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Stretchable active-matrix organic light-emitting diode display using printable elastic conductors

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Stretchability will significantly expand the applications scope of electronics, particularly for large-area electronic displays, sensors and actuators. Unlike for conventional devices, stretchable electronics can cover arbitrary surfaces and movable parts. However, a large hurdle is the manufacture of large-area highly stretchable electrical wirings with high conductivity. Here, we describe the manufacture of printable elastic conductors comprising single-walled carbon nanotubes (SWNTs) uniformly dispersed in a fluorinated rubber. Using an ionic liquid and jet-milling, we produce long and fine SWNT bundles that can form well-developed conducting networks in the rubber. Conductivity of more than 100 S cm^{-1} and stretchability of more than 100% are obtained. Making full use of this extraordinary conductivity, we constructed a rubber-like stretchable active-matrix display comprising integrated printed elastic conductors, organic transistors and organic light-emitting diodes. The display could be stretched by 30–50% and spread over a hemisphere without any mechanical or electrical damage.

Recently, new technologies have been emerging in stretchable electronics[1–8], motivating intensive efforts to grant various kinds of surface some form of 'intelligence'[9–17]. The largest obstacle, however, has been the development of stretchable or elastic electrical wiring that is both highly conductive and highly stretchable. Various types of high-conductivity stretchable material have been developed, such as wavy thin metals[1–7], metal-coated net-films[15], graphene films[18] and single-walled carbon nanotubes[19,20] (SWNTs)/fluorinated copolymer composite[8]. These stretchable materials exhibit excellent conductivity and mechanical stretchability by exploiting structures such as waves and nets; the manufacturing process used is vacuum evaporation, photolithographic patterning or mechanical punching and/or cutting. However, because these manufacturing processes are not scalable, it is difficult to apply to large-area electronics. One solution would be the use of direct printing technologies using printable inks that have high conductivity and stretchability, but none has been reported thus far. Our group has reported an elastic conductor comprising SWNT–fluorinated copolymer composite[8]; however, the material was incompatible with printing owing to its low viscosity and tendency for reaggregation before drying and, furthermore, it required an extra rubber coating and mechanical process to improve the elasticity.

We have developed a printable elastic conductor comprising SWNTs uniformly dispersed in a highly elastic fluorinated copolymer rubber by using an imidazolium ion-based ionic liquid[21–23] and a jet-milling process. Taking advantage of a process that can uniformly disperse finer bundles of SWNTs in a rubber matrix without shortening the length of the nanotubes, the SWNT–rubber composite gel becomes increasingly viscous, and consequently this material can be finely patterned using direct printing technologies. Furthermore, a printed elastic conductor requires no extra coating and mechanical process, and can stretch by 118% and has an extraordinarily high conductivity of 102 S cm^{-1}. To the best of our knowledge, this is the highest value reported for a stretchable, printable conductor so far. This printable elastic conductor can be used for stretchable wires and contacts in electrical integrated circuits. We have integrated such printed elastic conductors with organic transistors[24–27] and organic light-emitting diodes[28] (LEDs) to realize a truly rubber-like active-matrix organic LED display. A 16 × 16 grid of transistors was used for driving the display and the effective size of the active matrix was 10 × 10 cm^2. The display could be stretched by 30–50% and spread over a hemisphere without any mechanical or electrical damage. Furthermore, it remained functional even when folded in two or crumpled up, indicating excellent durability.

Stretchable, conductive inks

A schematic representation of the manufacturing process is shown in Supplementary Fig. S1. As a chemically stable and highly conductive dopant, we used super-growth SWNTs of high purity and with a high aspect ratio (>99.98% in purity, >1 mm in length and 3 nm in diameter)[20]. Typically, a mixture of super-growth SWNTs (30 mg), an ionic liquid (1-butyl-3-methylimidazolium bis(trifluoromethanesulphonyl)imide, BMITFSI, 60 mg) and 4-methly-2-pentanone (20 ml) was stirred at 25 °C with a magnetic stirrer (>700 r.p.m.) for 16 h. The resulting swollen

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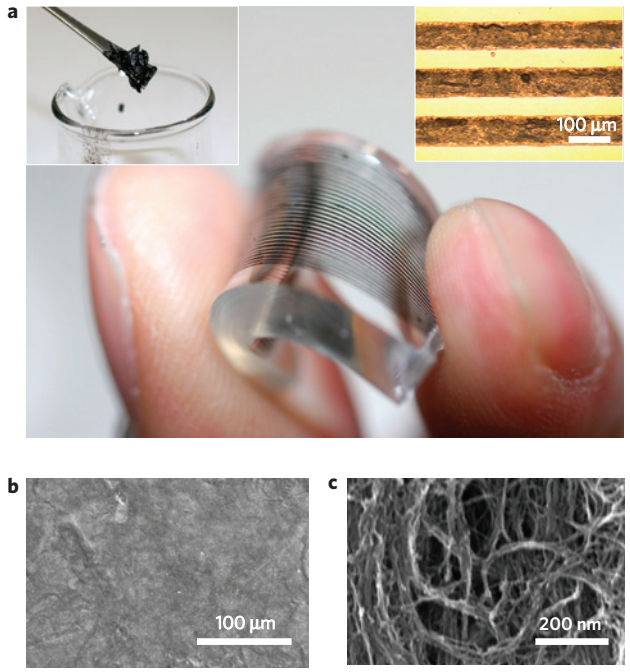

Figure 1 | Printable elastic conductors. a, Printed elastic conductors on a PDMS sheet. Printed elastic conductors patterned by screen printing can be stretched by 100% without electrical or mechanical damage. The insets show SWNTs dispersed in paste and a micrograph of printed elastic conductors with a linewidth of 100 μm. The picture was taken without stretching. **b**, An SEM image of the surface of printed elastic conductors. SWNTs were uniformly dispersed in a fluorinated copolymer matrix (Daikin, Daiel-G912), which is a very soft rubber, using a room-temperature ionic liquid (BMITFSI) and jet-milling. **c**, A magnified SEM image of the elastic conductor. Finer or exfoliated bundles of SWNTs were uniformly dispersed in the rubber, and formed well-developed conducting networks.

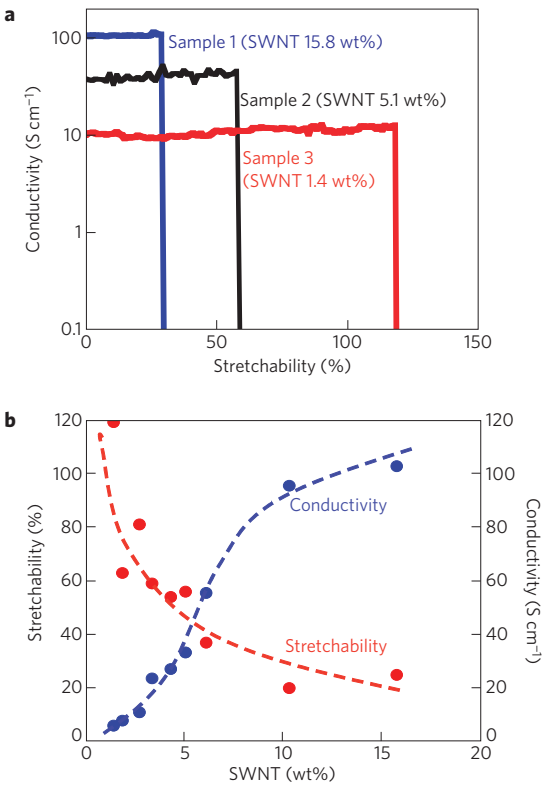

Figure 2 | Electrical and mechanical characteristics of printed elastic conductors. a, Conductivity of printed elastic conductors as a function of stretchability. Typical elastic conductors with 15.8, 5.1 and 1.4 wt% SWNTs are shown. Conductivity increases with SWNT content, whereas stretchability decreases. Furthermore, conductivity does not change with stretching. **b**, Stretchability and conductivity as a function of SWNT content.

gel was processed on a high-pressure jet-milling homogenizer (60 MPa; Nano-jet pal, JN10, Jokoh), giving a black paste-like substance, referred to as bucky gel. To the gel were successively added 4-methly-2-pentanone (80 ml) and a fluorinated copolymer, vinylidenefluoride–tetrafluoroethylene–hexafluoropropylene (50–1,500 mg; Daiel-G912, Daikin; referred to as G912 henceforth), and the mixture was stirred at 25 °C for 16 h, poured onto a glass plate by drop-casting and air dried for 6 h to afford a SWNT–rubber composite gel (referred to as SWNT paste henceforth; Fig. 1a). The paste viscosity was approximately 10 Pa s. Furthermore, when the SWNT paste was air dried for more than 12 h, an elastic conductor was formed.

The SWNT paste was patterned on rubber sheets made of a silicone elastomer (polydimethylsiloxane, PDMS; Sylgard 184, Dow-Corning) using screen printing through shadow masks. The patterned SWNT pastes were air dried for more than 6 h to afford fine SWNT-based elastic conductors with a linewidth of 100 μm (Fig. 1a). The resolution is currently limited by the adhesion between the SWNT paste and the PDMS substrate. The printed elastic conductors can be stretched by approximately 100% without mechanical damage or delamination from the PDMS matrix. Note that the stretchability can be enhanced when printed elastic conductors are sandwiched between rubber substrates because encapsulation prohibits the elastic conductors from delamination. More detailed information on the printability can be found in Supplementary Information.

Figure 1b shows an image of the surface of printed elastic conductors taken using a scanning electron microscope (SEM, S4300; Hitachi). The smooth surface confirms that the bundles of SWNTs were made much finer through the process using ionic liquid and jet-milling. In fact, the SWNTs were significantly untangled or exfoliated after the jet-milling process (see Supplementary Fig. S2). We clearly demonstrated that the process including an ionic liquid and jet-milling significantly untangled or exfoliated the bundle of SWNTs, and formed well-developed nanotube networks in a rubber, as shown in Fig. 1c.

The electrical and mechanical properties of the printed elastic conductors under tensile stress were investigated using a precision mechanical stretching system (Autograph AG-X; Shimazu). Figure 2a shows a plot of the conductivity as a function of stretchability as the SWNT content was changed from 1.4 to 15.8 wt%. A SWNT content of 15.8 wt% produced an extraordinarily high conductivity of 102 S cm^{-1} and stretchability of 29%. In contrast, the conductors with 1.4 wt% SWNTs exhibited an extraordinarily high stretchability of 118% and conductivity of 9.7 S cm^{-1}. Importantly, the elastic conductors did not show significant changes in conductivity or any sign of mechanical damage when fully stretched. For comparison, a conventional conducting rubber containing carbon particles was also tested. Although the stretchability of the conventional conducting rubber exceeded 150%, the conductivity was as low as 0.1 S cm^{-1}, which is insufficient for electronic circuit applications. In fact, low conductivity of electrical wirings may result in a low-speed response, large voltage depression and large power consumption.

There are three important factors determining the conductivity and stretchability of the printable elastic conductors. The first is the mixing ratio of SWNTs, the ionic liquid (BMITFSI) and the fluorinated copolymer matrix (G912). The SWNT content was changed from 1.4 to 15.8 wt% (Fig. 2b) while the BMITFSI content

was maintained at twice the SWNT content. When the SWNT content was greater than 6 wt%, the conductivity was higher than 50 S cm^{-1}, and the stretchability was less than 40%. In contrast, at less than 6 wt% of SWNTs, the stretchability was higher than 50%, and the conductivity was less than 40 S cm^{-1}. Importantly, we found that the elastic conductors have very smooth surfaces and high film quality over the wide range of SWNT contents. This is due to the compatibility of the constituent materials, as described later. Furthermore, Fig. 2b clearly indicates that the important parameters—conductivity and stretchability—can be systematically controlled and tailored to the demands of electronic applications, although there exists a trade-off between the two parameters. One method to achieve a better balance between conductivity and stretchability would be the use of mechanical punching to form a net-shaped structure, although this manufacturing process is more complex.

The second factor is the method used to exfoliate the bundles of SWNTs and make them finer so that they are uniformly dispersed in the elastic fluorinated copolymer matrix. Typical dispersing techniques such as ultrasonication, grinder-milling and ball-milling can make the bundles finer, but at the same time, they make the SWNTs shorter, thus reducing the conductivity. We used a jet-milling high-pressure homogenizer that breaks materials into fragments with a high-pressure jet. It is particularly worth noting that this process enables us to uniformly disperse finer bundles of SWNTs into a rubber matrix without shortening the length of the nanotubes during the process. The resulting longer and finer bundles of SWNTs can form well-developed conducting networks in rubbers, as shown in Fig. 1c, thus leading to higher conductivity and stretchability, simultaneously. The formation of SWNT networks in rubbers also realizes printability of this material because the SWNT paste becomes increasingly viscous (more than 10 Pa s), which is very suitable for high-definition screen printing.

Besides the mixing ratio and dispersing process, the compatibility of the materials—SWNTs, ionic liquid and copolymers—is also very essential. We carried out material compatibility tests to determine the combination of elastic polymers and ionic liquid that would be compatible with each other (see Supplementary Fig. S3). We examined three different fluorinated copolymers: G912 and vinylidene fluoride/hexafluoropropylene copolymers with composition ratios of 0.78:0.22 (Daiel-G801, Mw = 150,000, Daikin; referred to as G801 henceforth) and 0.88:0.12 (KYNAR-FLEX2801, Mw = 30,000, Arkema; referred to as Kyner henceforth). G912 is very soft and elastic, but this often results in separation of the G912 and ionic liquid phases. We used BMITFSI, which is compatible with several fluorinated copolymers. In fact, when G912 and BMITFSI were used, the resulting SWNT rubber films were very smooth, flat and uniform (Fig. 1b and Supplementary Fig. S2c), and hence exhibited high conductivity and stretchability. In contrast, when Kyner (which is a hard resin) was used, the resulting films exhibited a conductivity of only 21 S cm^{-1} and were very fragile and not stretchable. When G801 (which is also a soft rubber) was used, the conductors exhibited a stretchability of 87% and conductivity of 3 S cm^{-1} with 1.8 wt% SWNTs, and a stretchability of 24% and conductivity of 22 S cm^{-1} at 15.7 wt% SWNTs. The results of the compatibility tests convinced us that combination of G912 and BMITFSI is the best from the points of view of both the electrical and mechanical performance in the process with jet-milling.

We also investigated the durability under stretching cycles. Although durability depends on stretchability, the elastic conductors with 1.4 wt% SWNTs, 2.8 wt% BMITFSI and 95.8 wt% G912 did not show changes in conductivity after 1,000 cycles of stretching by 70%.

Carbon-nanotube-based conducting materials or carbon-nanotube/conducting-polymer composites have also been produced by other groups[29–32]. In particular, Spinks *et al.* and Shim *et al.* reported composites with excellent conductivities of the order of 100 S cm^{-1} by taking advantage of highly conductive polymers[30,31]. Recently, Mukai *et al.* demonstrated the fabrication of SWNT films comprising millimetre-long SWNTs and an ionic liquid, which also exhibited a high conductivity of 169 S cm^{-1} (ref. 32). However, these materials reported so far cannot be stretched.

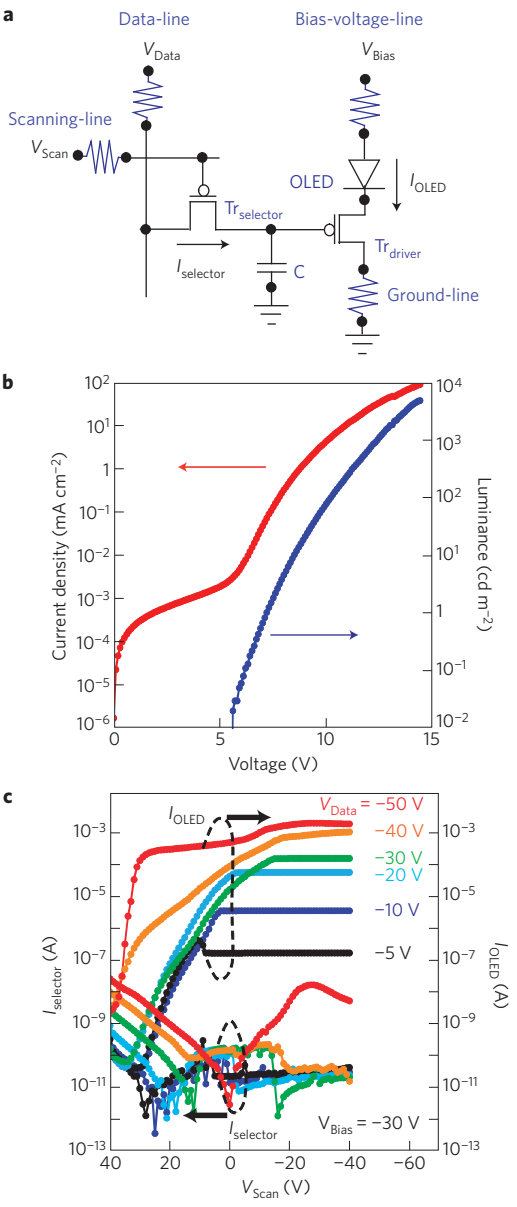

Figure 3 | Stretchable display cells comprising an organic LED and a 2T1C driving cell. a, Circuit diagram of one stretchable display cell comprising an organic LED and a 2T1C driving cell. $I_{selector}$ and I_{OLED} represent currents passing through the selector transistor $Tr_{selector}$ and through the organic LED, respectively. Display cells are interconnected with each other through printed elastic conductors that work as scanning-, data-, bias-voltage- and ground-lines. V_{Scan}, V_{Data} and V_{Bias} represent voltages applied to scanning-, data- and bias-voltage-lines, respectively. The capacitance of the circuit (C) is 215 pF. The resistor symbols represent printed elastic conductors. **b**, Current density and luminance of a typical stand-alone organic LED as a function of bias voltage. Experiments were carried out in ambient air. **c**, $I_{selector}$ and I_{OLED} as a function of V_{Scan} when V_{Bias} is constant at −30 V and V_{Data} is varied from −5 to −50 V.

a

V_{Scan}	+40 V	−5 V	−10 V	−30 V	−40 V
Cu wiring	6.7 nA, 0.0 cd m^{-2}	1.7 μA, 0.3 cd m^{-2}	35 μA, 6.1 cd m^{-2}	381 μA, 88.3 cd m^{-2}	1.3 mA, 408 cd m^{-2}
Elastic conductor	7.0 nA, 0.0 cd m^{-2}	1.6 μA, 0.28 cd m^{-2}	31.6 μA, 5.5 cd m^{-2}	343 μA, 77.2 cd m^{-2}	1.2 mA, 364 cd m^{-2}
Conventional conducting rubber	2.6 nA, 0.0 cd m^{-2}	280 nA, 0.0 cd m^{-2}	6.6 μA, 1.2 cd m^{-2}	19.4 μA, 3.4 cd m^{-2}	40 μA, 7.1 cd m^{-2}

b

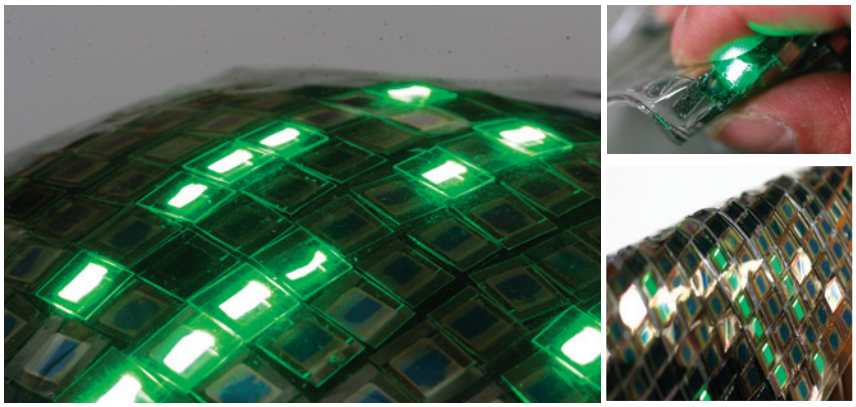

Figure 4 | Luminance of a stretchable display. a, Luminance of a display cell as a function of V_{Scan}. Luminance was investigated for three configurations of display cell: 1, an organic LED integrated with a 2T1C driving cell and electrical wirings made of 1- μm-thick Cu; 2, an organic LED integrated with a 2T1C driving cell and electrical wirings made of printed elastic conductors (conductivity, 50 S cm^{-1}; length, 100 mm; linewidth, 500 μm; thickness, 100 μm); 3, an organic LED integrated with a 2T1C driving cell and electrical wirings made of conventional conducting rubber (conductivity, 0.1 S cm^{-1}; length, 100 mm; linewidth, 1 mm; thickness, 100 μm). V_{Bias} and V_{Data} were −30 V and −40 V, respectively. I_{OLED} and luminance are also shown. The effective size of the light-emitting area was 3 × 4 mm^2. **b**, A demonstration of a stretchable display that can be spread over arbitrary curved surfaces. The stretchable display is functional even when folded in two or crumpled, indicating excellent durability.

We previously demonstrated the SWNTs were uniformly dispersed in a fluorinated copolymer rubber matrix by grinding with an imidazolium ion-based ionic liquid and the fabrication of a SWNT–rubber composite with a conductivity of 57 S cm^{-1} and a stretchability of 134% (ref. 8). However, the previous composite could not be printed owing to its low viscosity and tendency for reaggregation before drying and, furthermore, it required an extra rubber coating and mechanical process to improve the elasticity. In this work, we found that through the process, the SWNT paste becomes increasingly viscous (more than 10 Pa s), and consequently this material can be finely patterned using direct printing technologies. The spatial resolution of the printing is approximately 100 μm on PDMS substrate (see Supplementary Information). Furthermore, the new material requires no extra coating and mechanical process, and exhibits the highest conductivity reported for a stretchable conductor so far. It can be stretched by more than 100% and, more importantly, it has an extraordinarily high conductivity of 102 S cm^{-1}, which changes negligibly little with stretching (Fig. 2 and Supplementary Fig. S4). Importantly, the performance can be easily controlled by changing the mixing ratio. This is mainly due to the use of jet-milling as well as ionic liquid that could prevent unfavourable aggregation of nanotube bundles, thus resulting in the uniform dispersion of finer and longer bundles of SWNTs in the very elastic rubber matrix. The ionic liquid has the important role of plasticizing agents in the elastic conductors. This enables the SWNT content to be increased up to 16 wt% without reducing the mechanical flexibility or softness of the copolymer, G912 (see Supplementary Fig. S5).

Stretchable active-matrix display

Taking full advantage of printable elastic conductors with excellent conductivity and stretchability, we manufactured a rubber-like stretchable active-matrix organic LED display comprising 16 × 16 driving cells with two organic transistors and one capacitor (2T1C), organic LEDs, elastic conductors and pastes. The manufacturing processes for the organic 2T1C driving cells and organic LEDs can be found in Supplementary Information. A circuit diagram of one display cell with a 2T1C driving cell and an organic LED is shown in Fig. 3a. Each driving cell comprises a selector transistor ($Tr_{selector}$), a driver transistor (Tr_{driver}), a capacitor (C) and printed elastic conductors: selector transistors enable addressing of each organic LED for light emission, and the driver transistors control the luminance. Capacitors have a major role for stabilizing light emission with high-frequency operation (see Supplementary Fig. S9). After manufacturing the organic LEDs and organic driving cells independently on separate

PDMS sheets, the two sheets were laminated to construct a rubber-like stretchable display.

An active matrix comprising a sheet of 16 × 16 driving cells was fabricated on a flexible polyimide film using vacuum evaporation. These cells were mechanically processed to isolate and uniformly distribute them on a stretchable PDMS rubber sheet. The periodicity is 5 mm. Each isolated driving cell on the PDMS sheet was electrically connected using printed elastic conductors as scanning-, data-, bias-voltage- and ground-lines (Fig. 3a). The organic transistors for both $Tr_{selector}$ and Tr_{driver} had a 50-nm-thick pentacene channel and 250-nm-thick parylene gate dielectric layers; the channel length and width were 20 μm and 6 mm, respectively. The manufactured organic transistors exhibited a mobility of 0.3 $cm^2 V^{-1} s^{-1}$ and an on/off ratio of 10^6. Details of the transistor performance characteristics can be found in Supplementary Information (see Supplementary Fig. S7).

Figure 3b shows the typical current density and luminance of an organic LED as a function of the applied voltage. From 0 to 15 V, the luminance increased to more than 5,000 cd m^{-2}, indicating excellent light-emission characteristics. An encapsulation layer made of plastic barrier films acted as an excellent gas barrier so that the organic LEDs were not degraded even when operating in ambient air; the lifetime in air is at least three months.

The electrical characteristics of a 2T1C driving cell with printed elastic conductors were also investigated. Figure 3c shows plots of the currents through a selector transistor ($I_{selector}$) and a driver transistor and organic LEDs (I_{OLED}) as functions of the voltage from scanning-lines (V_{Scan}) when the voltages of the data-lines (V_{Data}) were gradually changed from −5 to −50 V; the bias voltage of the organic LEDs (V_{Bias}) was constant at −30 V. $I_{selector}$ is very small because the drain electrodes of the selector transistors were not grounded, whereas the selector transistors can supply the voltage V_{Data} to the gate electrodes of the driver transistors. Therefore, when one selector transistor is addressed, the connected driver transistor is activated. In fact, I_{OLED} increased from several picoamperes or nanoamperes to several milliamperes with increasing V_{Scan} and V_{Data}. The on/off ratio of I_{OLED} was found to be 10^6 when controlling V_{Scan}. This result clearly indicates that driving cells with the 2T1C structure can operate organic LEDs even though large-area organic LEDs require large currents of the order of several hundred microamperes to several milliamperes for light emission.

Figure 4a shows the light-emitting properties of the display cells at different V_{Scan} when V_{Bias} and V_{Data} were −30 V and −40 V, respectively. The light-emitting properties of an organic LED integrated with a driving cell and Cu wirings, an organic LED integrated with a driving cell and printed elastic conductors (50 S cm^{-1}) and an organic LED integrated with a driving cell and commercially available conventional conducting rubbers (0.1 S cm^{-1}) are shown, for comparison. The data in Fig. 4a were obtained from a single organic LED and a single organic driving cell to evaluate the performance of the elastic wirings. The experimental procedure can be seen in Supplementary Information. When printed elastic conductors were used, I_{OLED} decreased slightly by 8% to 1.2 mA, resulting in a high luminance value of 364 cd m^{-2}, which is sufficient for display applications. In contrast, when electrical wirings were formed with conventional conducting rubbers (0.1 S cm^{-1}), I_{OLED} decreased by 97% to 40 μA, producing a luminance of only 7.1 cd m^{-2}. This indicates that printed elastic conductors with high conductivity are crucial for stretchable display applications.

By integrating a driving-cell active matrix and organic LEDs using printed elastic conductors, we manufactured a 16 × 16-pixel stretchable active-matrix organic LED display. Figure 4b shows a demonstration of the display, which can be spread over an arbitrary curved surface. Bending or crumpling caused no mechanical or electrical damage because of the excellent conductivity and mechanical durability of the printed elastic conductors, organic transistors and organic LEDs manufactured on the PDMS rubber.

Miniaturization of display cells for a higher resolution is, of course, one of the most important requirements. We successfully patterned printable elastic conductors with linewidths of 100 μm using screen printing. However, screen printing can be used to pattern pastes to resolutions of 10 μm. Organic LEDs can be manufactured using printing technologies[33], and furthermore, printing technologies of organic semiconductors and electrodes with resolutions of 1 μm have been reported[34–36].

The stretchable materials and integration technology used in this study can also be used to create other types of functional electronics such as a rubber-like electrical artificial skin with an integrated, stretchable active matrix and pressure sensors[14,15] or a rubber-like mechanical actuation system with an integrated, stretchable active matrix and actuators that can create the sensation of touch electrically[16,17]. The combination of stretchable sensors, actuators and displays can be used to create real, tangible displays and user-friendly human–machine interfaces on all kinds of surface. In this manner, the printable elastic conductor developed in this work enables the construction of electronic integrated circuits that can be spread over any surface, including arbitrary curved surfaces and movable parts. This would significantly expand the areas where electronics can be used. This is an important step towards the development of the infrastructure for the imminent era of ambient electronics, in which a multitude of electronic devices such as ambient sensors, actuators and display networks function as user-friendly human–machine interfaces that can be used in daily life to enhance security, safety and convenience.

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Author contributions

T.Se. and T.So. planned the project and data analysis. T.Se., H.N., H.M, T.F., T.A., K.H. and T.So carried out experimental work. (T.Se., T.F., T.A., T.So.: elastic conductors; T.Se. and T.So.: organic transistors; H.N. and H.M.: organic LEDs; K.H.: carbon nanotubes.)

Additional information

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