a pentacene p-channel TFT and a F_{16}CuPc n-channel TFT) fabricated on this substrate. The transfer characteristics were measured in ambient air in the flat state and while the substrate was bent into a radius of 300 μm . There is no discernible change in the electrical characteristics during bending. The schematic, a photograph and the output signal of a five-stage complementary ring oscillator operated with a supply voltage of 3 V are shown in Fig. 4c. The signal delay at this supply voltage is 4.5 ms per stage, similar to circuits made on glass²⁵. The signal delays in the range of a few milliseconds are fast enough for certain sensor applications such as artificial skins², as highlighted in Figs 1 and 5. Furthermore, as can be seen, the circuit continues to oscillate in the bent state, and the output signal is unaffected by bending of the ring oscillator into a radius of 300 μm . The circuits shown in Fig. 4 were fabricated on a polyimide substrate and therefore relax back into the flat state when released from the cylinder around which they had been wrapped.

An interesting option we have also explored is the fabrication of circuits on a shape-memory polymer film³¹. The substrate is initially in its flat state to facilitate fabrication of high-performance organic devices. After wrapping the substrate around a cylinder into a helix, it is heated to a temperature of about 150 $^{\circ}\text{C}$ and then allowed to cool, which causes the substrate to permanently adapt the helical structure, as shown in Fig. 5b.

To demonstrate a possible application for organic TFTs that operate in the bent state, we have manufactured a thin catheter that measures the spatial distribution of mechanical pressure. The sensor was fabricated by laminating three sheets: a foldable 4×36 array of pentacene TFTs, a pressure-sensitive rubber sheet and a 12.5- μm -thick polyimide sheet with a gold counter electrode. The picture is shown in Fig. 1d and demonstrated in Fig. 5 and Supplementary Fig. S9. The source contacts of all 144 transistors are connected to the rubber sheet, and the counter electrode is in contact with the opposite surface of the rubber sheet. When mechanical pressure is exerted on the catheter, the electrical resistance between the rubber's top and bottom surfaces decreases (see Supplementary Fig. S9). A potential of -3 V applied to the counter electrode is supplied to the TFTs in those positions where pressure is applied,

and thus the spatial distribution of pressure can be obtained by interrogating the TFTs in the active-matrix array.

We have demonstrated that low-voltage organic transistors that are fabricated on planarized polymeric substrate and covered with an encapsulation stack that places the devices into the neutral strain position operate reliably and with excellent performance characteristics while being folded into a bending radius as small as 100 μm . This may open a wide range of opportunities for electronic applications that require a high degree of mechanical flexibility combined with operating voltages accessible by small batteries. As an example we have illustrated a thin catheter that measures the spatial distribution of mechanical pressure using a helical sensor array enabled by ultraflexible organic transistors.

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Author contributions

T. Sekitani and T. Someya designed the concept. T. Sekitani, U.Z., H.K. and T. Someya carried out experimental work, data analysis and wrote the paper. T. Someya supervised the project.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturematerials. Reprints and permissions information is available online at <http://npg.nature.com/reprintsandpermissions>. Correspondence and requests for materials should be addressed to T. Someya.