

Recent Advances in Flexible Sensors for Wearable and Implantable Devices

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ABSTRACT: Flexible devices are emerging as important applications for future display, robotics, *in vitro* diagnostics, advanced therapies, and energy harvesting. In this review, we provide an overview of recent achievements in flexible mechanical and electrical sensing devices, focusing on the properties and functions of polymeric layers. In the order of historical development, sensing platforms are classified into four types: electronic skins for robotics and medical applications, wearable devices for *in vitro* diagnostics, implantable devices for human organs or tissues for surgical applications, and advanced sensing devices with additional features such as transparency, self-power, and self-healing. In all of these examples, a polymer layer is used as a versatile component including a flexible structural support and a functional material to generate, transmit, and process mechanical and electrical inputs in various ways. We briefly discuss some outlooks and future challenges toward the next steps for flexible devices. © 2013 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 130: 1429–1441, 2013

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INTRODUCTION

Recent studies on flexible mechanical and electrical sensing devices have demonstrated great potential in a wide range of applications, such as displays,^{1,2} robotics,^{3–6} *in vitro* diagnostics,^{7–9} advanced therapies,^{10,11} and energy harvesting.^{2,12–14} Such remarkable advances have been achieved through various solutions through the use of electrical active matrices on flexible rubberlike substrates to obtain various functions, such as high bendability,^{15,16} ultrasensitivity,^{7,17} transparency,^{1,2,18–21} or well-established human–device interfaces.^{8,9,22,23} In general, to be able to measure multifunctional mechanical and electrical signals, a number of circuit elements involving organic/inorganic matrix arrays,^{3,4,17,24–31} hybrid composites,^{32–38} graphene,^{39,40} and nanowires (NWs) or nanotube assemblies^{41–44} need to be integrated on various flexible substrates.⁴⁵

About a decade ago, flexible electronic skins (e-skins) for pressure sensing were first introduced with polymer-based switching matrices for future displays, robots, and prosthetics of mechanical communications [Figure 1(a)].^{3,4,17,41} Recently, e-skins have evolved to wearable or skin-attachable electronic devices for

motion detection on clothing or a diagnostic tool to monitor body signals [Figure 1(b)].^{7,8,42} Here, the polymer plays an important role as a biocompatible, less irritating support layer. In parallel, sensors have been used as essential components of biomedical mapping systems in surgical operations through the exploitation of conformal contact with the surfaces of human organs, such as the brain and heart [Figure 1(c)].^{10,11,46} To address some shortcomings of the current flexible devices, several innovative features have been incorporated into sensor systems, including transparency,^{1,21} self-healing capabilities,⁴⁷ and energy harvesting [Figure 1(d)].²

To achieve high flexibility in sensing devices, various fabrication methods are currently available with polymeric materials, as shown in Figure 2, such as low-temperature deposition,⁴⁸ solution processing,^{49–53} transfer printing,^{31,54–56} and nanomolding/micromolding.^{2,7–9,17} Typically, transistor arrays made of organic semiconductors or inorganic transferred nanomembranes/ribbons have been considered as key elements because of their defect-free layout, high-resolution processing, and superior carrier mobility.^{31,57} In addition, a solution-based process with nanotubes/

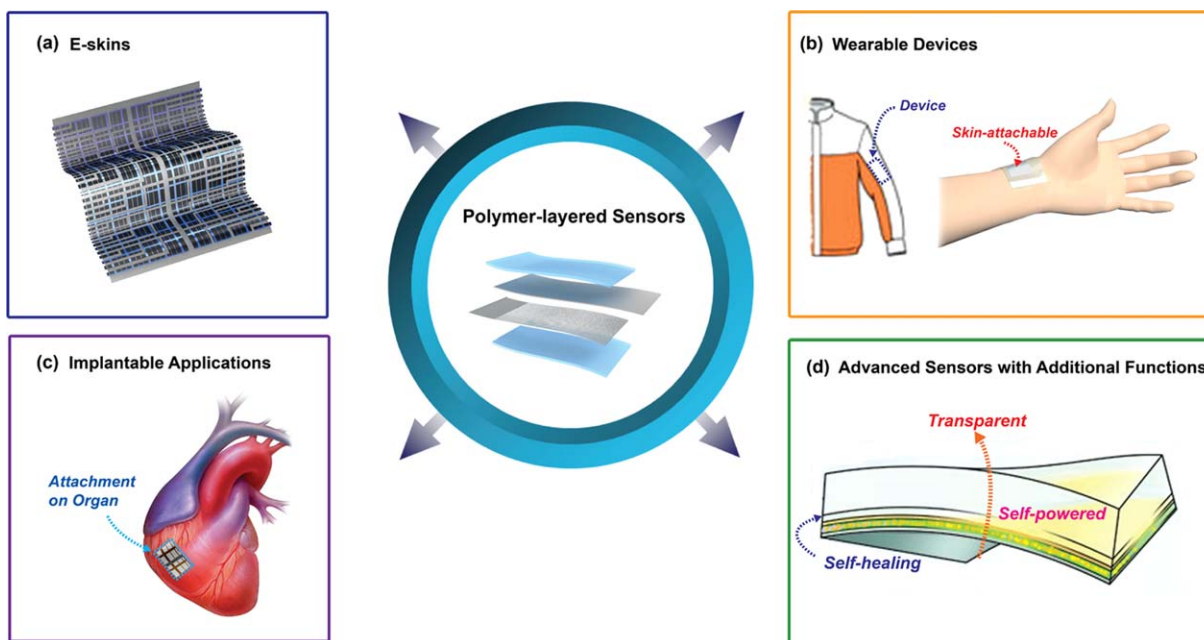


Figure 1. Illustrations of flexible mechanical and electrical sensors: (a) E-skins, (b) wearable and skin-attachable sensors, (c) implantable devices for *in vivo* diagnostics, and (d) advanced sensors with additional functionalities. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

NWs has been used as a facile method for fabricating large-area devices.^{1,41} More recently, bio-inspired nano-interlocking structures have been proposed as a high-sensitivity element to detect various physical forces in a simple and cost-effective manner.⁷

It is worthwhile to note that the proper use of a polymer gets more important as the sensing device becomes more complex and multifunctional in a multilayer shape. In general, a polymeric supporting layer contributes significantly to the mechanical flexibility or stretchability; the device can be bent, stretched, and adapted into an arbitrary shape. For the multifunctional sensors described here, the polymer also plays key roles in various ways

for high-sensitivity detection,^{2,17} the self-programmable active layer,^{1,7} less irritating conformal contact on the skin,^{8,9} and biocompatible/waterproof sealing for implantable devices.⁴⁶

In this review, we provide an overview of recent advances in flexible mechanical and electrical sensing devices with particular emphasis on the properties and functions of the polymeric layers. In the order of historical development, the flexible sensing devices are categorized into four areas: (1) e-skins, (2) wearable devices, (3) implantable devices, and (4) advanced sensors with additional features such as transparency, self-power, and self-healing. For each application, the role of polymers is briefly described in terms

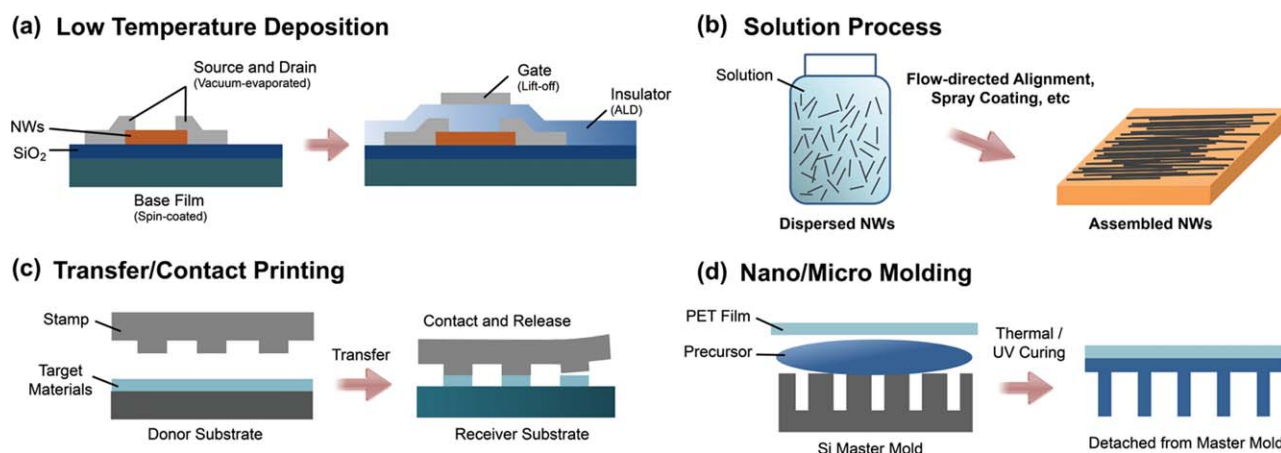


Figure 2. Schematic diagrams of current fabrication methods for flexible sensors: (a) low-temperature deposition method for an OFET (ALD: Atomic Layer Deposition), (b) solution-assisted assembling process, (c) transfer printing and contact printing method, and (d) nanomolding/micromolding for the construction of polymeric structures. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

of their material properties and structural characteristics. In the concluding section, we provide some perspectives and future directions for the next steps of these flexible devices.

CLASSIFICATION OF FLEXIBLE SENSING DEVICES

Figure 1 shows the classification of flexible sensing devices: e-skins, wearable/skin-attachable sensors, implantable sensors, and advanced sensors with additional features. For the e-skins shown in Figure 1(a), an array of organic field-effect transistors (OFETs) is used as an active layer; this is considered an essential element in the large-area fabrication of soft and flexible substrates.^{3,4,17,58} In addition, a parallel array of semiconductor NWs was explored as a versatile route in the fabrication of high-performance, bendable transistors for sensors on polymer supports.⁴¹ Next, a motion detector in the form of a skin-attachable electronic device was demonstrated with various organic and inorganic materials.^{7,42} In particular, skin-attachable devices can characterize the mechanical/electrical signals on various dynamic and complicated surfaces, such as skins with the help of biocompatible, less irritating properties of supporting layers [Figure 1(b)]. Recently, an innovative approach was introduced in diagnostic devices for medical surgery of brain and cardiac therapy with high-resolution mapping of bio-signals, as shown in Figure 1(c).^{10,11} Finally, some additional features have been incorporated into flexible mechanosensing devices, as shown in Figure 1(d), including flexible and transparent characteristics,¹ energy harvesting via a triboelectric effect,² and a self-repairing ability with novel functional polymer composites.⁴⁷ The aforementioned sensing devices consist of various assemblies with organic or inorganic materials, such as silicon (Si) NWs, carbon nanotubes (CNTs), conducting/metalized polymers, and metal electrodes.

Table I shows the summary of various flexible sensors categorized in Figure 1 in terms of active layers, the role of polymers, and fabrication methods. As shown in Table I, polymeric materials have been employed extensively as an essential component in flexible devices in a wide range of applications. To provide more details, the role of polymeric materials is described in three aspects as described in the following.

Supporting and Packaging Layer for Flexibility and Stretchability

Flexibility and stretchability, which are associated with the reversibility of polymeric backbones, are of paramount importance for sensing applications. These mechanical properties can be used to wrap uniformly complex curvilinear and time-dependent dynamic surfaces. In particular, the bendability of polymeric supports can be significantly elevated with the help of a specific shape, such as mesh,^{10,43} web,⁵⁹ or porous membrane.⁶⁰ Despite the versatility of polymer substrates, however, some inherent properties of polymers hinder the fabrication via conventional high-temperature processes with nanotube networks, nanoribbons, and nanomembranes. To overcome this problem, a number of low-temperature fabrication methods have been developed to stack inorganic NWs, including solution assembly,^{49–51} transfer printing,^{31,54,55} spray coating,^{1,61,62} and contact printing.^{41,56,63,64} As a result, various smart circuits in

the form of multilayer NW stacks, wave-patterned membranes, and embedded OFETs have been used for stable electric performance on plastic substrates upon bending or stretching. In some cases, a sacrificial layer made of a dissolvable material [e.g., silk fibroin, poly(methyl methacrylate) (PMMA), or poly(vinyl alcohol) (PVA)] has been used as an adhesive layer and for easy handling^{10,24,46}

Functional Layer for Programmable Active Matrices

Typically, an OFET array has been frequently used as a switchable matrix in part because of its low-temperature processing and high flexibility. Also, a polymer microstructure has been integrated as a gate dielectric; this further strengthens the deformability and mechanosensitivity of the device.¹⁷ More recently, it was demonstrated that the reversible and restorable characteristic of polymer contributes significantly to (1) the elastic self-reconstruction of polymer fibers when they are engaged in an interlocking geometry⁷ and (2) the programmable alignment of spray-deposited CNT films.¹ In these approaches, the elastic behavior of the polymer-based active layer gives rise to the restoration of signal outputs during multiple loading–unloading cycles.

Wettable Interface between Humans and Devices

To achieve skin-attachable/implantable devices, an adhesive layer for stable fixation on human skin or an organ needs to be addressed. Here, an interfacial layer between the human and the device is important as a supporting layer; this allows for a reliable transfer of vital or mechanical signals to the sensor matrix. A representative example can be found in a recent work by Rogers et al.,⁵⁹ in which an ultrathin and low-modulus sheet was used to detect the dynamic signals of a physical strain and temperature variation on the epicardial surface. In parallel, an organ-attachable, waterproof sealing with a polymeric substrate is essential for biomedical applications or monitoring medical surgeries.⁴⁶

By exploiting the previous features of polymeric layers, it would be possible to develop a more economically viable sensor system with high performance (e.g., fast response time and high sensitivity) and excellent mechanical properties (e.g., ultrathin flexibility and stretchability). Next, we describe four representative applications of these flexible sensors: (1) e-skins, (2) wearable devices, (3) implantable devices, and (4) some unique sensors with additional features.

E-SKINS

The name *e-skin* was originated on the basis of its similarity to human skin, which has the ability to sense the spatiotemporal distribution of an external stimulus with a soft substrate. The requirements of high flexibility and high sensitivity for e-skins are driving various research directions in design and fabrication. The earlier version of artificial e-skin consisted of flexible organic transistors; it was proposed by Someya and coworkers,^{3,4} as shown in Figure 3(a–i). Here, the device could detect an external pressure distribution over a large area (~ 64 cm²) with a gate dielectric of a spin-coated polyimide (PI) layer on the base substrate of a poly(ethylene naphthalate) (PEN) film. The drain currents of the sensor array were responsive to

Table I. Classification and Descriptions of Mechanical and Electrical Sensing Devices in Terms of Materials and Fabrication Methods

Category	Features	Active layer	Polymer layer (function)	Fabrication method	Reference
Electric skins	Flexible pressure sensors	Organic transistors	PEN/rubber (substrate)	Laser drilling/PVD	3
			PI (gate insulator) PET/PDMS (protection)		
Wearable devices	Flexible strain-gauge sensor	Ge/Si NWs	PI/rubber (substrate)	Photolithography/ etching/contact printing	41
			Parylene (insulator)		
			PET (substrate) PDMS (dielectric layer)		
Wearable devices	Flexible strain-gauge sensor	Pt-coated nanofibers	Micropyramids/ITO/organic transistors	Micromolding/PVD	17
			PET (substrate) PDMS (dielectric layer)		
			PET/PDMS (substrate) PUA (nanointerlocking)		
			PDMS (substrate)		
Implantable devices	Mapping brain signals	Au electrode patterns	PDMS (substrate)	Photolithography/ etching/ Transfer printing	10
			PDMS (substrate)		
			PE (contact to skin) PVA (temporary supports)		
			Silk (dissolvable substrate)		
Advanced sensors	Mapping cardiac electrophysiology	Si-based circuits	PI (substrate and dielectric layer) Epoxy (dielectric layer)	Spray coating	11
			PDMS (substrate) Ecoflex (filler)		
Advanced sensors	Transparent pressure sensor	CNTs/F4TCNQ	PET (substrate) PDMS (dielectric layer)	Micromolding	2
			PDMS (substrate) Ecoflex (filler)		
Advanced sensors	Self-powered sensor	Microspyrramids/ITO	Empol 1016 and DETA (self-healing) PET (substrate)	Mixing and compression molding	47
			Polymer- μ Ni composites		

PUA, poly(urethane acrylate); DETA, diethylenetriamine; F4TCNQ, tetrafluorotetracyanoquinodimethane.

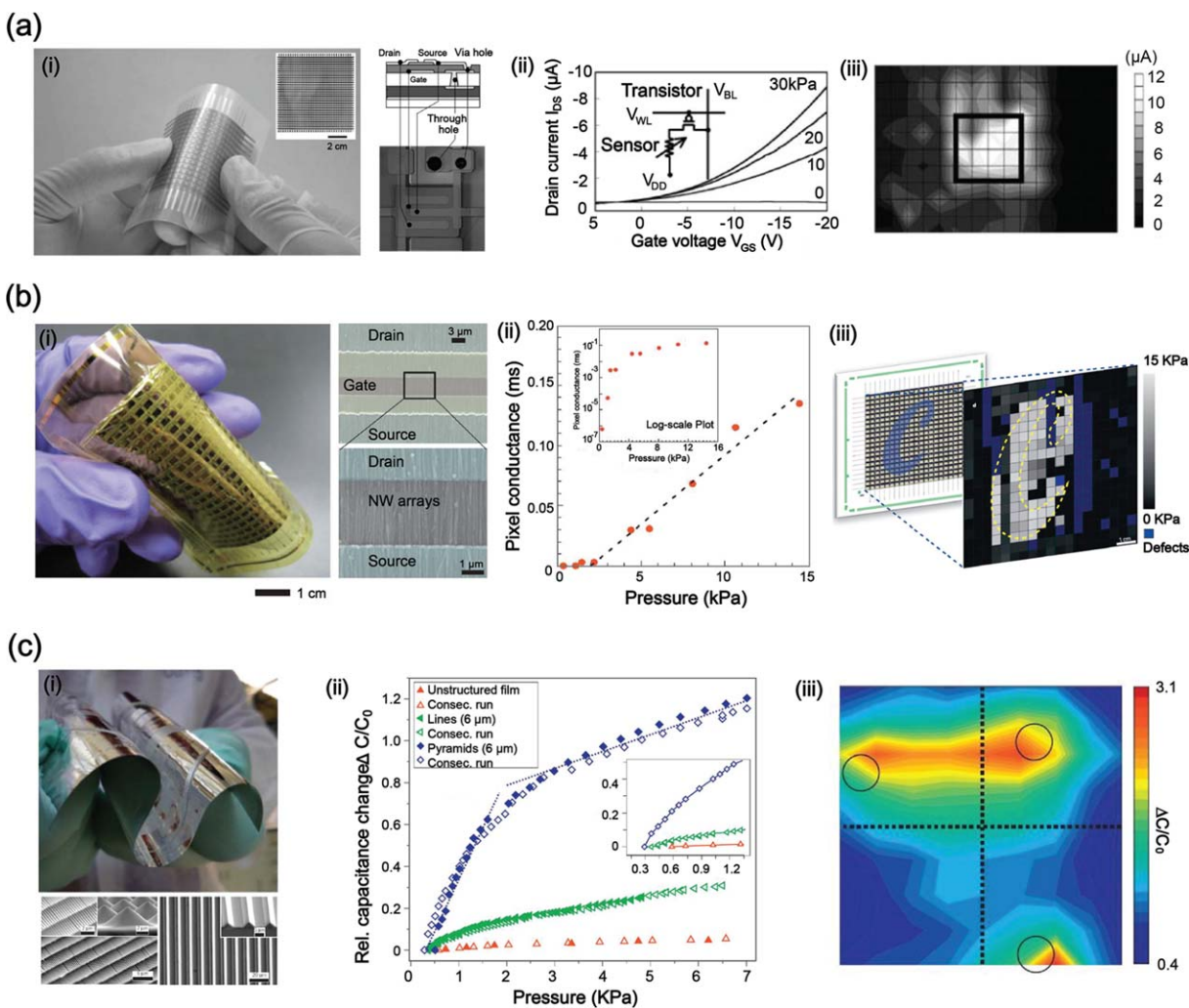


Figure 3. Various e-skin sensors. (a) An array of OFETs with (i) images of overall sensor matrix (32×32 arrays) and a single cell, (ii) I–V (current–voltage) characteristics under various pressures, and (iii) measured current distribution. (Reprinted with permission from ref. 3. Copyright 2004 National Academy of Sciences.) (b) Integrated Ge/Si NW arrays with a (i) photograph of fabricated e-skin device and scanning electron microscopy images of aligned NW arrays, (ii) linear response of the NW field-effect transistor, and (iii) a map of measured pressure distribution (C). (Reprinted with permission from ref. 41. Copyright 2010 Nature Publishing Group.) (c) OFETs with micropatterned rubber dielectric layer with (i) a wafer-scale molding of micropatterned PDMS thin film, (ii) capacitance change curves for different microstructures, and (iii) a spatial distribution map by external pressure. (Reprinted with permission from ref. 17. Copyright 2010 Nature Publishing Group.) C_0 : capacitance without applied pressure; V_{BL} : bit line; V_{WL} : word line; V_{DD} : supply voltage; V_{GS} : gate voltage. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

the loading pressure within a dynamic range of 10–30 kPa [Figure 3(a–ii,iii)].³ Many years later, the same group demonstrated the use of an organic thin-film transistor network on an ultra-thin polymeric substrate with extreme flexibility.¹⁵

In addition, Javey et al.⁴¹ demonstrated an artificial e-skin with aligned germanium (Ge)/Si NW circuits in a large-scale active system (19×18 pixels of $7 \times 7 \text{ cm}^2$ areas). In general, a highly ordered NW assembly is challengeable, despite its several advantages, such as high resolution, low power, and high sensitivity.⁶⁵ Here, the integration of a well-aligned NW array was achieved by the contact printing of NWs grown by chemical vapor deposition, as shown in Figure 3(b–i), in which PI and a pressure-sensitive rubber were used as a base substrate and a sensing element, respectively. The performance of an individual sensing component was recorded as a normal pressure up to about 15

kPa; this resulted in a spatial mapping of the local distribution of loads with a character C, as shown in Figure 3(b–ii,iii).⁴¹ There were some defects in the flexible devices because of the difficulty associated with handling NWs in the fabrication process. A stretchable system based on a honeycomb-shaped PI mesh substrate, which could cover a high curvilinear, three-dimensional object such as a baseball, was also reported.⁴³

Another notable achievement for e-skin was demonstrated by Bao et al.¹⁷ with a microstructured poly(dimethyl siloxane) (PDMS) rubber for dielectric layer, as shown in Figure 3(c). In this study, an indium tin oxide (ITO)/poly(ethylene terephthalate) (PET) film was prepared with different types of micropatterned PDMS structures [Figure 3(c–i)]. The microstructured PDMS could alleviate viscoelastic behavior of the thin PDMS film and result in a higher sensitivity and a faster response.

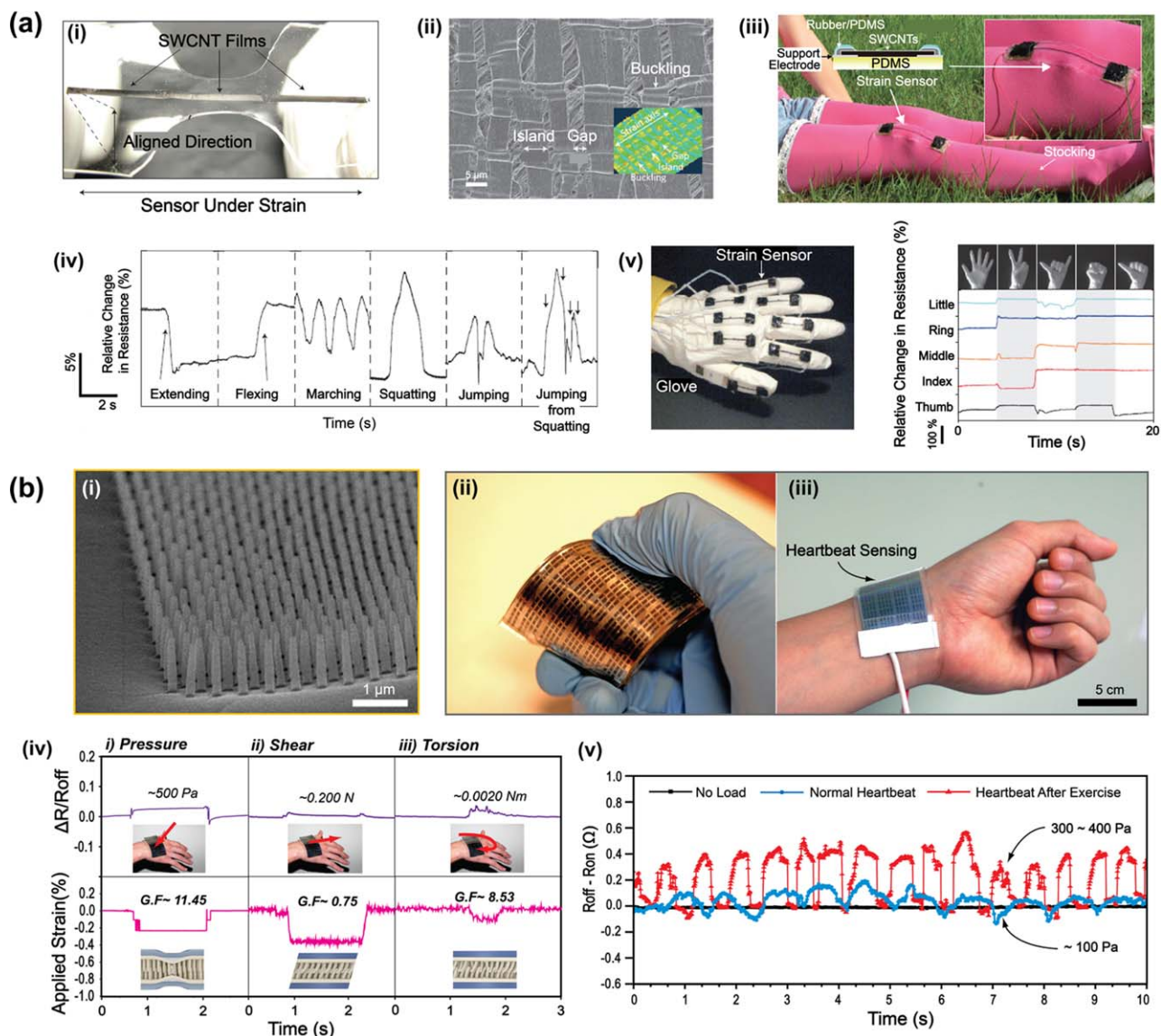


Figure 4. (a) Stretchable human-motion detector: (i) photograph of a stretched SWCNT film strain sensor, (ii) scanning electron microscopy image and three-dimensional view (inset) of the regularly fractured SWCNT structure, and (iii–v) strain-gauge sensors and signal patterns of attachment on knee and a glove. (Reprinted with permission from ref. 42. Copyright 2011 Nature Publishing Group.) (b) Flexible strain-gauge sensor: (i) scanning electron microscopy image of a high-aspect-ratio nanofiber array, (ii–iii) images of an assembled sensor and attachment on the artery of the wrist, (iv) graphs showing the ability of decoupling the external stimulus to pressure, shear, and torsion, and (v) plots of the heart rate under normal and postexercise conditions. (Reprinted with permission from ref. 7. Copyright 2012 Nature Publishing Group.) ΔR : difference in the electrical resistance; (Roff–Ron) Roff: resistance when unloading; Ron: resistance when loading; G.F.: gauge factor. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Here, the device with a pyramidal shape (6- μm feature size) demonstrated the best performance with a minimum detectable pressure as small as about 3 Pa; this corresponded to the weight of a tiny fly. [Figure 3(c–ii)]. Also, the pressure sensor pad (64 pixels of 64- mm^2 areas) addressed the applied forces from multiple points at the same time, as shown in Figure 3(c–iii).¹⁷

Currently, it appears that e-skins are ready to be used in various fields, such as multitouch sensing in flexible displays⁶⁶ and humanoid robotics and prosthetics.^{26,57,67,68} The use of polymeric layers should increase because of industrial demands for cost-effective fabrication with high sensitivity and resolution.

WEARABLE DEVICES

In addition to the advances in e-skins, a number of innovative approaches have been proposed for wearable or skin-attachable devices for the detection of complicated human motions and *in vitro* diagnostics on human skin. For this purpose, the integration of various functional parts seems essential; this has been demonstrated in a number of ultrathin electronic devices by the integration of multiple components, including sensors, light-emitting diodes, signal transmitters, and power generators.^{24,69–73} A wearable diagnostic or therapeutic device has the potential to transform future ubiquitous healthcare, where technology can monitor and improve a patient's condition.^{8,9,74} Recently,

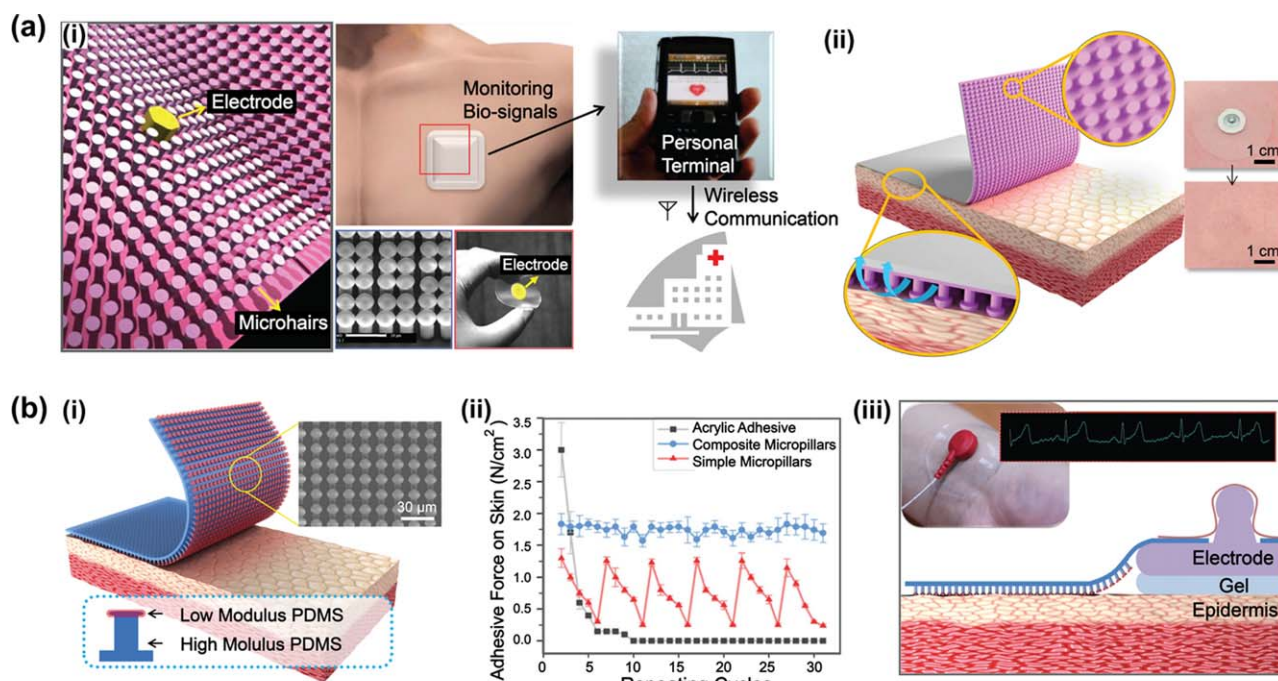


Figure 5. Skin-attachable applications for measuring biosignals with the aid of a dry adhesive skin patch. (a) Dry adhesive medical skin patch with (i) schematics of skin adhesive integrated with an electrode and concept of a real-time healthcare system and (ii) illustrative advantages of a dry adhesive patch (Reprinted with permission from ref. 8. Copyright 2011 John Wiley and Sons.). (b) Enhanced skin adhesive patch with composite micropillars with (i) a schematic illustration and SEM image of heterogeneous micropillars, (ii) plot of the enhancement of adhesion, and (iii) ECG signal measurement with an electrode attached on the skin. (Reprinted with permission from ref. 9. Copyright 2013 John Wiley and Sons.) [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

bio-inspired approaches have been used for skin-attachable sensors through the mimicking of unique structural features from the gecko lizard. For example, one can take advantage of hairy structures in a dry adhesive patch with less surface contamination and more ventilation of air, moisture, and skin residues.^{8,9}

In parallel, an innovative wearable device for sensing human motion was demonstrated by Hata et al.,⁴² as shown in Figure 4(a). To assimilate with various dynamic motions of humans, a high stretching capability is considered a critical property. Here, the device was fabricated by an alignment process of single-walled carbon nanotubes (SWCNTs) on a thin PDMS substrate. Strikingly, it could be stretched up to 280%⁴² before the substrate ruptured. Such extreme stretchability was attributed to the fracturing of the SWCNT film, with the aid of a mesh structure with gaps and islands [Figure 4(a–i,ii)]. Nonetheless, viscoelastic creep was inevitable in the highly stretched state; thus, the device needed about 5 s to recover its original shape when it was elongated at 100% strain. When the sensor was attached to the knee joint, various knee motions were detected, such as initial knee flexing and quick knee extension, as shown in Figure 4(a–iii,iv)]. Also, the sensors on a glove were able to measure different motions of each finger [Figure 4(a–v)]. The wearable device is notable for its nonrestrictive capability of detecting human motions for human-friendly rehabilitation.^{42,75}

More recently, a notable sensing mechanism was introduced to enable the detection of various types of mechanical stimuli,

even on human skin. Suh et al.⁷ demonstrated a layered strain-gauge sensor based on nanoscale mechanical interlocking between metal-coated, high-aspect ratio nanofibers, as shown in Figure 4(b). Interestingly, this van der Waals force-assisted interlocking phenomenon can be found in the wing-locking device of a beetle to operate reversible fixation of delicate flying wings via sensing organs.^{76,77} In contrast to other sensing devices with switching layers,^{1,24,41,49,78} the nano-interlocking mechanism did not involve any complex, integrated nanomaterial assemblies, or multiple arrays. Hence, it may allow for a simple yet robust sensing platform for the production of highly sensitive, large-area strain-gauge sensors.⁷ In particular, the sensor could measure and distinguish three different mechanical loads in the form of normal pressure, shear, and torsion like human skin does by interpreting the gauge factor of each case (~ 11.5 for pressure, ~ 0.75 for shear, and ~ 8.53 for torsion) with a high sensitivity and repeatability [< 8000 cycles; Figure 4(b–iv)].⁷ For potential applications in *in vitro* diagnostics, the physical force of a heartbeat was monitored in real time after the sensor was attached to the artery of a volunteer's wrist with medical adhesive [Figure 4(b–iii)]. As shown in Figure 4(b–v), the heartbeats were monitored in real time under two distinct conditions: normal (~ 60 beats per min with an average intensity of ~ 100 Pa) and exercise conditions (~ 100 beats per min with an average intensity of 300–400 Pa). The signals could be differentiated by discernible magnitudes and frequencies; this suggested that the sensor could be used as a diagnostic tool to measure biosignals,

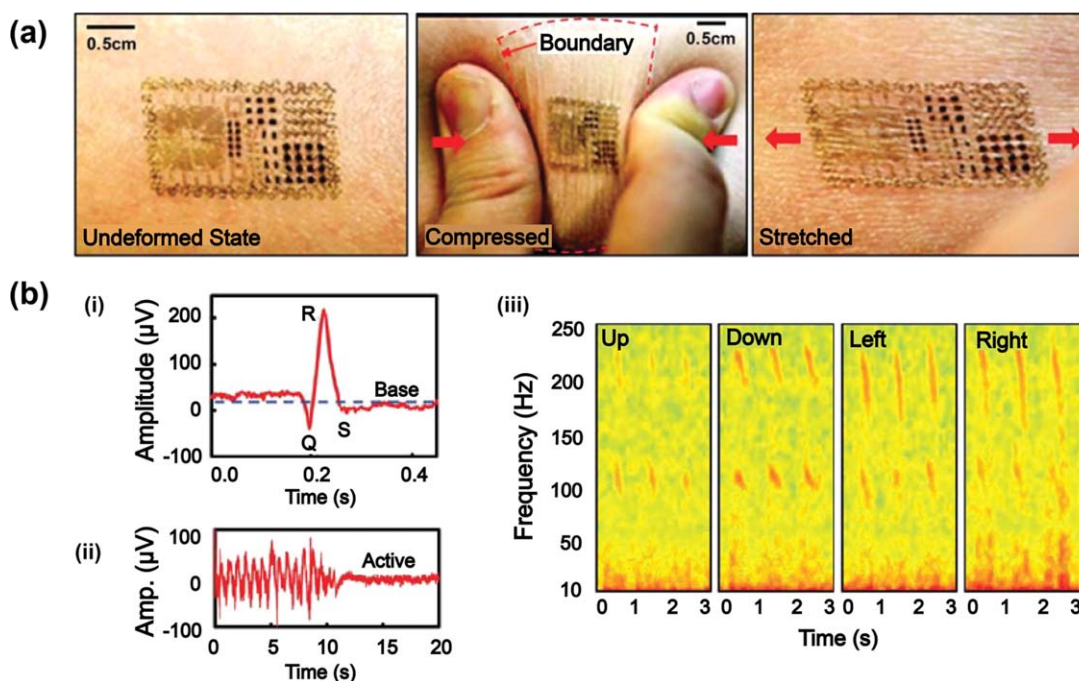


Figure 6. EES. (a) Images of conformal attachment on the epidermis under compressed or stretched conditions. (b) Measured EEG signals by the attachment of EES on the chest: (i) EMG signals measured on the (ii) right leg and (iii) neck. (Reprinted with permission from ref. 24. Copyright 2011 The American Association for Advancement of Science.) Q, R, and S are part of the QRS complex (Q: Q wave, R: R wave, S: S wave). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

for example, unique patterns of beating frequency and level of blood pressure.⁷

To enhance the adaptability on a rough skin surface, the same group also developed a dry adhesive patch for *in vitro* diagnostics.^{8,9} To this end, they exploited the unique structural characteristics of gecko foot hair; the resulting structures were high-density micropillars with a bulged tip to maximize normal and shear adhesion, as shown in Figure 5.^{8,9} It turned out that such mushroom-shaped pillars made of soft PDMS were less affected by surface contamination, oxidation, and other environmental factors than conventional acrylic adhesives and provided better long-term biocompatibility [Figure 5(a–ii)].⁸ To strengthen the mechanical stability, modulus-tunable composite micropillars made of stiff and soft PDMS materials were used, as shown in Figure 5(b).⁹ This dry adhesive was used as a fixation unit to monitor an electrocardiogram (ECG) for a time period of 48 hours on two locations of a volunteer's skin (chest and wrist), as shown in Figure 5(b–iii).⁹ Although such a skin patch was still difficult to use under highly dynamic conditions, such in running, no side effects such as an allergic reaction, redness, or skin damage were observed in most daily activities (e.g., walking, sitting, sleeping).

A striking platform for reading various physiological signals with epidermal electronic systems (EES) was introduced by Rogers et al.²⁴ through the integration of a collection of multiple components, including transistors, light-emitting diodes, circuit elements, wireless power coils, and radio-frequency communication modules to establish a multifunctional biosignal detection system. The silicon-based electric circuits on an ultra-

thin polyethylene (PE) layer were fabricated by a transfer technique with a high-quality inorganic semiconductor/metal arrays, and a water-dissolvable PVA sheet was used as a temporary substrate for easy handling. This ultrathin structure maintained a tattoo-like, conformal attachment on the epidermis even under an extremely bumpy state because of its low effective modulus and minimized deformation-induced elastic energy [Figure 6(a)].^{22,24}

The same group also demonstrated the measurements of various biosignals with lamination onto the surface of a target position, including electroencephalograms (EEGs) from the brain, ECGs from the heart [Figure 6(b–i)], and electromyograms (EMGs) from muscle tissue [Figure 6(b–ii)]. Here, the device was attached to the chest without a conductive gel and showed high-quality signals with information on all phases of the heart rate, including the rapid depolarization of the cardiac wave.²⁴ In particular, EMGs on the neck during vocalization were measured with four different words: *up*, *down*, *left*, and *right* [Figure 6(b–iii)]. Despite such remarkable performance from the device, there is still room for improvements, including repeatable attachment and enhanced air permeability for long-term fixations. A reversible interconnection system between EES and data acquisition could be one possible solution.⁷⁹

IMPLANTABLE DEVICES

An implantable device, which can be attached directly to an internal organ, is considered as an alternative clinical system for monitoring electric signals from the inner body, including electrocorticographic signals and epicardial electrogram

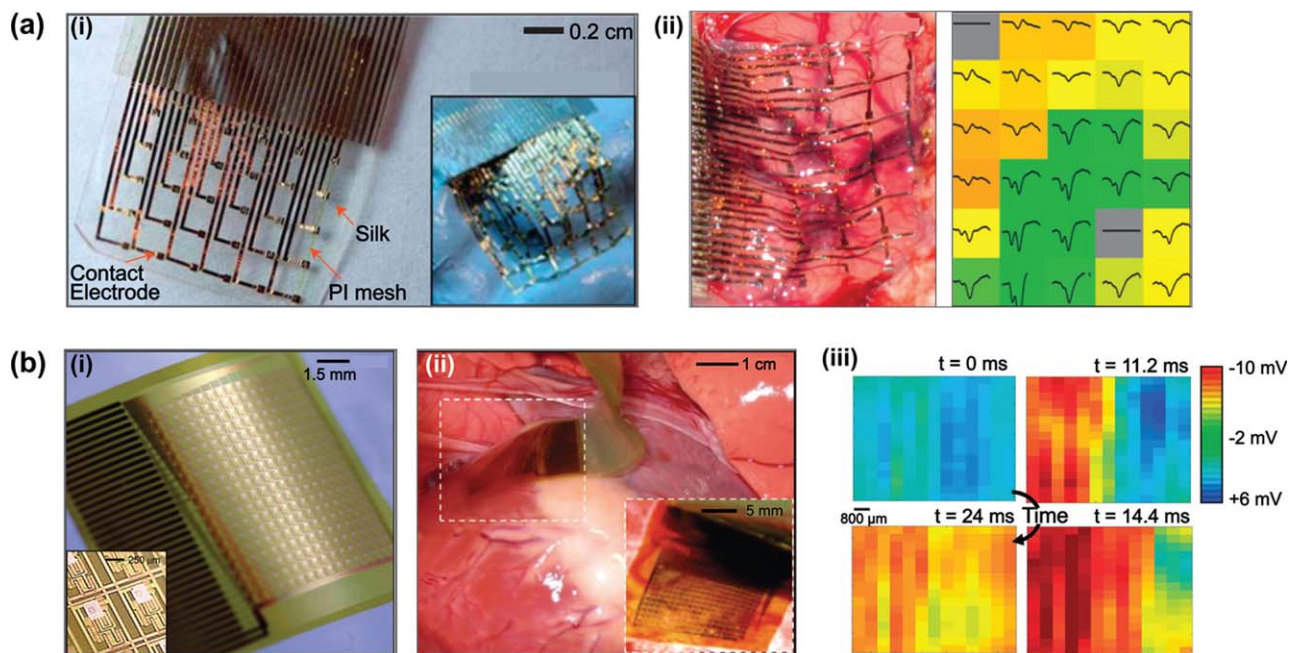


Figure 7. Thin, flexible electrode array for mapping signals on an organ surface: (a) dissolvable films for bio-integrated electronics in with (i) images of electrodes on a silk substrate and lamination on a brain model (inset) and (ii) a meshed electrode array on the brain and measurement of signals from a cat brain. (Reprinted with permission from ref. 10. Copyright 2010 Nature Publishing Group.) (b) Bio-interfaced electronics for cardiac electrophysiology with images of (i) an electrophysiology mapping device, (ii) adaptation on the porcine heart, and (iii) high-resolution maps of a normal cardiac wave. (Reprinted with permission from ref. 11. Copyright 2010 The American Association for the Advancement of Science.) t: time. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

signals.^{10,11,25} It seems that conventional devices based on microelectrode arrays have potential limitations, including insufficient spatial density and the possibility of damaging adjacent soft tissue with their intrinsic rigidity.^{80–84} Hence, a minimally invasive and nonpenetrating measurement system is required for *in vivo* diagnostic and therapeutic devices with the capability of having large-area coverage and providing high performance and sufficient spatial resolution.

Recently, one solution was proposed by Rogers et al.¹⁰ with an ultrathin (2.5 μm) inorganic electrode array on a meshed PI substrate, which demonstrated the capability of making conformal contact on the brain of a cat (feline) due to its low bending stiffness as shown in Figure 7(a–i). Here, an absorbable silk fibroin substrate was laminated on the device as a sacrificial layer, which was dissolved in biofluid in less than an hour. Then, the gapless attachment between the device and highly uneven brain surface *in vivo* was demonstrated. Subsequently, a mechanical visual stimulus evoked the real-time mapping of physiological signals with spatial distribution [Figure 7(a–ii)]. Later on, a more integrated system with a multiplexed active transistor array was implemented to maintain the high spatial resolution of the mapping of the brain signals.⁸⁵

In the case of the heart, monitoring electric activity seems critical to diagnosing and treating heart disease, such as cardiac arrhythmias.^{11,53} However, a conventional catheter with a single- or few-point measurement system is insufficient to infer cardiac electric signals for diagnostics at a high resolution and in real time.^{86,87} Therefore, the integration of stretchable and multi-

functional circuits on the surface of a balloon catheter could be a suitable solution for cardiac therapy.⁸⁸ Also, a bio-integrated system with a multilayered active circuit array or a multifunctional web could enable epicardial signal mapping in a minimally invasive manner, as shown in Figure 7(b–i,ii).^{11,59} In such an amplified and multiplexed circuitry, the PI and epoxy (SU-8) thin layers play a role as dielectric layers, preventing mechanical failure even under a highly bent and stretched condition. With the proper encapsulation and mechanical stability, an *in vivo* experiment successfully recorded the voltage data of cardiac activation with 288 measurement points; this showed the spatial movement of cardiac pacing signals with a current of 10 mA and a pulse width of 2 ms [Figure 7(b–iii)].

SOME UNIQUE SENSOR SYSTEMS

To expand the realm of flexible sensing devices, several innovative concepts have been combined with the current sensing platforms. Examples include transparent,¹ self-energy-harvesting,² and self-healing devices.⁴⁷ To achieve these goals, the integration of an additional function is required with new material properties and processing. First, a transparent and skinlike pressure sensor was demonstrated by Bao et al.¹ with PDMS elastic films of CNTs. First, a CNT solution was spray-coated onto a UV/ozone-treated PDMS sheet through a stencil mask, and the morphology of the CNT bundles was tailored by the amount of external strain. Here, compressible capacitors were fabricated with a silicon elastomer (Ecoflex), as shown in Figure 8(a–i). It turned out that the capacitance was linearly dependent on the

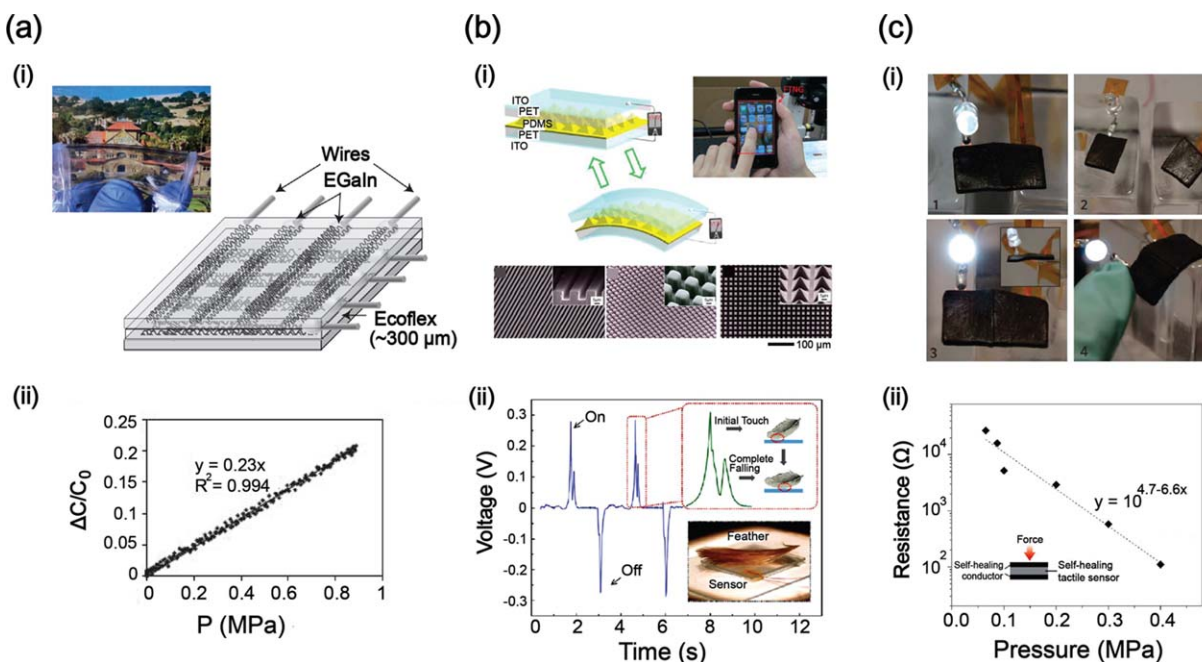


Figure 8. Advanced concepts of flexible sensors. (a) Highly transparent pressure sensor with aligned CNTs with (i) a schematic illustration and image of a sensor and (ii) the capacitance change under external pressure. (Reprinted from ref. 1. Copyright 2011 Nature Publishing Group.) (b) Transparent self-powered pressure sensor with illustrations of (i) the device with embedded microstructures and (ii) sensing of a tiny pressure with a feather. (Reprinted with permission from ref. 2. Copyright 2012 American Chemical Society.) (c) Mechanically and electrically self-healing sensor with (i) a demonstration of a self-healing procedure and (ii) linear responses of resistance changes under compression. (Reprinted with permission from ref. 47. Copyright 2012 Nature Publishing Group.) P: pressure. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

pressure from about 50 kPa to 1 MPa with high reversibility by programmable elastic networks [Figure 8(a–ii)].

A novel, flexible transparent nanogenerator was reported by Wang et al.² on the basis of the triboelectric effect. The device could be fabricated by the simple assembly of a PDMS microstructure and a PET/ITO substrate without pixel arrays, and it was operated by a capacitance change via frictional touch. Here, the efficiency of the device was optimized with micropyramid structures between two flexible electric substrates after various structures (flat, microline, microcubic, and micropyramid shape) were investigated, as shown in Figure 8(b–i).² This device displayed an effective power of about 18 V (output voltage) and 0.13 $\mu\text{A}/\text{cm}^2$ (current density). Also, the device's sensitivity was shown to be as high as 0.4 Pa; this was equivalent to a contact pressure of a falling feather [Figure 8(b–ii)]. Interestingly, although the micropyramid embedded device showed the best efficiency, the transmittance of visible light was decreased to the lowest value ($\sim 45\%$) among the previous various embedded structures because of its grating or light trapping effect (flat PET $\approx 85\%$).²

More recently, the idea of a self-repairing sensor was introduced with a composite material that was composed of a supramolecular organic polymer with embedded nickel (Ni) nanostructures.⁴⁷ The flexible device of self-healing at room temperature was fabricated by the mixture and compression molding of a conducting polymer with micro-Ni composites. These mechanical and electrical self-healing properties were unique and similar to the remarkable ability of human skin and shed light on bio-

prosthetic and emerging soft-robotic applications. Here, the repeatable and room-temperature operation of self-healing sensors was demonstrated with a high conductivity ($\sim 90\%$ restored) after a 15-s healing process, as shown in Figure 8(c–i).⁴⁷ Also, the mechanical properties were completely restored after about 10 min. The electrical properties of the sensor were tunable through changes in the portion of $\mu\text{-Ni}$ particles as high as 40 S/cm. In addition, the self-repairable sensor could detect the bending angle ($< 80^\circ$) and pressure (0.05–0.4 MPa), as shown in Figure 8(c–ii).⁴⁷

CONCLUSIONS AND OUTLOOK

In this review, we have highlighted recent advances in flexible mechanical and electrical sensing devices with a particular focus on the properties and functions of polymer layers in four representative areas (e-skins, wearable devices, implantable devices, and advanced sensors with additional functions, e.g., transparency, self-power, and self-healing). A number of key features have been identified as the advantages of polymers toward the development of a highly sensitive and multifunctional sensor: bendability/stretchability, self-programming ability, biocompatibility, elasticity, and low-cost processing with a geometrically controllable shape.

In terms of the choice of materials, PI is mainly used as a base substrate and an insulating layer because of its high thermal and chemical resistance and mechanical robustness. These properties would be particularly useful when combined with conventional semiconductor fabrication processes (e.g., chemical vapor

deposition, oxidation, chemical etching). PDMS is also widely used because of its ease of use and highly stretchable features. Nonetheless, PDMS would be limited in high-resolution patterning with a feature size less than approximately 1 μm if the aspect ratio was larger than unity. For other polymers, a proper selection guide is required on the basis of the processing conditions and material properties.

Another notable point is that the polymer is broadening its role in flexible devices from a simple flexible support to an active, functional layer to facilitate reliable attachment on the target surface or to transfer various mechanical and electrical inputs. To this end, a variety of structures in the forms of mesh, webs, high-aspect-ratio nanopillars, and pyramidal or mushroom-shaped microstructures are required; this can also benefit from the facile and versatile processing methods of polymer materials.

For further development and widespread use of the sensing devices, a number of issues are yet to be solved. For e-skins, an organic thin-film transistor based device is frequently used because of its inexpensive processing and high flexibility. However, several challenges need to be solved in views of robustness and power efficiency. In contrast, inorganic NW circuits for e-skin possess a high power efficiency, mobility, and reliability, but a cost-effective fabrication method with a high yield is necessary for further development. Also, transistor-free devices (e.g., fiber interlocking-based sensors) have been recently introduced as an alternative platform, but the ability to map the spatial pressure distribution with a high resolution should be implemented.

For wearable and skin-attachable applications, better adaptability on skin and organ interfaces needs to be investigated with biocompatible materials in highly integrated systems. Also, the bio-interface between human and device should be elaborated for a reliable transfer of vital or mechanical signals to the sensor matrix with minimal noise and leak signals. Finally, new concepts and applications need to be pursued in combination with recently introduced advanced sensors. At present, there are some limitations toward self-powered systems for which a wireless transmitting component would be one promising solution for supplying power along with the communication to the device.

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