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STATE-OF-THE-FIELD REVIEW

Flexible and Stretchable Electronics Paving the Way for Soft Robotics

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Abstract

Planar and rigid wafer-based electronics are intrinsically incompatible with curvilinear and deformable organisms. Recent development of organic and inorganic flexible and stretchable electronics enabled sensing, stimulation, and actuation of/for soft biological and artificial entities. This review summarizes the enabling technologies of soft sensors and actuators, as well as power sources based on flexible and stretchable electronics. Examples include artificial electronic skins, wearable biosensors and stimulators, electronics-enabled programmable soft actuators, and mechanically compliant power sources. Their potential applications in soft robotics are illustrated in the framework of a five-step human–robot interaction loop. Outlooks of future directions and challenges are provided at the end.

Introduction

Research on flexible electronics started almost 20 years ago[1,2] with the demand of macroelectronics,[3] such as paperlike flexible displays.[4,5] Organic semiconductors and conducting polymers were appealing materials for large-area electronics attributing to their intrinsic flexibility, light weight, and low cost, especially when merged with the roll-to-roll processes.[6,7] Methods to synthesize novel organic materials and their printing, patterning, and passivation techniques[8,9] were later applied to manufacture artificial electronic skins (E-skins) for robotics[10,11] and organic solar cells.[12,13] As of today, flexible displays based on organic light-emitting diodes are nearing commercial reality.

The other branch of flexible and stretchable electronics based on high-quality monocrystalline inorganic semiconductors started to emerge in the mid-2000s.[14,15] Inorganic semiconductors exhibit high carrier mobility and excellent chemical stability under ambient environments.[14] Well-defined properties and well-established manufacturing processes make them even more appealing. Their intrinsic stiffness and brittleness, however, greatly hindered their application in flexible/stretchable electronics until the discovery of unconventional mechanical behaviors of low-dimensional inorganic materials (micro-/nanowires, ribbons, and membranes), when bonded to polymer substrates.[16–19] Two prevailing design strategies, that is, wrinkled nanoribbons/nanomembranes[20–22] and isolated device islands interconnected by noncoplanar[23–25] or serpentine-shaped metal wires,[26–28] were proven to be effective for minimizing strains in inorganic electronic materials. When substrate materials are stiff in-plane, but thin and flexible (e.g., paper, leather, fabric), compliant interlayers laminated in between the substrate and the active device islands are also very effective for strain isolation.[29,30] Microtransfer printing technology developed for single-crystal inorganic semiconductors[31–33] has enabled the integration of fully functional flexible electronics, including flexible complementary metal oxide silicon circuits,[34] flexible displays,[35] high-efficiency flexible solar cells,[33,36] and bioinspired electronic eye cameras.[24,37] Because high-quality semiconductors are used as the active components, the electronic performance and long-term reliability of these devices are on par with wafer-based electronics while flexibility is still incorporated.

Flexible and stretchable electronics found their destined application in the late 2000s when the concept of biointegrated electronics was proposed.[38] While organisms are soft, curvilinear, and deformable, wafer-based electronics are not. Novel electronic systems with matched form factors and mechanical properties with biotissues can help establish long-term, intimate bioelectronic interfaces. So far, biointegrated electronics have enabled exciting applications, including epidermal electronics for vital sign monitoring,[27,39,40] brain–computer

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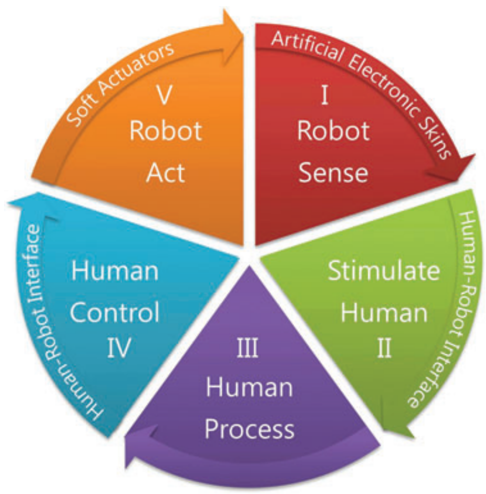

FIG. 1. Schematic diagram of the five-step human–robot interaction loop. Step I, the robot senses environmental stimuli (e.g., touch) through artificial electronic skins (E-skins). Step II, the robot passes the sensed environmental signal to the human through human–robot interface. Step III, the human processes the received stimulation. Step IV, the human sends out control signals through human–robot interface. Step V, robot (actuator) takes action according to the commands it received.

interfaces,[41,42] electrocardiogram mapping devices,[43,44] and smart or minimally invasive surgical tools.[26,45] More detailed materials and mechanics strategies for bioinspired and biointegrated electronics have been summarized in several recent review articles.[46–48]

Soft sensors and actuators enabled by flexible and stretchable electronics can offer transformational opportunities for soft robots.[49,50] One of the ultimate goals of modern robotics is their seamless interfacing with humans. Examples include prosthetic limbs and other types of wearable robots. In this review article, we limit our focus to noninvasive human–robot interactions. A five-step human–robot interaction loop is summarized in Figure 1. Starting from step I, the robot should be equipped with humanlike E-skins to *sense* the mechanical and thermal stimuli from the environment. The sensed signal should be transferred to humans by *stimulating* the human subject through human–robot interface, as illustrated in step II. Human subjects will *process* the received stimulation through their central nervous system, as illustrated in step III. A *control* signal will be passed from human to robot again through the human–robot interface, as illustrated in step IV. The robot will take mechanical *actions* through programmable soft actuators or the so-called artificial muscles following the control signal, as illustrated in step V. Once an action is taken by the robot, a new sensing–acting loop will be initiated. This review article is organized according to the five steps, with the following sections corresponding to steps I, II, IV, and V in order. Compliant power sources for soft robots as a special topic will be discussed in the section Stretchable Power Sources. Concluding remarks are provided in the last section.

Artificial E-Skin: Robot Sense

Flexible E-skins capable of tactile and temperature sensation are critical for humanlike robots or smart prosthetic limbs, which is highlighted as step I in Figure 1. It is nontrivial to mimic the human skin because it is highly deformable and can feel pressure and temperature with high sensitivity and spatial resolution. It can also differentiate applied normal pressure from its own bending/stretching. These challenges have been addressed by a variety of E-skins, as summarized in Figure 2.

Figure 2A shows the early version of large area, flexible E-skins.[10] The E-skin is composed of an $8\,\text{cm} \times 8\,\text{cm}$ pressure-sensitive rubber (PSR) integrated with an array of 16×16 organic field-effect transistors (OFET). It is bendable down to a 2 mm radius, and the resistance of PSR can drop by three orders of magnitude when subject to a pressure of 30 kPa, a typical pressure for human finger to grab an object.[51] A similar system is later made stretchable and conformable by using a perforated, that is, open mesh, active matrix, as shown in Figure 1B.[52] Rotation of each segment of the network can accommodate applied tensile strains up to 25%, which is comparable to the stretchability of human skin.[53] Thermal mapping was also achieved in this E-skin where organic diodes were fabricated and used as temperature sensors. E-skin shown in Figure 2C used a similar type of PSR as the pressure-sensing material but employed ordered inorganic semiconductor nanowires as transistors of the active matrix backplane.[54] Low carrier mobility of OFET demands an operating voltage of 20 V,[10] whereas inorganic nanowire-based FETs used in E-skins shown in Figure 2C are able to operate at voltages less than 5 V, indicating much lower energy consumption. The flexibility and robustness of nanowires were manifested by no performance degradation after 2000 bending cycles with a bending radius of 2.5 mm.

Aforementioned PSR-based pressure sensors are incapable of measuring pressure below 10 kPa and are susceptible to hysteresis.[55] Pressure sensors based on micro- and nano-patterned elastomers have demonstrated enhanced sensitivity and repeatability.[56,57] Figure 2D shows an ultrasensitive array of capacitive pressure sensors.[56] Microstructured elastomers can help minimize viscoelastic effects suffered by solid elastomers, resulting in a highly responsive and reversible system. This 8×8 flexible sensor array operated by OFET is capable of measuring the contact of a fly (3 Pa) with good repeatability. Nanostructured elastomers are also applied to create a multifunctional artificial skin that can detect pressure, shear, and torsion, respectively, as shown in Figure 2E.[57] The electrical resistance across two arrays of metal-coated polymer nanofibers placed face to face is very sensitive to the status of interlocking, which is, in turn, very sensitive to pressure, shear, and torsion applied to this bilayer. Detectable pressure can be as low as 500 Pa, and reproducible signals were obtained even after 10,000 loading–unloading cycles.

An outstanding challenge in E-skin is to decouple the pressure-induced signals from substrate deformation. This has been achieved by a recent development of tactile imaging arrays of piezotronic transistors made of vertical zinc oxide (ZnO) nanowires (Fig. 1F).[58] When the substrate is bent to adapt to a target curvature, the piezoelectric current is measured under this reference state. When a normal pressure is applied to the bent surface, a new current is recorded. Numerically subtracting the reference current from the new current gives rise to a spatial imaging of the additionally applied pressure, which was hard to resolve with past flexible tactile sensor arrays. Other strain decoupling strategies, such

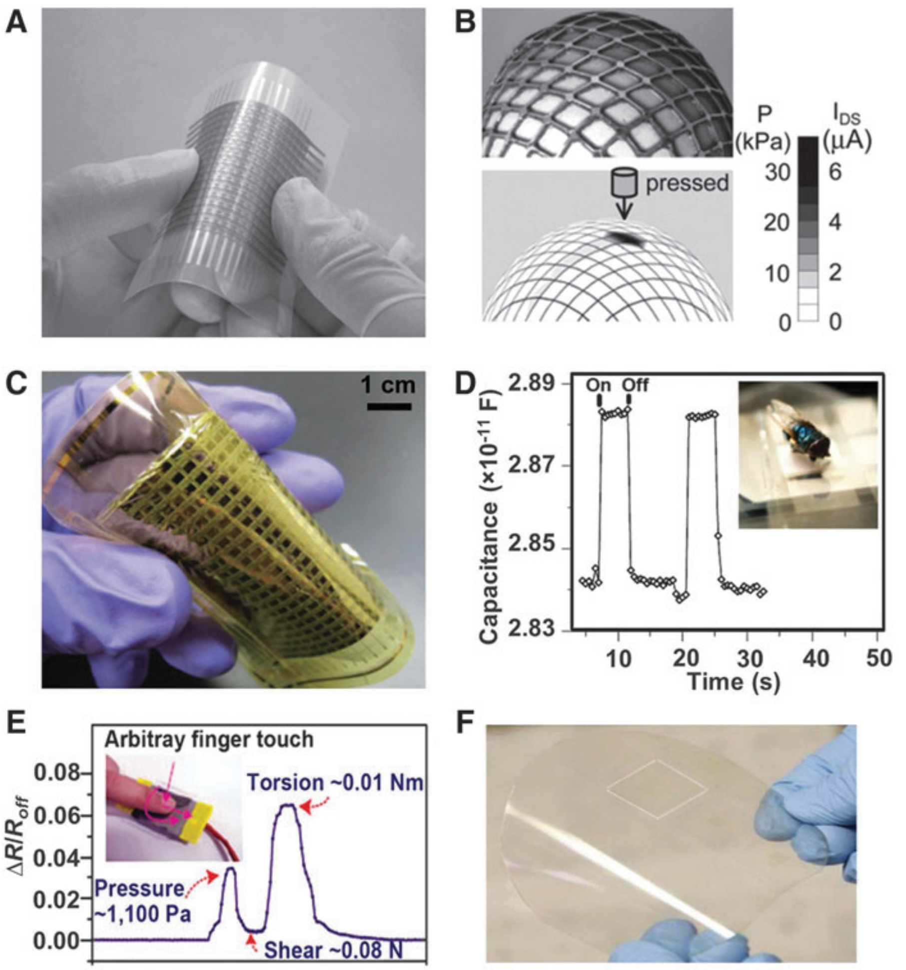

FIG. 2. Artificial E-skin capable of tactile sensation for step I in Figure 1. **(A)** Flexible E-skin made of pressure-sensitive rubber (PSR) integrated with organic field effect transistors (OFET) for multiplexing.[10] **(B)** Pressure-sensing matrix similar to **(A)**, made into open-mesh structure so that it can stretch and conform to 3D shapes such as an egg (upper frame) and map-applied pressure distribution (lower frame).[52] **(C)** A bendable E-skin based on PSR integrated with inorganic nanowire-array FETs.[54] **(D)** Highly sensitive capacitive pressure-sensor array operated by OFET capable of detecting the touch of a bluebottle fly (3 Pa).[56] **(E)** Flexible artificial skin based on interlocking nanofibers that can differentiate arbitrary pressure, shear, and torsion induced by a finger touch.[57] **(F)** Flexible array of tactile pixels (taxel) by virtue of vertical-nanowire piezoelectronic transistors.[58]

as stiff sensing pixels wired by compliant interconnections used in instrumented balloon catheters,[26] could potentially be applied for tactile sensor arrays as well.

Human–Robot Interface: Stimulate Human

E-skins paired with programmable tactile stimulators, either mechanical or electrical, could genuinely pass the touch sensation from a robot to a human, which is represented by step II in Figure 1. Tactile stimulation is a major means for the robot to send feedbacks to humans through human–robot interface (Fig. 1B). Figure 3 summaries three types of skin-interfaced tactile/electrotactile stimulators. Figure 3A shows a flexible, lightweight braille display.[59] Braille code, composed of a 2×3 dot pattern, is a well-established tactile writing system that can communicate information to the human finger in a simple and unambiguous manner. For the display shown in Figure 3A, each soft actuators (made of ionic metal polymer composites) can be turned on or off individually by an OFET. Hemispheres attached to the tip of each actuator can project upward to tap on human skin, enabling the transfer of information via the mechanical actuation. The limitation of this system is the relatively slow response time because of the charge and discharge of the actuator electrodes.

Figure 3B is another soft-actuator-enabled tactile display but is made to be wearable.[60] Dielectric elastomer as a common type of electroactive polymer (EAP) has been widely used in soft actuation. EAPs are intrinsically soft compared with inorganic piezoelectric materials and can change shapes in response to electrical stimulation, so EAPs are also referred to as artificial muscles. Advantages of EAPs also include fast response speeds, small hysteresis, strain levels far above those of traditional piezoelectric actuators, and elastic energy densities even higher than those of piezoceramics.[61,62] These merits make EAPs the most popular choice of material for soft actuation so far and have been discussed in several review articles.[63,64] More examples of EAPs will be mentioned later when we discuss soft actuators for robots taking mechanical actions. In the current example of human–robot interface, both the dielectric elastomer and the electrically conductive polymer electrodes are highly soft and deformable, enabling the shape adaptation and easy wearing of the tactile display.

Electrotactile stimulation could be acute and timely controllable by passing a properly modulated electrical current into the skin, which can excite cutaneous mechanoreceptors.[65] Figure 3C features a wearable finger tube that integrates high-performance, inorganic electronics for electrotactile stimulation.[66] A stretchable array of gold anode–cathode pairs multiplexed by silicon diodes has been transfer-printed on the inner surface of the finger tube. Despite the intrinsic stiffness and brittleness of inorganic electronic materials, the ultralow profiles of the devices and the serpentine-shaped interconnections have enabled conformal and intimate contact between the stimulating electrodes and human skin. Modulated

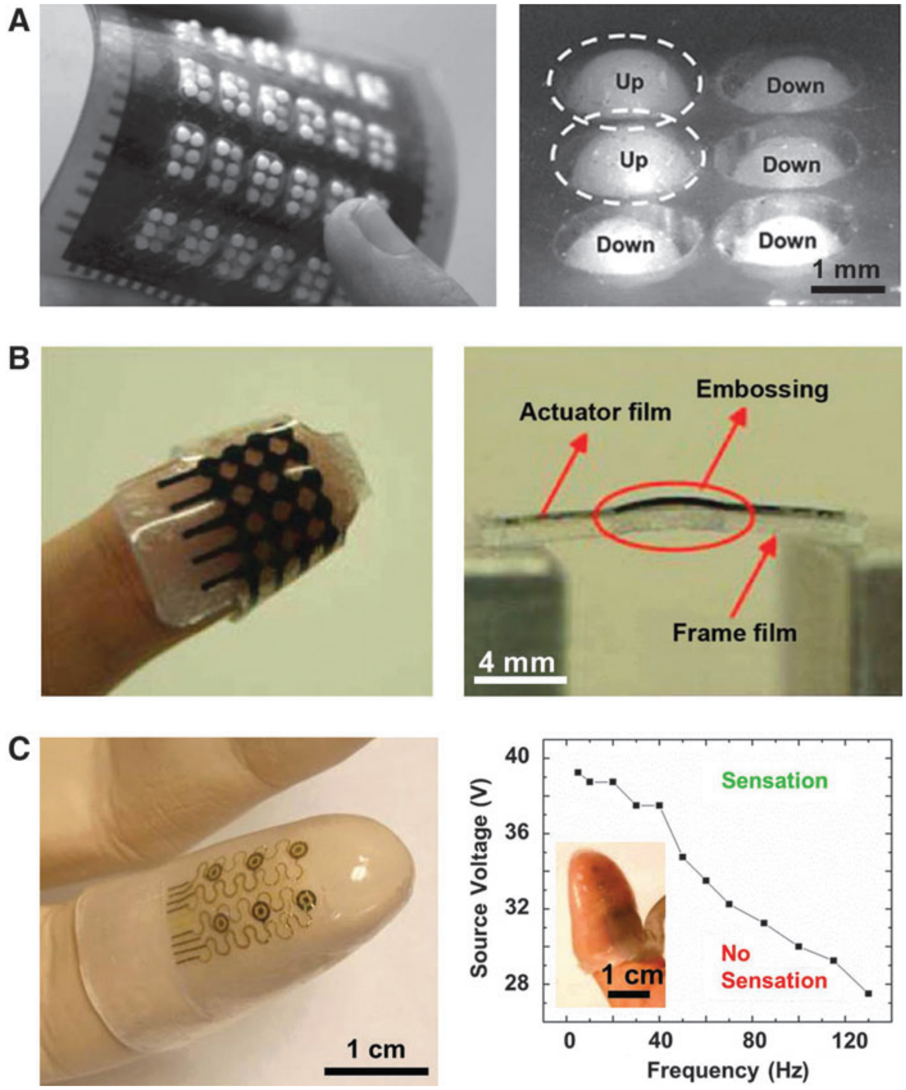

FIG. 3. Flexible and wearable tactile/electrotactile stimulators as part of the human–robot interface for step II in Figure 1. **(A)** A flexible Braille display sheet based on ionic polymer metal composite (IPMC) actuators and organic transistors.[59] **(B)** A wearable tactile display composed of dielectric elastomer actuators with carbon-based electrodes.[60] **(C)** A wearable, conformable finger tube equipped with electrotactile stimulators to generate tingling sensation with suitably modulated current.[66]

currents sent through each pair of electrodes can create a localized tingling feeling known as the electrotactile sensation. The voltage–frequency combination to enable sensible stimulation is provided in the right frame of Figure 3C. Detailed calibrations of voltage and frequency will need to be carried out to achieve pixel-to-pixel differentiation.

Human–Robot Interface: Human Control

As a two-way communicating channel, in addition to the stimulation of humans, the human–robot interface should also be able to sense biosignals from humans to control the robots, as illustrated by step IV in Figure 1. Human commands can be sent via a variety of biosignals, such as electroencephalogram (EEG), electromyogram (EMG), and mechanical motion, which can be readily measured through biointegrated sensors. Biointegrated electronics as an emerging area of flexible/stretchable electronics have been thoroughly reviewed recently,[46–48] and we just briefly capture the spirits here. Conventional EEG and electrocardiogram sensors are poorly suitable for long-term application outside of research labs or clinical settings because of rigid power and communication components, bulky wires, irritating adhesives, and so on.[67] Multifunctional electronic systems that are self-adhesive, conformable, and mechanically invisible to human skin have created new opportunities for studying disease states, improving surgical procedures, monitoring health/wellness, establishing human–machine interfaces, and many others. In this review, we will just focus on their sensing capabilities.

Figure 4 illustrates three measurements carried out by skin-integrated electronics. Figure 4A demonstrates EEG measurements with epidermal electronic systems laminated on the human forehead, in a manner that is mechanically invisible to the user, much like a temporary transfer tattoo.[27] Since attachment is purely enabled by van der Waals force and hence does not require conductive gels, these systems can function for a prolonged period. The bottom frame presents a spectrogram showing the alpha rhythm that corresponds to eye opening and blinking events. With this technology, mind control of wearable robotics is getting closer to reality. Figure 4B shows a Sokoban game controlled by neck EMG measured with epidermal electronic systems.[27] When a human subject speaks "up," "down," "left," and "right," the corresponding neck EMG can be uniquely identified through pattern recognition. The identified commands will be used to move a box in the video game from red locations to green locations. Such

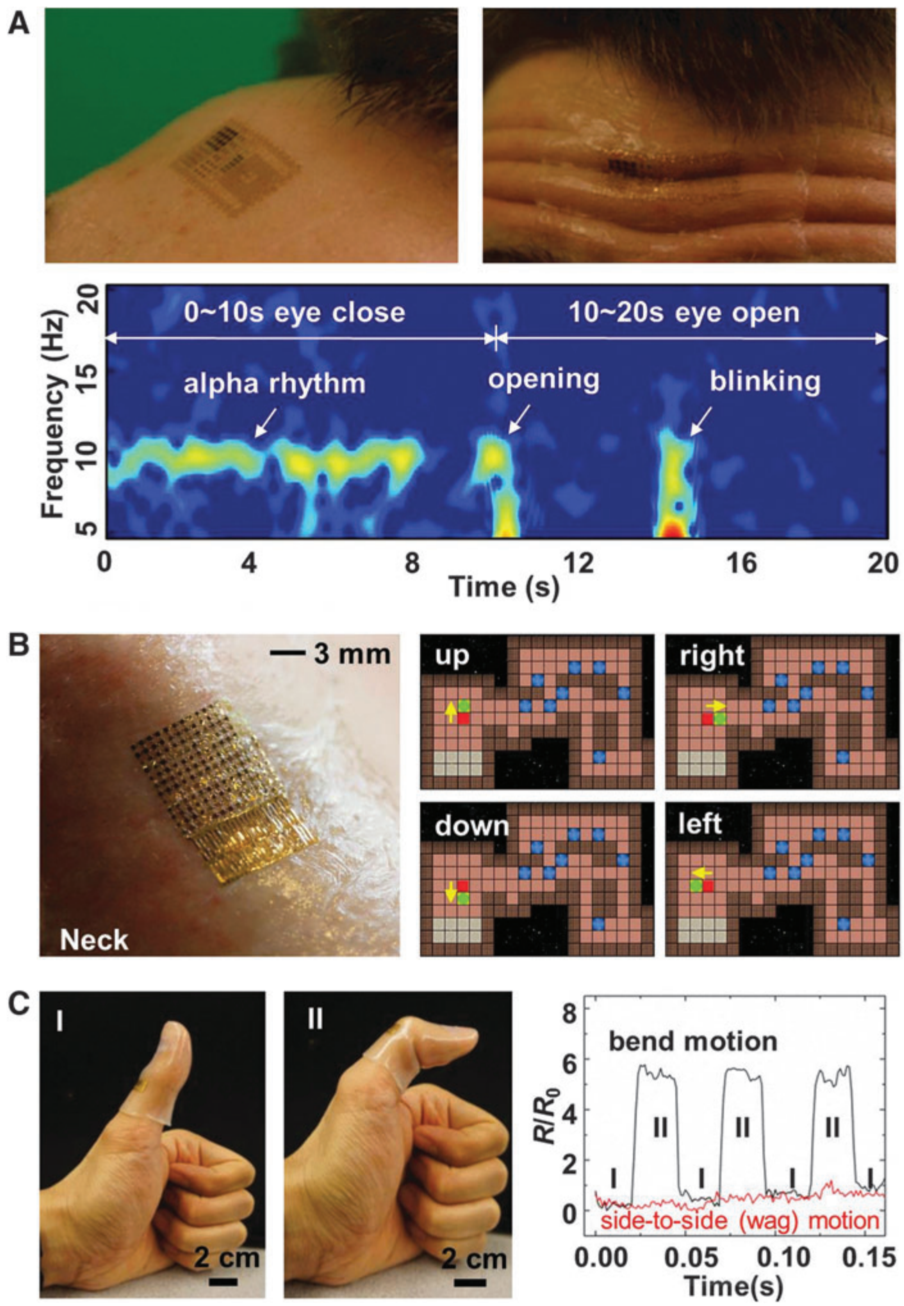

FIG. 4. Biointegrated sensors as part of the human–robot interface for step IV in Figure 1. **(A)** Ultrathin, ultrasoft epidermal electronic system (EES) laminated on the human forehead (upper frame) to read a human EEG (lower frame).[27] **(B)** A similar EES attached to the human neck measures neck EMG to control a video game.[27] **(C)** Stretchable strain gauges integrated on the wearable finger to detect the bending motion of human fingers.[66]

technology is beneficial to people who have lost speaking capability to, for example, a laryngectomy, or in circumstances when vocal communication is not possible. Figure 4C illustrates the bending movements of the human finger measured by highly deformable strain gauges integrated on a smart finger tube 66. The right frame demonstrates that the strain gauges can be arranged in a way that is only responsive to finger bending, not wagging side to side. Unambiguous sensing of human signals is the foundation for accurate control of wearable robots.

Soft Actuators: Robot Act

Making robots to act as commanded is denoted as step V in Figure 1. A fully autonomous, mission-capable, humanmimetic robot has been a Holy Grail of roboticists. The development of such machines has been mostly hampered by the absence of actuation and power supply technologies that are comparable with even some of the simplest systems found in nature. Biological organisms have a great advantage over artificial mechanical systems in that muscle, nature's actuator of choice, has a favorable force-to-weight ratio and requires low levels of activation energy. The most frequently used actuators, electric motors and pneumatic/hydraulic cylinders, are far from equivalent to their biological counterparts. Compared with pneumatically/hydraulically driven actuators, soft active materials that are responsive to electrical, thermal, or optical cues can be readily controlled by integrated electronics or optics. Emerging research on soft active materials controlled by highly compliant and stretchable

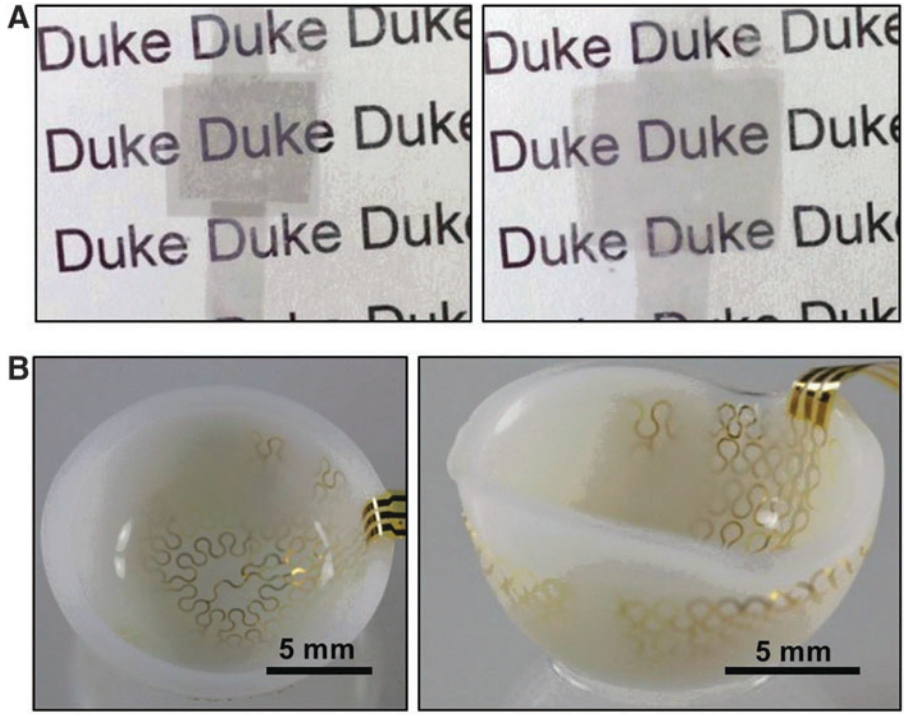

FIG. 5. Programmed motion enabled by integrating stretchable electronics with soft active materials for step V in Figure 1. **(A)** Dielectric elastomer being actuated by highly crumpled and stretchable graphene nanomembrane.[68] **(B)** Temperature-sensitive hydrogels being locally actuated by compliant electrodes working as microheaters.[72]

electronics start to demonstrate electrically programmable soft actuators capable of localized motion control. We have already introduced programmable soft actuation when we discussed tactile displays for human stimulation. Here in Figure 5, we will highlight two of the most recent efforts in this category.

Figure 5A features a soft actuator based on EAPs, similar to the ones used in the tactile display (Fig. 3B).[68] Figure 5A represents the advancement in stretchable electrodes for EAPs. Crumpled graphene electrodes are obtained through the relaxation of extremely high prestrains (450%) of the dielectric elastomer, a popular type of EAP as mentioned before. Highly stretchable electrodes have enabled the repeated area shrinking and expanding by over 100% in the dielectric elastomer. However, these polymers also require a high electrical field (>70 V/μm) to generate such high elastic energy densities (>0.1 J/cm^{-3}), making it difficult to safely integrate them in wearable robots. The development of stretchable and reliable passivation/insulating layers will be very helpful.

A softer and more biocompatible candidate is hydrogel, which is in general highly biocompatible and can repeatedly change volume by absorbing or expelling solvents when subject to stimulations, including heat, light, pH, humidity, and electrical fields.[69–71] Figure 5B features an electronically programmable two- and three-dimensional hydrogel structure.[72] To preserve the softness and deformability of hydrogels, ultradeformable, individually addressable electrodes are embedded in the hydrogel matrix as microheaters for reversible and nonuniform actuation.

Other soft active materials such as ferroelectric polymers[73] and shape-memory polymers[74] have also been made as soft mechanical actuators, but the integration of flexible electronics to those materials is still largely lacking.

Stretchable Power Sources

So far, the power source of the actuators often provides the greatest constraint on a robot's potential capabilities. Many important energy storage devices have been developed with flexible characteristics, including supercapacitors[75–77] and batteries.[77,78] In terms of energy-harvesting devices, flexible solar cells[33,79] and flexible nanogenerators[80,81] have experienced vast growth in the past decade. In particular, flexible solar cells were commercialized as early as 2008. Nevertheless, aforementioned bendable systems usually exhibit low stretchability and high stiffness in-plane, which prevents them from being mechanically compatible with soft actuators, especially the ones to perform large stretch or to conform to 3D curvilinear surfaces. Soft robotics will benefit significantly from the development of stretchable and compliant power sources, with both energy storage and harvesting capabilities, as summarized in Figure 6.

The stretchable alkaline battery as presented in Figure 6A is one of the early efforts toward soft energy storage systems.[82] In this example, compliant and stretchable conductive fabrics are embedded with manganese dioxide (MnO_2) and zinc (Zn) inks, respectively. In Figure 6A, MnO_2 (left, darker strip) and Zn (right, lighter strip) electrodes are mounted on a transparent and stretchable polyacrylic-acid-based polymer gel electrolyte. This cell has an open circuit potential of 1.5 V and a capacity of 3.875 mAh/cm^2, and is stretchable up to 100%. The stretchability and capacity records were later broken by stretchable lithium ion batteries as shown in Figure 6B.[28] Each molded microbattery cell contains a cathode of $LiCoO_2$ slurry, an anode of $Li_4Ti_5O_{12}$ slurry, and their respective electrodes. A silicone spacer filled with gel electrolyte is inserted between cathodes and anodes to provide a liquid-rich media for ionic transport. By virtue of "self-similar" metallic interconnects,

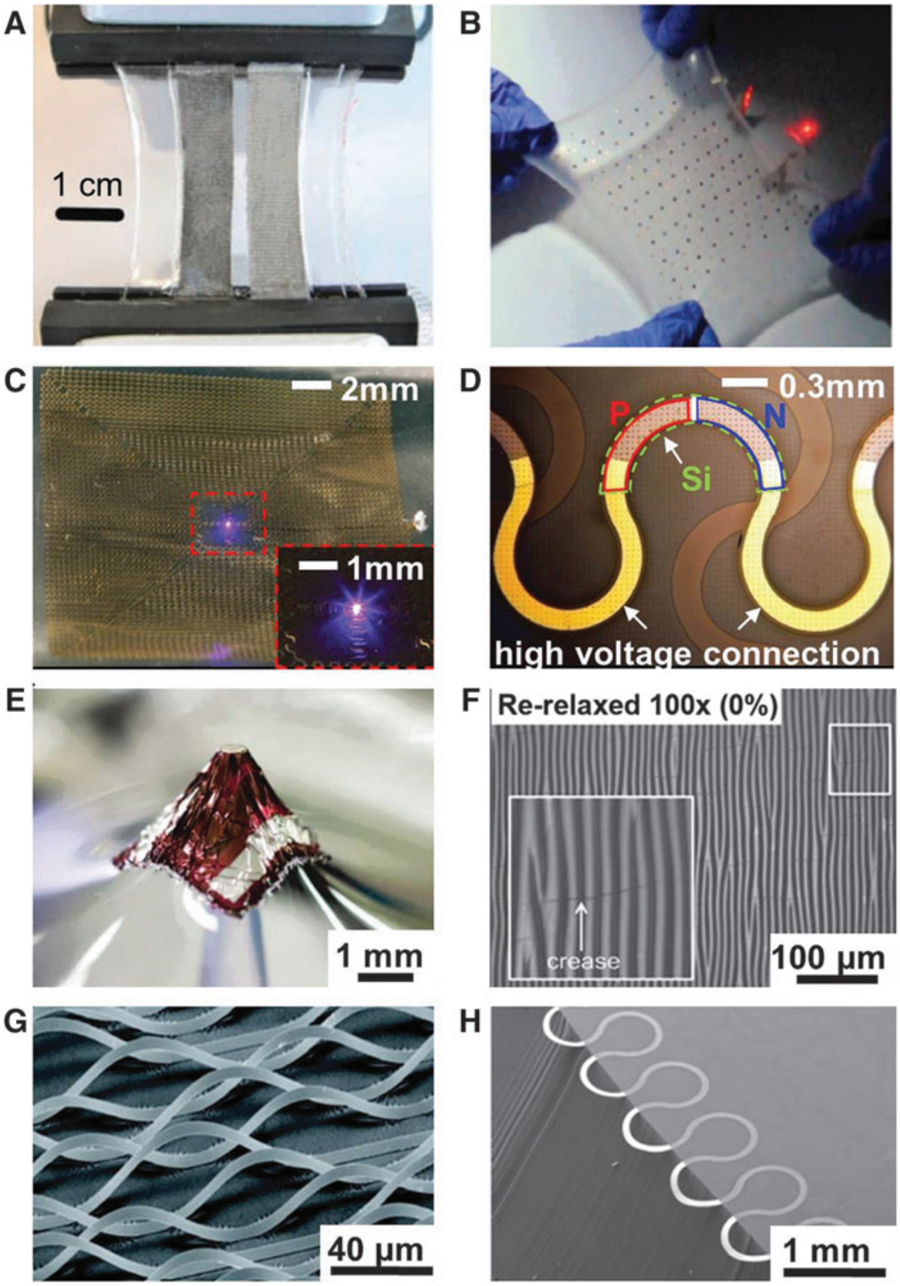

FIG. 6. Stretchable and compliant power sources. **(A)** The early effort of stretchable battery with electrochemically active materials embedded in compliant conductive fabrics.[82] **(B)** Array of lithium-ion battery microcells interconnected by "self-similar" serpentine electrodes with stretchability up to 300%.[28] **(C)** Serpentine-shaped stretchable inductive coils enabling wireless power transmission.[27] **(D)** Serpentine-shaped stretchable solar cells based on single crystal silicon nanomembrane.[27] Panels **(E)**[13] and **(F)**[12] depict stretchable solar cells based on wrinkled organic photovoltaic thin films on elastomer substrates. **(G)** Stretchable piezoelectric nanogenerator based on out-of-plane buckled lead zirconate titanate (PZT) nanoribbons.[87] **(H)** Stretchable piezoelectric nanogenerator based on zinc oxide (ZnO) nanoribbons forming in-plane serpentines on elastomers.[90]

the battery system can be repeatedly stretched up to 300%, while maintaining capacity densities of 11.1 mAh/cm^2.

The power-to-weight ratio is one of the key metrics of robot actuators. Biological muscles are reported to have power-to-weight ratios as high as 250 W/kg.[83] To maximize the power-to-weight ratio of energy storage modules on soft actuators, energy-harvesting components with a high power-to-weight ratio are very helpful. Popular energy-harvesting mechanisms include wireless transmission, photovoltaics, and piezoelectrics, as demonstrated in Figure 6C–H. Figure 6C shows a microscale light-emitting diode wirelessly powered by a stretchable coil through inductive coupling.[27] The coil is made of filamentary serpentine-shaped wires for compliance and comformability. Figure 6D–F highlights three types of stretchable solar cells. Stretchable solar cells can be made out of serpentine-shaped single-crystalline silicon ribbons, as shown in Figure 6D.[27] Solar cells comprised of organic semiconductors are potentially low-cost alternatives to devices made of crystalline silicon and other thin-film materials.[84] Figure 6E[12] and 6F[12] are two examples of compliant solar cells based on wrinkled thin films of organic solar cells. Although there is still a long way to go to overcome the inferior performance and environmental stability of organic semiconductors, the light weight of organic solar cells could yield a power-to-weight ratio as high as 10,000 W/kg with an energy conversion efficiency of just 4% (Fig. 6E[13]).

Compliant piezoelectric generators can be used to harvest energy out of routine mechanical motions of organisms or robots.[80,85–87] As a well-known piezoelectric polymer,[88] polyvinylidene fluoride has not been made into stretchable energy-harvesting devices yet. Figure 6G and H are two examples of stretchable piezoelectric generators, based on lead zirconate titanate (PZT) and ZnO nanoribbons, respectively. Figure 6G shows PZT nanoribbons with sinusoidal out-of-plane buckling on elastomer substrates after the relaxation of a prestrain of 8%.[87] More interestingly, local probing of the buckled ribbons reveals an enhancement in the piezoelectric effect of up to 70%, which is attributed to the flexoelectric effect of PZT nanoribbons, as reviewed in the literature.[89] Another popular strategy of stretchable electronics has been applied to piezoelectric ZnO nanoribbons, as shown in Figure 6H.[90] Serpentine ZnO nanoribbons are printed toward the edge of an elastomer with an overhang to accommodate tensile strains up to 30%. This structure minimized the strain effects on its natural frequencies in the audio frequency range, suggesting a capability of very stable energy harvesting even under large deformations.

Conclusions

In the past decade, flexible and stretchable electronics have demonstrated increasing impact in areas of sensing, actuation, and human–machine interface. New materials, mechanics principles, and microfabrication methods are the driving forces for the development of flexible and stretchable electronics. The intrinsic deformability, light weight, and low processing cost of organic electronics have enabled their application in large-area devices, including artificial E-skins and solar cells. While organic semiconductors have limitations in performances and reliability under wet and oxygen-rich environments, ultrathin, high-performance inorganic electronics have found their niche in biointegrated electronics. Soft robotics is a subject that can thoroughly enjoy the advancement of flexible and stretchable electronics, from artificial e-skins to biointegrated electronics and to compliant power sources. Within the framework of a five-step human–robot interaction loop, we have employed lots of examples to demonstrate how flexible/stretchable electronics can serve as building blocks in soft robotics. As we can see, while active research is being carried out in flexible/stretchable and even biointegrated sensors, programmable soft actuators for human stimulation or artificial muscles are still in their infancy. New opportunities are wide open for flexible/stretchable electronics to make more contributions to soft robotics.

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Author Disclosure Statement

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