Invited Review

Ultrathin Organic Electronics

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(Received Nov. 10, 2014)

We have succeeded in developing the world’s lightest (3 g/m²) and thinnest (2 µm) mechanically flexible organic photoelectric conversion devices [ultrathin organic solar cells and ultrathin organic light-emitting diodes (LEDs)] and electronic switches (ultrathin organic transistor integrated circuits) utilizing the intrinsic flexibility and processability at low temperatures of thin organic materials. Successful integration of a transistor and a p-n diode (LED or optical sensor), which are basic components of electronics, on a polymer film with a thickness of only 1 µm is expected to lead to the fundamental technology enabling the realization of ultrathin imperceptible wearable electronics that can change their shape according to the shape of the surface on which they are placed in the near future. In this review, the progress in the development and technical issues of ultrathin (~1 µm) flexible electronics as well as their future prospects including their applications in fields ranging from next-generation biomedicine to welfare are introduced.

Keywords: Mechanically flexible, Organic solar cells, Organic light-emitting diodes, Imperceptible devices

1. Background and purpose

Along with the declining birth rate and aging population, interest in medical services and health has been increasing among people. In addition, with the spread of internet technologies, information is more globalized and the lifestyle of people is diversified because they can easily access the worldwide web (WEB) anytime. In this social background, the electronics used in daily life have become increasingly diversified. The silicon-based technology supporting our modern society has realized high-density integration, significant power savings, and high-speed operation, along with the trend of miniaturization realized by photolithography technology, and has made rapid progress. In the diversified modern society, new performance indices different from those based on silicon-based technology, namely, light weight, durable, and user friendliness, have been sought and become widespread among people. In particular, wearable electronics have attracted attention since the beginning of this year.

Not only actuator-type devices, such as roboticorthoses and power suits, but also implantable medical devices, such as heart pacers and electrodes for deep brain simulation to treat epilepsy, can be included in wearable electronics in the broad sense. Recently, however, the term wearable electronics has been used as a general term for any device used to detect biosignals and perform operations. Google Glass and Intel Mimo are typical examples of such devices. The range of applications of these devices is extremely wide because they target population health, that is, the health of all human beings. Expectations have been growing for wearable electronics that can accurately obtain human biosignals within the social background of declining birth rate and aging population as well as the holding of the 2020 Olympic Games to be held in Tokyo.

To obtain accurate human bio-information, sensors should be placed as close to the human body as possible. In addition, flexible sensors are preferred because hard sensors attached to the soft surface of a human body may generate a sense of discomfort owing to the difference in hardness between the sensor and the skin. The research and development of the fabrication of electronics on substrates with favorable mechanical properties, such as flexible plastic and elastomeric films, has been extensively carried out in not only Japan but also other countries throughout the world. They are collectively called flexible or stretchable electronics (1-13). As materials for flexible semiconductor layers, which are the most important components of electronics, InGaAs/InAlAs (14), single-crystalline silicon nanomembranes (15), indium gallium zinc oxide (IGZO) (16), MoS₂ (17), graphene (18), nanotubes (19), amorphous silicon (20), and organic semiconductors (4, 5, 8, 10) have been used. The application of these flexible semiconductor layers to flexible displays, flexible sensors, and flexible radio-frequency identification (RFID) tags has been examined. Some of them have already been introduced into the market. The flexible and stretchable electronics technologies have started to be applied to wearable bioinstrumentation electronics technologies.

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Our research group has been developing large-area, flexible, lightweight, and thin electronics sensors, the presence of which cannot be perceived when worn, using organic-semiconductor flexible transistors as the fundamental technology. The goal of our study is to realize large-area, multipoint sensing using sensors that are thinner than the human skin and can cover a wide surface area even for complex shapes rather than one-point sensing using conventional sensors. We attempt to realize a next-generation bioinstrumentation system with a reduced impact on the human body by using organic electronics.

In this review, I will introduce fabrication methods and performance characteristics of ultrathin (1 µm) organic LEDs and organic solar cells (optical sensors), as well as of the organic transistors that make up the former two devices. Finally, I will discuss the future prospects of imperceptible biointerface electronics, which are realized by integrating the above devices.

2. Ultrathin organic LEDs

Organic LED displays and illumination equipment have been put to practical use and used in various aspects of daily life. Organic LEDs are excellent in terms of power consumption, color reproducibility, and response speed. Thus, they are expected to be applied not only for illumination and display, but also in medicine. Because of this trend, lighter and thinner organic LEDs are demanded. Our research group has...
established an original fabrication technology to laminate organic semiconductor or electrode materials onto commercially available ultrathin (1.2 µm) polymer (polyethylene terephthalate (PET) and polyethylene naphthalate (PEN)) films at low temperatures and low power consumption. We also succeeded in fabricating flexible organic LEDs that are the lightest and thinnest in the world21). Fig. 1 (a) shows a schematic of the ultrathin organic LED and its composite layers. The key to the development of the organic LED is the low-temperature process by which organic LEDs are fabricated on 1-µm-thick polymer films with a relatively rough surface (root mean square (rms) roughness, 1 nm) without damaging the films. Concretely, a conductive polyol that can be formed into films at low temperatures \( \leq 120°C \) and with low loss is used as the material of the electrode (anode), instead of an indium-tin oxide (ITO) transparent electrode that re-

![Figure 1](image1.png)

**Fig. 1** (a) shows a schematic of the ultrathin organic LED and its composite layers.

![Figure 2](image2.png)

**Fig. 2** Organic solar cell (optical sensor) fabricated on thin film

(a) Schematic showing composite layers and thickness of each layer, (b) Photograph, (c) Current–voltage (I–V) characteristic (d) Dark current (dashed line) and photovoltaic current (red solid line), (e) Comparison of amounts of electric power (Specific weight) among different solar cells with various semiconductor materials. The bar at the right end indicated by “Ultrathin OPV” shows the result for our organic solar cell.
requires high-energy processing. Light emission was realized using a luminescent polyphenylene vinylene-based polymer material \( \text{anthracene-containing poly (p-phenylene-ethynylene-alt-poly (p-phenylene-vinylene) (PPE-PPV) polymer (AnE-PVstat)} \) for the active layer. The organic LED has a thin substrate (1 µm); thus, it can accommodate flexing strain induced by folding and can operate electrically without any change in its properties even when it is crumpled. The luminance was 100 cd/m\(^2\) (cd means candela) when driven at 9 V [Figs. 1 (c) and 1 (d)]. Fig. 1 (d) shows the \( V-L-I \) characteristics of the LED with red-emitting AnE-PVstat (red curve) and that with a typical luminescent polyphenylene-based polymer material, orange-emitting poly [2-methoxy-5 (3,7-dimethyloctyloxy)-1,4-phenylene-vinylene (MDMO-PPV) (blue curve). The obtained luminance is equivalent to that necessary for indoor use. As shown in the chromaticity diagram [Fig. 1 (e)], the emission wavelength (red) is 640 nm. The ultrathin organic LEDs have high bending flexibility and their properties do not deteriorate when they are bent to a radius of curvature of 5 µm. The range of applications of the LEDs can be widened by utilizing their mechanical flexibility. For example, when the ultrathin organic LED film is pasted onto a prestretched flexible elastomer and then the stretched strain is released, an accordion-like wavy LED is obtained. The wavy organic LED was demonstrated to accommodate stretching of up to 100% or more. Stretchable LEDs that can realize a complex shape surface are expected to be applied to not only display media, such as displays, but also new light sources for medical purposes. The improvement of luminance and luminous efficiency of energy generated by flexible organic solar cells 22). Fig. 2 (a) and 2 (b) show a schematic of the composite layers and a photograph of the fabricated ultrathin organic solar cell, respectively. The developed solar cells can also be used as photodetectors (optical sensors). Namely, the devices can be used as ultrathin organic optical sensors by applying a reverse bias (negative and positive voltages to p-type and n-type semiconductors, respectively).

3. Ultrathin organic solar cells—Optical sensors—

With the increasing interest in environmental energy, the importance of solar cells as a renewable energy source that can compensate for the insufficiency of energy generated by conventional methods has been increasing. In addition, an ambient energy harvesting technology for obtaining energy from the environment is particularly important considering a future era when various types of electronics, such as sensors, will be installed not only in living organisms but also everywhere around us. Large-area solar cells should be realized at a low cost because electric power generation using solar cells is basically proportional to their area. In addition, lightweight and increase in impact resistance (flexibility) are required, along with an increase in the area of solar cells. However, the solar cells currently available on the market are fabricated on glass substrates using silicon, resulting in a low impact resistance. Furthermore, when the glass substrate is thinned, the solar cells may break during fabrication or use, hampering the realization of lightweight solar cells. In contrast, solar cells comprising organic semiconductors can be easily fabricated on polymer films by a liquid process such as printing. Therefore, the research on these solar cells has been vigorous with the expected realization of large-area, low-cost, and lightweight solar cells. However, organic solar cells with a high power conversion efficiency equivalent to that of glass substrates may not be fabricated on flexible thin polymer films by a liquid process; this issue must be resolved.

We succeeded in realizing the world’s thinnest and lightest flexible organic solar cells\(^-\). Fig. 2 (a) and 2 (b) show a schematic of the composite layers and a photograph of the fabricated ultrathin organic solar cell, respectively. The ultrathin organic solar cells were fabricated on PEN or PET films. Even when bent to a radius of curvature of 35 µm, they are not mechanically broken and maintain the power conversion efficiency of 4.2% [Figs. 2 (c) and 2 (d)]. In fact, the ultrathin organic solar cell can be wound around a human hair (radius, approximately 10 µm). The key technology in the development was the low-temperature film growth process, in which p-type semiconductor ink [poly (3-hexylthiophene-2,5-diyl) (P3HT)] and n-type semiconductor ink [(6,6)-phenyl-C61-butyric acid methyl ester (PCBM)] are mixed in an organic solvent and deposited onto a 1.4-µm-thick PET film. Poly (3,4-ethylenedioxythiophene) : poly (styrenesulfonate) (PEDOT : PSS), a conductive polymer that can be treated by a liquid process, was used as the transparent electrode. By placing the ultrathin solar cells on a prestretched elastomeric substrate, the solar cells can be stretched or compressed up to 300%. Thus, organic solar cells with a power conversion efficiency of 42% and steady properties even when bent to a radius of curvature of 35 µm or stretched or compressed up to 300% were successfully fabricated on polymer films. The electric power generation per gram of the ultrathin organic solar cell is equivalent to 10 W. Our organic solar cells are lighter, thinner, and more flexible than any other commercial solar cells [Fig 2 (e)].

The developed solar cells can also be used as photodetectors (optical sensors). Namely, the devices can be used as ultrathin organic optical sensors by applying a reverse bias (negative and positive voltages to p-type and n-type semiconductors, respectively).

4. Organic thin film transistors (TFTs)

Organic TFTs used as flexible electronic switches are easily fabricated on polymer films by a liquid process such as printing. Therefore, the research on organic TFTs has also been active with the expected realization of large-area, low-cost, lightweight, and flexible organic TFTs. However, organic transistor integrated circuits with a high electric performance...
equivalent to that of glass substrates may not be fabricated on flexible ultrathin polymer films, particularly those with thicknesses $\leq 10 \, \mu m$; this issue must also be resolved.

We developed the world’s lightest (3 g/m², which is much lighter than a feather) and thinnest (2.4 µm) organic TFT active matrix and its integrated circuit [Fig. 3(a)]. All the components constituting the system are directly deposited one by one onto a 1.2-µm-thick PEN film [Fig. 3(b)], and then whose weight is much lighter. The film can withstand bending to a radius of curvature 5 µm, can be crumpled like paper, and will not break upon dropping from heights exceeding 1 m. The mobility is $3 \, cm^2/Vs$ at a drain-to-source voltage ($V_{DS}$) of 3 V, demonstrating highly uniform electric characteristics [Figs. 3(c)-(e)]. The key to developing these organic TFTs is the...
successful fabrication of a highly uniform 19-nm-thick ultrathin insulating film on a polymer film with a surface roughness of ~1 μm with good adhesion. Concretely, we established an original room-temperature process based on anodic oxidation to form uniform aluminum oxide films with good adhesion to the substrate. In the conventional plasma oxidation method, the ultrathin polymer films under the aluminum oxide film are damaged by the plasma, leading to the generation of pinholes. The anodic oxidation method adopted in our study did not involve any high-energy processes, such as plasma processing, and the conventional problems were solved.

The organic transistor integrated circuits were found to be extremely durable. In addition, no significant deterioration in electric characteristics was observed after immersion in a physiological saline solution (with components equivalent to those of body fluid and sweat) for two weeks. Furthermore, no deterioration in electrical or mechanical characteristics was confirmed when both ends of a stretchable integrated circuit were fixed to a mechanical compression/stretching test apparatus and compressed/stretched up to 233%. Fig. 3 (f) shows a photograph of an ultrathin transistor integrated circuit attached to the back of a human hand. The circuit can follow the complex shape of the human hand because it is thin. This type of flexible system can be attached to curved surfaces to measure changes in pressure and temperature.

5. Future prospects

Imperceptible ultrathin and lightweight electronics are expected to be applied in various fields ranging from health care to medicine to welfare because of the progress in their device technologies (e.g., TFTs, LEDs, and photodiodes). Along with the development of thinner and lighter sensing systems, sensors that cannot be perceived when worn can be used to obtain bioinformation unobtrusively. It is expected that bioinformation can be obtained over 24 h in a stress-free manner while leading a normal life. In addition, bioinformation (e.g., body temperature and heart rate) can be obtained anytime, for example, during exercise, by using these electronics as highly impact-resistant sensors. When these electronics are combined with the world's lightest solar cells, optical sensors, and organic LEDs developed concurrently, their application is expected to open new fields such as those involving health care sensors for monitoring health conditions in a free-standing manner and semipermanently powered by electricity converted from indoor and outdoor light.

Acknowledgements

The achievements described in this article were obtained through the collaboration of researchers at the University of Tokyo (Prof. Takao Someya, Dr. Martin Kaltenbrunner, Dr. Tomoyuki Yokota, Dr. Kazunori Kurihara, and Mr. Takeyoshi Tokuhara) and those of Johannes Kepler University of Linz (Dr. Matthew S. White, Dr. Eric D. Glowacki, Dr. Michael Drack, Dr. Reinhard Schwoediauer, Dr. Ingrid Graz, Dr. Simona Bauer-Gogonea, Professor Siegfried Bauer, and Professor Niayzi Serdar Sariciftci).

References


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Prof. Sekitani received Ph. D. from the Department of Applied Physics, School of Engineering, at the University of Tokyo, in 2003. From 2003 to 2010 he was an Assistant Professor in the Department of Applied Physics at the University of Tokyo and worked with Prof. Takao Someya. In 2011, he advanced to the role of Associate Professor in the Department of Electrical Engineering and Information Systems at the University of Tokyo. In 2014, he has been Professor in the Institute of Scientific and Industrial Research, Osaka University.

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