Printed organic transistors: Toward ambient electronics

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Abstract

In the forthcoming ambient electronics era, multiple electronic objects are scattered on walls, ceilings or in imaginative locations and interact each other to enhance safety, security and convenience. For implementation of many electronic objects in our daily life, large-area, flexible devices, which would be printed on plastic sheet, cloth, and/or paper, are expected to play an important role. In this paper, we describe recent progress and future prospects of flexible, large-area electronics based on printed organic transistors.

Introduction

In view of the tremendous technical challenges for realizing next-generation information technology, organic semiconductors have attracted significant attention since the emerging electronics based on them have features that are complimentary to main stream electronics based on silicon. Thanks to the recent advent of organic transistors (FETs) [1-5], the emergence of a new class of electronics makes full use of the unique features of organic semiconductors, such as the ultralow cost, low weight, and flexibility, is becoming more realistic. With this background, our group discerned that large-area circuits could be easily fabricated using organic transistors, which are essential for certain applications, and has developed large-area sensors and actuators using organic transistors. More accurately, we have integrated various types of sheet-type sensors and sheet-type actuators with organic transistors on plastic films and have demonstrated the world's first electronic artificial skin (E-skin) [6-8], a sheet-type scanner [9], a sheet-type Braille display [10], a wireless power transmission sheet [11], a communication sheet [12], an ultrasonic proximity sensor sheet [13], stretchable displays [14], and many other sheet-type devices (Fig. 1).

In the forthcoming ambient electronics era, multiple electronic objects are scattered on walls, ceilings or in imaginative locations and interact each other to enhance safety, security and convenience. For implementation of many electronic objects in our daily life, large-area sheet-type devices are needed and organic transistors are expected to play an important role.

In this paper, we will report recent progress and future prospects of organic transistor-based flexible, large-area sensors and actuators. Moreover, the issues and the future prospect of organic transistors will be addressed from the view point of ambient electronics.

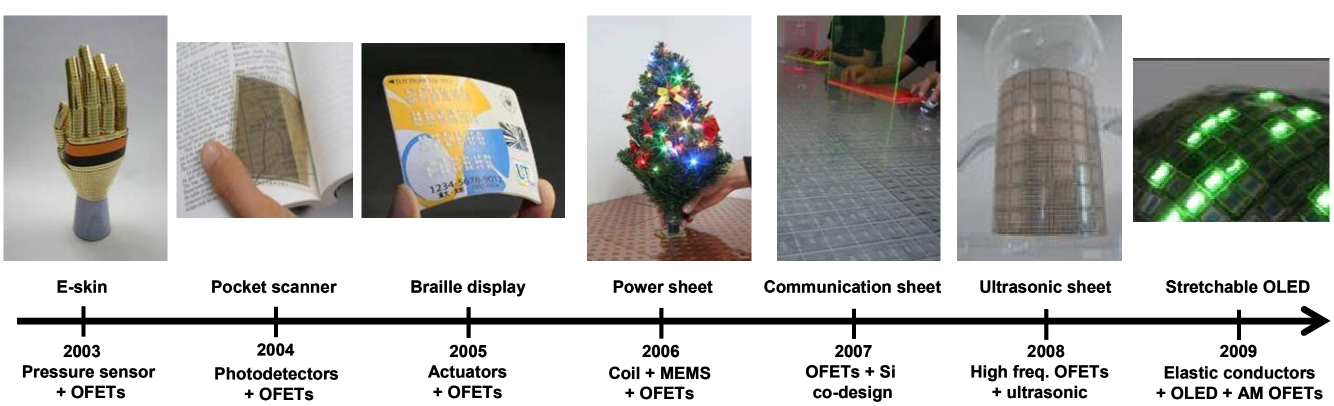

Fig. 1: Large-area, flexible sensors, actuators, and displays that are realized by organic field-effect transistors. (From the left) an electronics artificial skin (E-skin), a sheet-type image scanner (pocket scanner), a sheet-type Braille display for blind persons, a wireless power transmission sheet, a sheet-type communication system, a sheet-type ultrasonic system, and a stretchable active matrix OLED display.

Printed organic transistor active matrix

Organic transistors were manufactured using inkjet printing (Ricoh Printing Systems Co., Ltd.; IJP-1), screen printing (Micro-tec Co., Ltd.; MT-550), and vacuum evaporation methods. The base film (substrate) was a polyimide film with a thickness of 75 μm. Ag nanoparticles (Harima Chemical Co., Ltd.; NPS-J-HP) were patterned by inkjet printing with picoliter accuracy and cured at 180 °C to form 300-nm-thick gate electrodes. Epoxy partitions were formed around the gate electrodes by using screen printing. Poly-chloro-para-xylylene (i.e., parylene, Daisan-kasei Co., Ltd.; diX-SR) were coated at room temperature to form 500-nm-thick gate dielectric layers. Ag paste was patterned by screen printing and cured at 150 °C to form source and drain electrodes. A 50-nm-thick pentacene layer was deposited using vacuum evaporation with rotation mechanics through printed shadow masks [15] to form a channel layer. The channel length and width were typically 30 μm and 500 μm, respectively. Finally, a 5-μm-thick layer of poly-chloro-para-xylylene, parylene was uniformly coated to form a passivation layer. The fabrication process is similar to the method reported in ref. [16]. Figure 2 shows a magnified view of organic transistors. Channel length with 30 μm and printed electrode width with 500 μm were clearly formed using state-of-the-art screen printing.

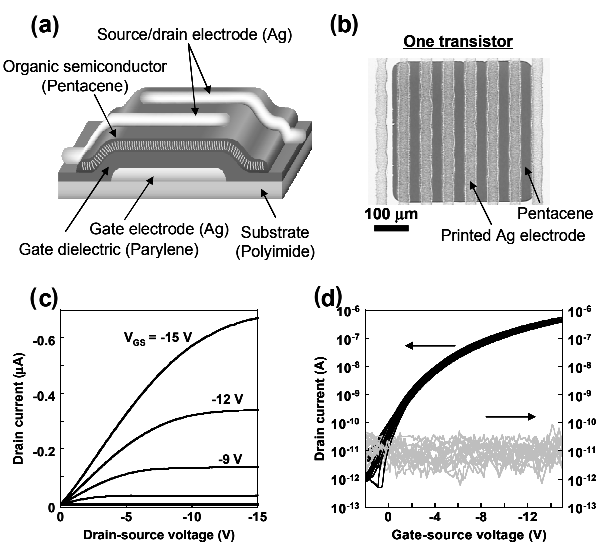

Fig. 2: Organic transistor active matrix using printing technologies: (a) Cross-sectional illustration of one organic transistor. (b) Top view of transistor's channel. Linewidth and space are 30 μm. (c) Drain currents as a function of drain-source voltage. (d) Transfer curve and gate currents obtained from ten transistors.

We characterized pentacene transistors using printed electrodes. The drain current of the transistors was measured as a function of the source-drain voltage (V_{DS}) as shown in Fig. 2c: The gate-source voltage (V_{GS}) was varied from 0 V to −15 V in steps of −3 V. Figure 1f shows the corresponding transfer curves of the same device: V_{GS} is varied from 2 V to −15 V with the application of V_{DS} = −15 V. A hysteresis was very small. Note in Fig. 2d that on-off ratio was about 10^6, showing that leakage current through gate insulator was sufficiently low. The mobility in the saturation regime was 0.1 cm^2/Vs when gate voltage was −15 V. In multilayered structures formed by solution processes, it is important to choose a solvent carefully so that a solvent may not melt a front layer. The present result shows that the choice of the solvents is reasonable.

Printed organic transistor with 1-μm channel length

In order to fabricate very fine top contact organic transistors using printing technology, we employed sub-femtoliter inkjet technology [17,18]. A sub-femtoliter inkjet can pattern very fine Ag nanoparticles (NPS-J-HP) directly on pentacene. The lines having a width of 2 μm can be written uniformly and reproducibly.

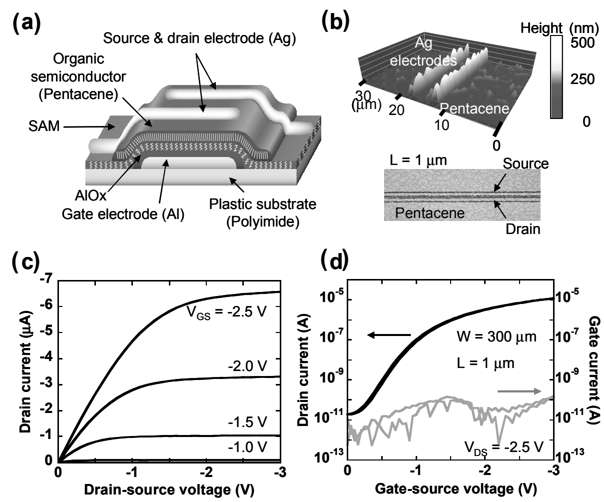

Fig. 3: Organic transistor manufactured using sub-femtoliter inkjet on self-assembled monolayer gate dielectric: (a) An illustration of one organic transistor using a self-assembled monolayer as a gate dielectric. (b) AFM image and magnified picture of printed source/drain electrodes. (c) Drain currents as a function of drain-source voltage. Gate-source voltage (V_{GS}) is changed. (d) Drain and gate currents as a function of gate-source voltage.

Taking full advantages of self-assembled monolayer gate dielectric and sub-femtoliter inkjet, we fabricated very fine printed organic transistors that can operate with very small voltages. A cross-sectional illustration of the organic transistors is shown in Fig.3a. Ag nanoparticles were patterned using sub-femtoliter inkjet to form the source and drain electrodes. The manufactured organic transistor has a channel length of 1 μm and width of 300 μm (Fig. 3b), which is the smallest channel length among printed organic transistors without pre-patterning, pre-treatments, or surface modifications.

The DC characteristics of the manufactured transistors were measured in air using a semiconductor parameter analyzer (Agilent 4156C). Figure 3c shows the drain current (I_{DS}) as a function of the drain-source voltage (V_{DS}). The gate-source voltage (V_{GS}) shifts from 0 to –3 V in steps of –0.5 V. Saturation was clearly observed even at 3 V. Figure 3d shows the corresponding transfer curves under an applied V_{DS} of –1.5 V. The transistors exhibit a high mobility of as high as 0.3 cm^2/Vs, V_{th} of –1.1 V, and their on/off current ratio exceeds 10^5. The present sub-femtoliter inkjet technologies can be applied not only to p-type organic semiconductors such as pentacene but also to n-type organic semiconductors such as F_{16}-CuPc. The transistors exhibit a mobility of 0.02 cm^2/Vs in air, V_{th} of –0.32 V, and an on/off current ratio in excess of 10^4.

Despite the short channel length of 1 μm, the off state current at V_{GS} = 0 V is less than 20 pA, and the on/off current ratio is about 10^6. Despite the small thickness of the room temperature gate dielectric, the maximum gate current at V_{GS} = 3 V is only about 100 pA.

These mobilities are comparable to those obtained with evaporated Au source/drain contacts, suggesting that the mobility is not affected by the 130 °C calcination. This is consistent with our previous work [19] and with our observation that the morphology of the pentacene films (examined by AFM) does not change during sintering at 130 °C.

E-skins

The first application of large-area sensors is a flexible pressure sensor [6-8], which is suitable for electronic artificial skin (E-skin) for the next-generation robots. Although the mobility of organic semiconductors is approximately two or three orders of magnitude less than that of poly- and single-crystalline silicon, the slower speed is tolerable for most applications of large-area sensors. In particular, for the fabrication of E-skins, the integration of pressure sensors and organic peripheral electronics avoids the drawbacks of organic transistors, while taking advantage of their mechanical flexibility, large area, low cost, and relative ease of fabrication.

Figure 4 (a) shows an image of an artificial skin system and (b) its corresponding circuit diagram. A 16 × 16 active matrix of organic transistors, row decoder, and column selector are assembled by a physical cut-and-paste procedure to develop integrated circuits for data readout. Three functional films — an interconnection layer, a pressure-sensitive rubber sheet, and a top electrode for power supply— are then laminated together with the organic ICs. Pressure images were obtained by a flexible active matrix of organic transistors whose mobility is as high as 1.4 cm^2/Vs. These sensors can be bent to a radius of 2 mm, which is sufficiently small for the fabrication of human-sized robot fingers.

However, human skin is more complex than transistor-based imitations demonstrated thus far. It performs certain functions including thermal sensing. Furthermore, without conformability, the application of E-skins to three-dimensional surfaces is impossible. Based on an organic semiconductor, we have developed conformable, flexible, wide-area networks of thermal and pressure sensors. A plastic film with organic transistor-based electronic circuits was

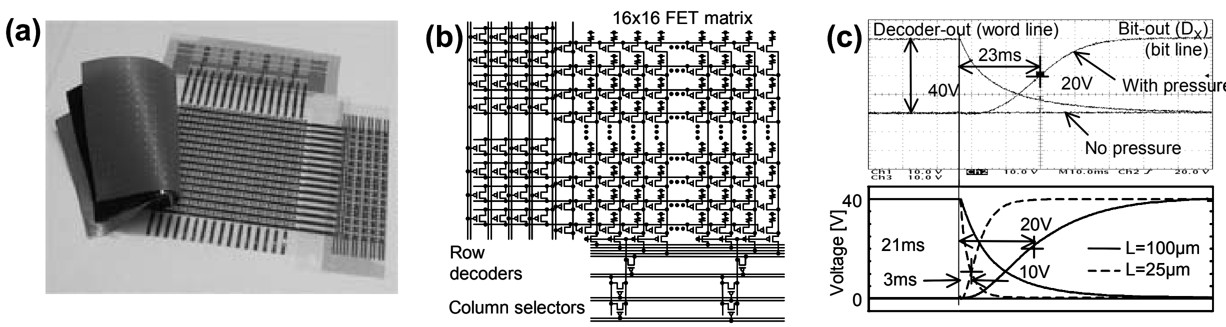

Fig. 4: An artificial skin for robots. (a) A picture of an artificial skin system and (b) a corresponding circuit diagram. A 16×16 organic transistor active matrix, a row decoder and a column selector are manufactured on plastic films separately and then assembled to make integrated circuits for data readout. (c) a measured waveform (upper panel) and a simulation (lower panel). The delay from the row decoder activation to bit-out is measured to be 23ms. The cycle time can be within 30 ms, which means that the time needed to scan the whole 16 × 16 sensor matrix is about 2 seconds.

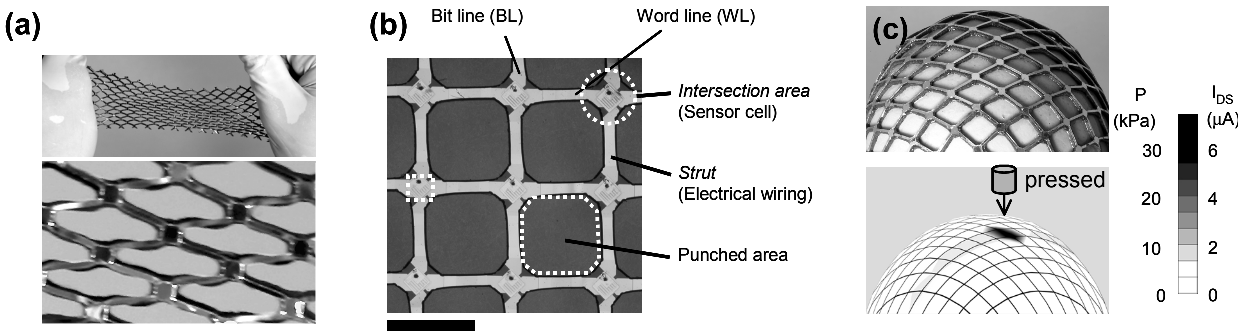

Fig. 5: A conformable network of pressure sensors. (a) A plastic film with organic transistors and pressure sensitive rubber is processed mechanically to form a unique net-shaped structure, which makes a film device extendable by 25%. (b) A picture of the 3×3 sensor cells. Scale is 4 mm. (c) An image (upper panel) of pressure sensor matrix put on an egg is shown together with a spatial distribution of pressure (lower panel). The current of each sensor cell is measured by applying a voltage bias of $V_{DS} = -20$ V and $V_{GS} = -20$ V under the application of local pressure.

processed to form a net-shaped structure that allows the E-skin films to be stretched by 25%. The net-shaped pressure sensor matrix was attached to the surface of an egg and pressure images were successfully obtained in this configuration (Fig. 5).

Moreover, a similar network of thermal sensors was developed using organic semiconductors. A possible implementation of both pressure and thermal sensors on various surfaces is presented. By using laminated sensor networks, the distributions of pressure and temperature are simultaneously obtained.

In order to realize truly rubber-like stretchable e-skins, elastic conductors have been developed [20] using single-walled carbon nanotubes (SWNTs) [21,22] as a conducting dopant. SWNTs were uniformly dispersed as chemically stable dopants in a vinylidene fluoride-hexafluoropropylene copolymer matrix by using an ionic liquid [24] of 1-butyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide, and manufactured SWNT composite films. The measured value of conductivity is as high as 57 S/cm. Such high conductivity is achieved because the content of SWNTs in the conductor can be increased up to 20 wt% without sacrificing mechanical flexibility and softness. The SWNT elastic conductor can be stretched to approximately 134% of its original size without significant mechanical damage. The elastic conductors are used as wirings in large-area stretchable organic FET ICs (Fig. 6). These ICs, which have a high electronic performance, can be stretched by up to 80% without any degradation in their mechanical or electronic properties. This is an important step in the development of ICs that can be used on freely curved surfaces and in smart surfaces.

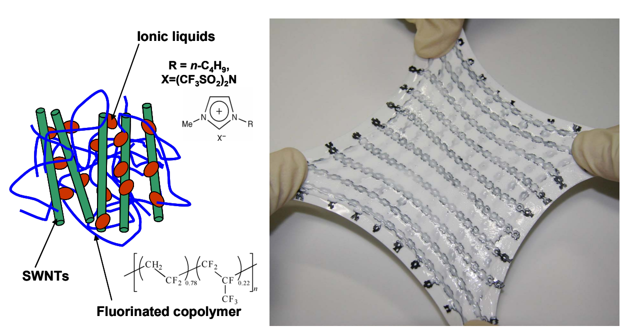

Fig. 6: A truly rubber-like stretchable organic FET active matrix. Elastic conductors, which are fabricated with mixing single-walled carbon nanotubes (SWCNs), ionic liquids, and fluorinated copolymers, are used as wirings to connect contacts of printed organic FETs.

Sheet-type ultrasonic sensors

The second example is an ultrasonic sensor, which realizes the three-dimensional (3D) sensing of the position of people and objects in free space. Although position sensing in free space has commonly been carried out using light, radio, and millimeter waves, ultrasonic imaging can offer complimentary attractive features such as a simple architecture and real-time nondestructive 3D imaging in free space at an ultralow cost [23].

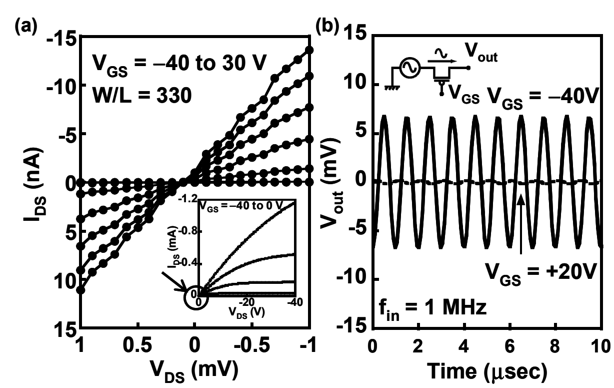

Fig. 7: High-speed printed organic FETs for ultrasonic sheet. (a) Operation of the organic FETs with a small source-drain voltage (V_{DS}). A plot of the source-drain current (I_{DS}) vs. V_{DS}. (b) 1 MHz operation of the organic FETs with a grounded-gate.

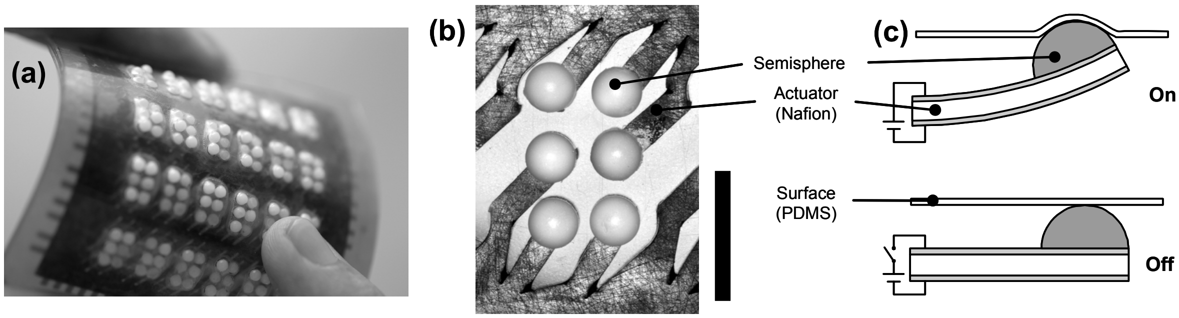

Fig. 8: A Braille sheet display. (a) An image of a pocket *Braille sheet display*. It was manufactured on a plastic film by integrating the active matrix of high-quality organic transistors with a plastic sheet actuator array based on a perfluorinated polymer electrolyte membrane. The device is mechanically flexible, very thin, and lightweight. (b) One character is displayed by a 3 × 2 array of rectangular actuators (4 mm in length and 1 mm in width). A semisphere of radius 0.9 mm is attached to each actuator, which bends and lifts the semisphere. The scale is 4 mm. (c) Principle of Braille motion.

A large-area, flexible, ultrasonic 3D imaging system, by integrating an ultrasonic transducer array sheet with a printed organic FET active matrix [13]. The ultrasonic sensing cell comprises one organic transistor and one ultrasonic transducer. A transistor active matrix sheet and an ultrasonic transducer array sheet are manufactured separately and electrically connected to each other. The system comprises 8 × 8 ultrasonic sensing cells or others, with a printed area of 25 × 25 cm^2. It is mechanically flexible and can be wrapped around a cylindrical bar; this configuration is suitable for obtaining a viewing angle of 360º for a medium-length (~ several meters) proximity robotic skin-like sensor.

The organic FETs are fabricated on a 75-µm-thick polyimide film. The source, drain, and gate electrodes are deposited by an inkjet printing or a vacuum evaporation. A 500-nm-thick polyimide gate dielectric layer is formed by inkjet or spin coating, and a 50-nm-thick pentacene channel layer is deposited by vacuum evaporation by using a printed shadow mask. The mobility in the saturation regime is 0.5 cm^2/Vs, while an on/off ratio is 10^7 in DC measurements. The transistors operate efficiently in the small signal regime of the ultrasonic transducers (~1 mVp-p). The transistors with the grounded gate can switch 40 kHz signals with an on/off ratio of greater than 10^4.

A 28-µm-thick piezoelectric polyvinylidene fluoride (PVDF) film [24], whose surfaces are covered by metal, is used as a sheet-type ultrasonic transducer. The transducers are used in the bending mode. To adjust the mechanical resonance frequency to 40 kHz, the transducer is bended to improve the sensitivity using a spacer sheet. The ultrasonic transducer has an output voltage of ~0.8 Vp-p and a narrow pulse width of ~100 µs after an amplification of 40 dB by the incidence of an ultrasonic wave. The ultrasonic transducers can detect a stiff object that is hidden inside a cloth. It also shows broad directivity (> –5 dB for ± 60º).

The characteristics of the integrated ultrasonic sensing cell comprising one ultrasonic transducer and one organic FET are examined. The signal from the ultrasonic transducer is switched by the organic FET at 40 kHz. The integrated sensing cells have an on/off ratio of 10^4 at 40 kHz. 3D ultrasonic images can be obtained for multiple target objects over this sheet. This system can detect target objects behind a cloth and a paper.

A pocket scanner

As a new development in the field of large-area electronics, we have developed a large-area, flexible, and lightweight sheet image scanner based on organic semiconductors [9]. We report its principle of imaging, manufacturing process, and electronic performance. The device is manufactured on plastic films by integrating organic FETs and organic photodiodes. Organic photodetectors distinguish between the black and white parts of paper based on the difference in their reflectivity. The effective sensing area of the integrated device is 5 × 5 cm^2; resolution, 36 dpi; and the total number of sensor cells, 5,184. Furthermore, by using two-dimensional arrays of a photodiode matrix without the organic transistors, we have successfully obtained images of several characters whose size is approximately 1 × 1 mm^2. The present sheet-image scanners are mechanically flexible, lightweight, and very thin; therefore, suitable for human-friendly mobile electronics.

A sheet-type Braille display

Organic transistors are suitable for applications to plastic large-area actuators. We have developed a flexible, lightweight *Braille sheet display* that is fabricated on a plastic film, by integrating high-quality organic FETs with plastic actuators [10]. A small hemisphere which projects upwards from the rubber-like surface of the display is attached to the tip of each rectangular actuator (Fig. 8).

A wireless power transmission sheet

We have also demonstrated the implementation of a large-

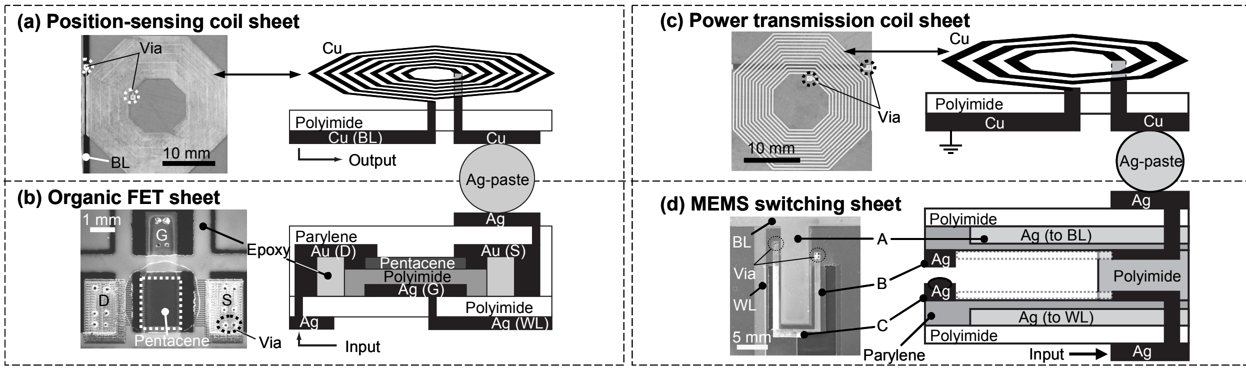

Fig. 9: A wireless power transmission sheet. The contactless position-sensing sheet comprising (a) position-sensing coil sheet and (b) organic FET sheet. S, D, and G represent the electrodes for source, drain, and gate, respectively. The organic FET sheet is laminated with the position-sensing coil sheet using silver paste islands. A resonance frequency of 2.95 MHz is used in the position-sensing system. WL and BL represent word-line and bit-line, respectively. The power transmission sheet comprising (c) power transmission coil sheet and (d) MEMS switching sheet. Electrodes for electrostatic attraction connected to the word-line (WL) and bit-line (BL) of MEMS switch. B and C: Electrodes for power transmission connected to coils and a power generator.

area wireless power transmission system [11]. The sheet-type large-area wireless power transmission system has been manufactured using organic transistors and MEMS switches (Fig. 9). The position of electronic objects on this sheet can be contactlessly sensed by electromagnetic coupling using an organic transistor active matrix. Then, power is selectively fed to the objects by an electromagnetic field using a two-dimensional array of copper coils that are driven by a printed plastic switching matrix. The effective power transmission area is 21 × 21 cm^2. Due to selective power transmission, we achieved a coupling efficiency of power transmission of 81.4%, and a power of 40 W was wirelessly received. The thickness and weight of the entire sheet are 1 mm and 50 g, respectively.

Remaining issues and future prospects

We describe the issues of organic transistors from the viewpoint of ambient electronics. Stability and reliability are the main concerns for the organic transistors. Degradation may be induced by oxygen and/or moisture like electroluminescent (EL) devices; therefore, it can be suppressed drastically by appropriate encapsulation. Although plastic films usually exhibit high gas penetration, applications that require mechanical flexibility, such as electronic skins and sheet image scanners, also require flexible substrates with low gas permeability. Thus, it is very important to develop flexible base films with low gas permeability.

Finally, we would like to briefly describe a futuristic life that would be possible by ambient electronics. Please image that the interiors of a car will be covered with large-area sensors. Based on flexible biometrics installed in the steering wheel, the driver is identified. When I fasten the seat belt, which is an invisible flexible sensor, it will start monitoring the heartbeat. If the grip pressure and the heart rate decrease, the car perceives that the driver's concentration is declining. Subsequently, stimulus was returned to the drivers by actuators. Our prototypes demonstrate that organic transistors can be used to extend the field of electronics into large areas. One day high-speed integrated circuits will be printed everywhere you want such as surfaces on plastic films, paper, and cloth. Subsequently, electronics can jump out of a confined small box of a computer into a real world and substantial progresses in electronics will continue.

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