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Skin-Patchable Electrodes for Biosensor Applications: A Review

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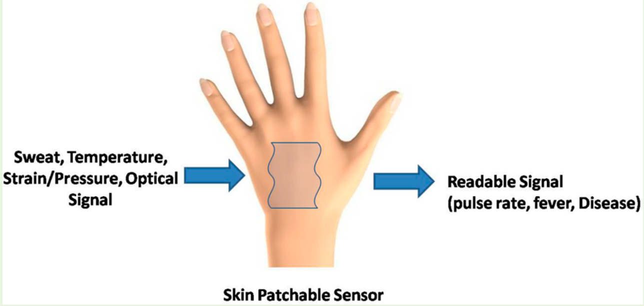
ABSTRACT: Health care monitoring is an extremely important aspect of human life that can be accomplished using wearable skin-patchable sensors. Upon interfacing with the skin or epidermal surface of the body, the sensing patches can monitor the movements of human parts such joints, legs, and fingers as well as tiny vibrations caused by respiration, blood flow, and heart beat. Wearable skin patches have shown improved promise in monitoring the body temperature and fever in addition to quick measurement of blood pressure and pulse rate along with breathing rate. Sensors can also analyze the sweat contents when in contact with the skin as well as other analytes such as diabetes-based volatile organic compounds (VOCs) and organophosphate nerve stimulating agents. Hence, the sensors can be of immense help in the early prediction of malfunctions of the body organs such as heart and lungs, leading to timely and effective treatment. This review covers different important aspects of skin-patchable sensors including mechanical strength and flexibility, sensitivity, transparency, self-healing, self-cleaning, and self-powering ability as well as their latest applications in medical technology.



KEYWORDS: *sensors, wearable, skin patchable, biomedical measurement, bioelectronics, health monitoring*

1. INTRODUCTION

Real-time health care monitoring is quite useful in the early prediction and treatment of various diseases.[1] With the advances in portable devices, thin, flexible, and wearable skin-patchable electrodes have gained considerable attention.[2−4] These can be very useful in monitoring the daily physiological problems related to human health,[3,5−7] leading to increased interest in developing next-generation biosensors offering a high flexibility.[8−13] There is also a need for a separate and flexible energy source to power such devices, such as charging and replacing, and charging heavy devices like batteries may be an obstacle for further development.[9,12,14−18] In order to fabricate fully flexible wearable biosensors, it is necessary that all the components must have high mechanical strength, especially the electrodes, as these can transmit body signals to an external circuit.[4,19−27] The development of wearable biosensors therefore requires interfacing the biomaterials and electronic components by assembling them onto a flexible and thin substrate, which can transform the biological interactions to readable electronic signals.[19,28−31]

Conventional biosensors, which are based on electrochemical interactions among the biomaterials and the analytes, are some of the earliest and more common types of devices.[20,32−34] The wearable sensors (in the form of wristbands and watches) may not only offer a more convenient monitoring of some of the critical parameters such as heart beat and blood pressure[35−38] but also allow noninvasive analysis of some important biochemical markers through sweat, saliva, tears, and interstitial fluids (ISF).[14,28,39−42] Thus, the noninvasive diagnosis with the help of these biofluids could provide more accurate health and fitness information.[28,43,44] Traditional analytical techniques require few point contacts that rely on flat electrode pads, which are kept in contact with the skin via adhesive tapes and sometimes with conductive gels that are applied to minimize the contact impedance between the skin and the electrode.[6,34,45,46] However, these suffer from a loss of adhesion and discomfort arising from the unfavorable nature of the skin−electrode interface.

The present review covers the developments on flexible and wearable skin-patchable electrodes used in the fabrication of wearable biosensors. A very typical approach to monitor human activity via wearable sensors is to measure the strain induced in the body by the muscle movements and internal organ functions.[29,42,47,48] Sensors attached to the skin areas near or on moving joints can reveal valuable information about large

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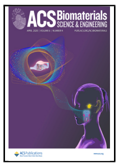

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body motions to measure large strains, and hence, sensors must have high stretchability and good mechanical strength.[46,49−52] On the other hand, some sensors can detect small or lesser intensity strains, which are mainly induced by the muscular movements because of the functioning of the internal organs.[47,53] Such sensors require a high sensitivity toward smaller strains. In the majority of the cases, the idea about proper functioning of internal organs can be assessed by measuring respiration rate, pulse rate, and heart beat by interfacing the strain sensor with the neck, wrist, and chest.[54−58] However, fabrication of sensors that can record high-quality signals when kept in contact with the skin is a challenging task, but a handful of sensors are available in the literature.[4,59,60] In subsequent sections of this review, different prospects and latest developments of skin patchable or wearable sensors will be discussed with suitable examples.

2. SOME ESSENTIAL PROPERTIES OF AN EFFECTIVE SKIN PATCHABLE ELECTRODE

Effective skin-patchable sensor and its components should possess some of the essential properties for its proper functioning (Figure 1). These include linearity, sensitivity, mechanical strength and flexibility, self-healing, self-powering ability, transparency, and biocompatibility. High mechanical strength, flexibility, and biocompatibility are quite essential for an effective integration of a sensor to the skin, and these features are described.

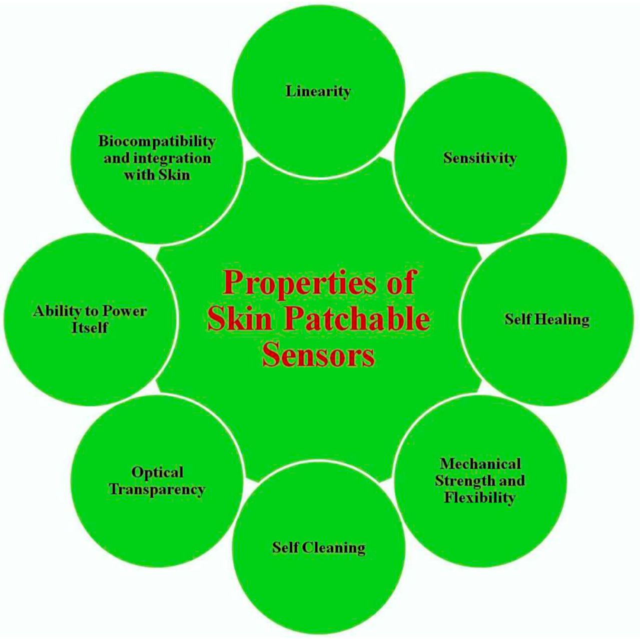

Figure 1. Desirable features of skin-patchable sensors.

2.1. Linearity in Measurement. Linearity in measurement is an important factor regarding the patchable skin sensors because they experience very large strains. Deviation in linearity leads to complexities in the calibration process, and it is a prominent limitation in most of the resistive type sensors. Nonlinearity also arises when the sensors undergo stretching, which is mainly due to the transition of microstructure from uniform to nonuniform morphology.[61]

2.2. Sensitivity. Sensitivity is defined as the slope of relative changes in electrical signal (resistance and capacitance) vs applied strain or stress. Stretchable conductors with a high peizo-resistivity are more eligible for skin-patchable sensor fabrication. Sensitivity in such sensors relies upon the mechanism, which is based on the propagation of cracks, tunnelling, and disconnection between the constituents as well as micro and nanostructures.[62] In this respect, fractured or crackled microstructure designs mediate the conductive interconnections to have high tunnelling peizo-resistance and sensitivity for high pressure.[63] A variety of mechanisms and designs, which when put together may lead to increment in sensitivity.

2.3. Mechanical Strength and Flexibility. One of the essential factors to be considered while fabricating the skin-patchable electrode is to get an intimate contact between the skin and the sensor with a minimal invasiveness and contact resistance.[64,62] This requires greater emphasis on the design of constituent materials with high mechanical strength and flexibility. The deformation of a typical human skin is up to 15% of strain with a elastic modules of 10 kPa to few hundred kPa.[65] Thus, patchable skin sensors should have sufficient stretchability to keep them attached to the skin and to efficiently adapt to the mechanical bending and stretching during the body motion. While fabricating the sensor, it is therefore necessary to modify the flexural strength of its constituent materials, as flexibility is proportional to the third power of thickness of the material.[66]

Fabrication and integration of ultrathin devices has been made possible by the recent advances in thin film techniques and nanotechnology. Single-crystalline Si nanomembranes (100−200 nm thickness) have transferred from silicon to insulator wafers to thin polymer substrate that could enable such an integration to promote the bending to small radii of curvature without any fracture. This also causes decrement in bending stiffness by several orders of magnitude.[67,68] Innumerable reports are available regarding the construction of devices having organic or inorganic constituents on very thin substrates, which could lead to very small bending radii of the order of micrometers even after using materials of relatively large elastic moduli.[69,70] The use of materials having high fracture resistance like CNTs, graphene,[71] some metal oxides, hydrogels, and polymers can be a more effective approach to obtain mechanically robust devices. Apart from incorporating active materials and reducing the thickness, structural, and morphological design of the device also plays an important role in its mechanical stability.[72] In this respect, soft lithography technique is very promising, as it offers soft molds for imprinting targeted materials, thereby allowing the generation of complex 3D morphologies. It also enables the utilization of elastomeric materials as stamps for the incorporation of materials in nanosize regime onto the planar and nonplanar topographic surfaces at reduced cost.[73]

Another preferred procedure is to build an island-bridge type of layout where conductive bridges or interconnects are linked to the active components, called islands.[74−76] These interconnects tend to accommodate an overall stretching in the device and decrease the strain in individual functional components. Therefore, It is necessary that these interconnects must withstand the repetitive strains as a result of daily motion of the human body. Hence, these must be designed and fabricated in such a way that they can undergo only elastic deformation during day-to-day use, as the plastic deformation will lead to crack formation and increased electrical impedance.[77] Matsuhisa et al.[78] reported a printable elastic conductor containing

AgNPs, which are formed in situ by mixing of nanosized Ag flakes, fluorine rubbers, and a surfactant. AgNP formation was influenced by the surfactant, heating process, and molecular weight of the elastomer. The printable elastic composite had conductivity higher than 4000 S cm^{-1} at 0% strain and 935 S cm^{-1} when stretched up to 400%.

There is yet another technique, called additive printing (3D and inkjet printing) for preparing skin patchable devices with better scalability.[43,79−84] This opens up a wider choice of materials such as biomaterials, metal nanoparticles, semiconductors, polymers, and ceramics.[79,85−87] Also, hybrid combinations of such materials can lead to the formation of functional devices that can generate optical or electrical signals after interacting with the target skin region.[70,71,88,89]

2.4. Ability to Self-Heal. Self-healing is very important, as the device components are prone to wear/tear and even damage during daily use.[90] Self-healing allows different components to repair themselves and re-establish their original role in the device functioning.[91,92] The self-healing materials possess a high tolerance to damage or small cracks and prevent their propagation, leading to an increase in device robustness. There are many materials such as self-healing conductors used as constituents in the stretchable and flexible electronic devices like electronic skins.[87,93−95] However, many self-healing polymers from which devices are fabricated have low mechanical strength and are viscoelastic. To overcome this limitation, Kang et al.[90] reported a cross-linked polymer via rationally designed multistrength hydrogen-bonding interactions. This has led to the formation of a supramolecular network in polymer film having exceptional mechanical stretchability and self-healing even under artificial sweat conditions.

Another challenge is the integration of different self-healing components into multifunctional electronic systems. To resolve this issue, Son et al.[96] observed the reconstruction of conducting nanostructures when they are in contact with a self-healing dynamically cross-linked polymer network. The self-bonding feature of the polymer enabled the integration of different devices to a heterogeneous multicomponent device or a single multifunctional system. In another study, Liu et al.[91] reported wearable hydrogels having self-healing and self-adhesive properties, which have the ability to transform mechanical stimuli of deformation of epidermal skin tissues to the readable electrical signals.

2.5. Ability to Self-Clean. The property of self-cleaning assures proper functioning and stability of skin patchable electrode sensors. Recently, Kar et al.[97] prepared a self-cleaning electronic skin capable of mimicking the pressure-sensing feature of natural human skin. It was observed[98] that carbon-based nanoparticles impart a sensor surface with a superhydrophobicity with contact angle 150° and sliding angle 10°. The superhydrophobic nature of the surface let the water droplets roll out along with dust particles and contaminants.[99]

2.6. Optical Transparency. For convenience and comfort, it is necessary that skin-patchable sensors should be transparent such that they are not visible when used on the face and neck.[47,100] Lan et al.[100] prepared optically transparent thermotherapy pads consisting of Ag nanowires on the poly(vinyl alcohol) (PVA) matrix. This film has an optical transparency of 93.1% with excellent flexibility and controllable heating with a rapid thermal response. Recently, Chun et al.[101] prepared thin and lightweight transparent pressure sensor using graphene applicable to an electronic skin sensor. In this protocol, a single graphene layer was grown by CVD onto polymethyl methacrtylate (PMMA) interlayer-coated polydimethylsiloxane (PDMS) substrate. Here, graphene acted as an intact conductive sensing layer.

2.7. Ability to Power Itself. A number of techniques have been developed to accommodate the energy generating and energy storage devices into wearable skin patchable electrode-based sensors.[102,103] Since energy autonomy is necessary for skin patchable devices, they can be designed to harvest their power from the human body itself or from the surrounding environment.[104−106] From the human body, power can be harvested by the mechanical motion of the body,[107,108] which can be converted up to electrical energy. Power can also be harvested from human sweat as in case of wearable biofuel cells,[109] solar energy[110] and electromagnetic energy in the radio frequency (RF) range.[104,105]

TENG is the latest power-generation technology reported for the first time in 2012.[111] This works on the principle of triboelectrification according to which static opposite charges are created between two different materials that are arranged face-to-face.[112] These materials have electrodes at their back side and the charges flow between these electrodes via an external circuit under the potential bias. TENG has been widely used to power a variety of wearable devices such as skin-patchable electrodes. Hwang et al.[47] reported the fabrication of a transparent self-powered patchable sensor in which a triboelectric nanogenerator (TENG) was integrated with a supercapacitor and was used for detecting strain on human skin. In an another report, Pu et al.[113] described the fabrication of an ultrastretchable and transparent TENG that is soft skinlike, which enables energy harvesting and tactile sensing that was achieved by a combination of an ionic hydrogel acting as an electrode and an elastomer, which is the electrification layer.

2.8. Biocompatibility and Interfacing with Skin. Biocompatibility is an important factor for a proper integration of the sensor with the skin such that it may not cause any allergies or rashes on human skin like rashes and etching. There are the three strategies of integration of sensors with skin that are based on different methodologies of attaching the sensor to skin such as epidermal or tattoo-like integration,[106] hard−soft integration,[114] and as functional substrates.[115] The materials that are attached to the skin as temporary epidermal tattoo have the elastic modulus similar to that of the skin and this allows for contact and adhesion between the skin and the sensor.[116] Silicone materials like PDMS, Ecoflex, and Solaris have also been used as substrates in most of the epidermal tattoo sensors.

Apart from silicone materials polymers such as poly(vinyl alcohol) (PVA), polyethylene terapathalate (PET), polyester, and polyimide have also been used as substrates that can be integrated with the human body at different locations. On the other hand, the hard−soft integration consists of a combination of commercial off-the-shelf chips and flexible metallic interconnections on soft and stretchable substrates that can be mounted on the skin.[114] This strategy allows building of skin-mountable integrated circuits. The third strategy of functional substrates involves the combination of different functional substrates and thin films for the fabrication of sensors for a particular application.[117]

Hence, it is necessary to consider all these factors before even choosing an active material for skin patchable electrodes. The other most important factor is the interface between skin and the sensor. The key point at the interface is the better adhesion of the sensor with the skin so that it can actively analyze strain, sweat, blood pressure, etc. Also, biocompatibility is an important factor.

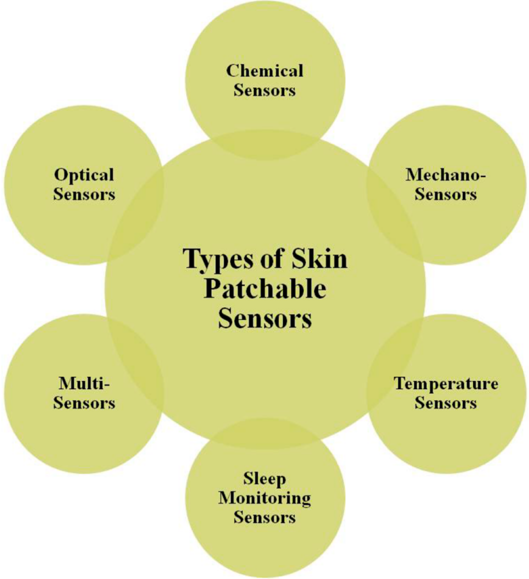

Figure 2. Different types of skin-patchable sensors.

3. TYPES OF SKIN-PATCHABLE SENSORS AND THEIR APPLICATIONS

Six types of skin-patchable sensors (Figure 2, Table 1) are considered in this review, which have been primarily classified based on their applications into categories such as chemical sensor, sleep-monitoring sensor, and temperature sensor. These are also differentiated by their working principles as optical sensors and mechanosensors under which strain and pressure sensors fall. The sixth type is multisensing devices, which combine the two sensing devices in a single substrate.

3.1. Skin-Patchable Chemical Sensors.

Flexible and wearable chemical sensors that can quickly detect different biomarkers in the human body are necessary for day-to-day monitoring of human health. These can be the effective noninvasive techniques to monitor at the molecular level providing information on some vital signs of the disease onset. Such sensors have been used in a number of attempts for the diagnosis of body fluids such as saliva, sweat, blood, exhaled air, breathing air, etc. An ultrasensitive chemical sensor based on 3D biomimetic butterfly wing template was developed by Wang et al.[118] A graphene sheet coated porous 3-D structure has shown to highly selective detection for diabetes-based volatile organic compounds (VOCs) with a fast response time of <1 s at the low detection limit of 20 ppb.

Wearable sweat sensing has gained much attention because of its immense potential in health diagnosis.[119] A novel wearable potentiometric tattoo biosensor for real-time monitoring of G type nerve agent stimulant was fabricated by Mishra et al.[89] This sensor was fabricated by screen printing electrodes on a tattoo paper (Figure 3A–D) and interfaced to a conformal electronic interface to enable wireless data transmission. The sensor could withstand large mechanical stresses without any decrement in performance. It has a fast response time and is selective toward fluorine-containing organophosphate nerve stimulant agent, namely, diisopropyl fluorophosphate (DFP), in both vapor and liquid phases. A microfluidic and flexible sweat-sensing patch containing spiral patterned microfluidic component incorporated with ion-selective sensors and electrical impedance-based sweat rate sensor mounted onto a flexible plastic substrate was fabricated.[120] The patch could perform sweat analysis by interfacing with the sensing component, which is an on-site signal conditioning, analysis, and transmission circuit (Figure 3E). Here, the pressure induced by secreted sweat governs the sweat flow in the microfluidic device to enhance the sweat sampling as well as electrochemical detection of ions viz., H^+, Na^+, K^+, and Cl^- by a sweat collection chamber. The sweat sensor consisted of the electrodes selective for each particular

Table 1. Representation of Different Skin-Patchable Sensors and Their Applications

sensor material	sensor types	applications	parameter sensed/analyte detected	ref
chtiosan/rGO composite	chemical	sensing of diabetes related VOCs	acetone	118
potentiometric tattoo sensor	chemical	G-type nerve-simulating agent detection	DFP	89
microfluidic sweat sensing patch	chemical	sweat analysis	H^+, Na^+, K^+ and Cl^-	120
PDA/PVA hydrogel	pressure/strain	epidermal strain	facial expressions, pulse beat, and limb movements	91
Au micromesh/PDMS	pressure/strain	epidermal strain	eye blinking, chewing, and gestures	76
CNTs/PDMS array sensor	pressure/strain	epidermal strain	epidermal/muscle movement of throat and wrist	123
resistor-type composite pNI-PAM/PEDOT/CNTs	temperature	skin temperature	fever diagnosis	136
stretchable SWCNT-based TFT	temperature	skin temperature	fever diagnosis	137
3D printed "earnable" smart device with liquid metal interconnect	temperature	core body temperature	fever diagnosis	139
TENG-based aluminum leaf patterned film (APLF)	sleep-monitoring sensor	sleep monitoring	sleep monitoring	102
AgNPs/CNT/PEDOT:PSS	multisensor	strain and temperature	ECG, temperature, acceleration	148
graphene-based ISFET	multisensor	sweat and temperature	sweat pH and skin temperature	1
regioregular narrow band gap PIPCP polymer	optical	photoplethysmogram	blood volume changes	151
NIR-PPG (h-PPG) sensor	optical	photoplethysmogram	heart rate variability and pulse pressure	152
SERS-based biocompatible poly-(e-caprolac-tone) film	optical	in situ identification of different analytes	MG molecule	153
nanocavity array incorporated into 3D nanocup plasmonic substrate	optical	biosensing	carcinogenic antigen	154

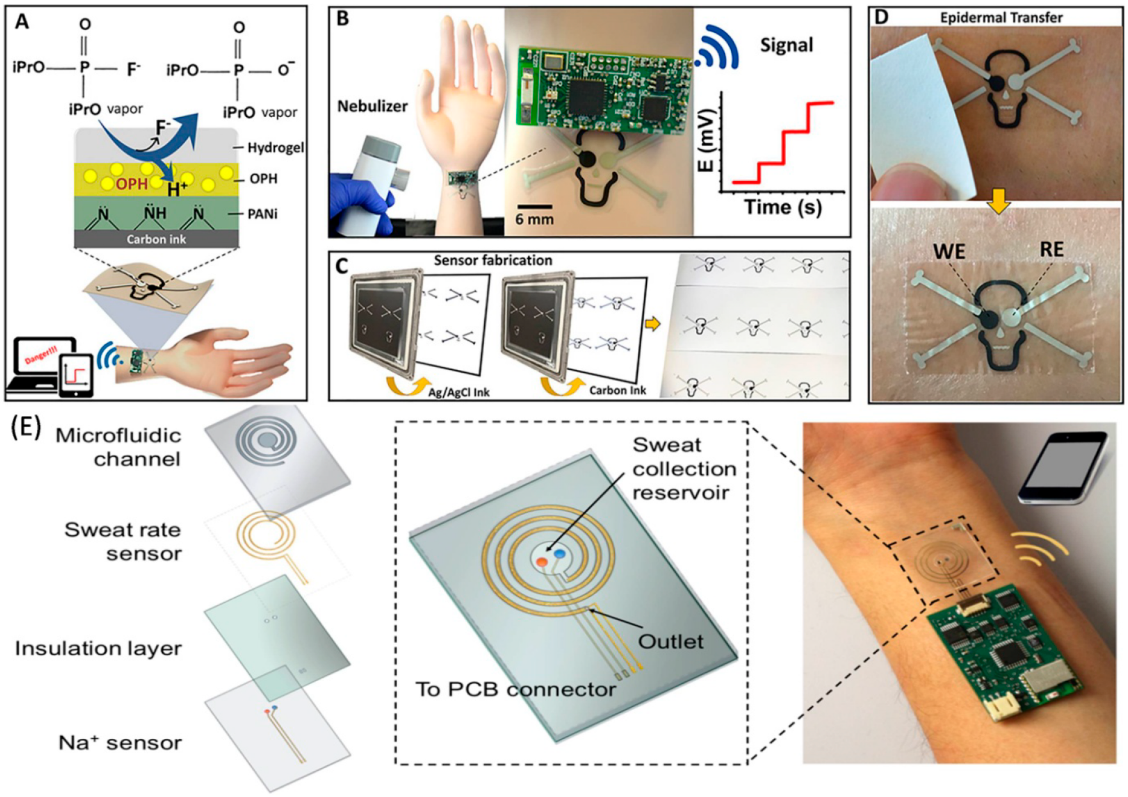

Figure 3. (A–D) Skin-patchable potentiometric tatoo biosensor. (E) Microfluidic channels based sweat sensor. (A–D) Reproduced with permission from ref 89. Copyright 2018 Elsevier. (E) Reproduced with permission from ref.120. Copyright 2018 American Chemical Society.

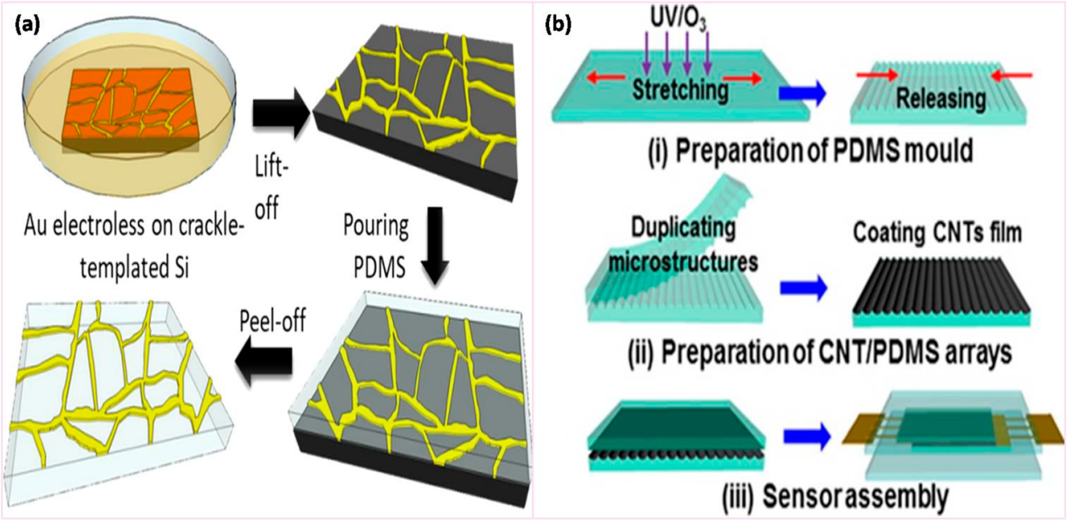

Figure 4. (a) Fabrication process of Au nanomesh/PDMS strain sensor by crackled approach, (b) fabrication of CNT/PDMS pressure sensor. (a) Reproduced with permission from ref 76. Copyright 2018 American Chemical Society (b) Reproduced with permission from ref 123. Copyright 2018 IOP Science.

ion integrated upon the flexible substrate. The experiments carried out on Na^+ selective electrode showed the sensitivity of 56 mV/decade at a constant flow rate of 1 μL/min and sensor shows very rapid response to sudden changes in flow rate.

Gao et al.[121] made a flexible microfluidic pressure sensor consisting of PDMS that was capable of undergoing strains up to 200% without getting failed. The as-fabricated sensor consisted of the Wheatstone bridge type circuit, which was sensitive for both tangential and radial strains with a high sensitivity of 0.0835 kPa^{-1} with the change in output voltage that can operate in the temperature range of 20–50 °C. It has also been found that the liquid is more deformable than the solids so the sensors containing the liquids confined in soft templates as sensing components represent ideal platform for applications such as flexible sensors.[122]

3.2. Patchable Pressure/Strain Sensors. In order to keep a watch on real-time live movements of the human body parts, wearable sensors have been developed to study different body movements such as tiny epidermal movements related to pulse beats, throat vibration, and facial expression changes as well as larger body movements like fingers and legs. In a recent study,[91] a self-adhesive and self-healing epidermal sensor was prepared by the addition of polydopamine (PDA) and poly(vinyl alcohol) (PVA) hydrogel. Because of their self-adhesive and compliant nature, they can be easily affixed onto the skin epidermis without using any external adhesive. Being very sensitive, it can detect small epidermal movements such as pulse rate, throat vibration, and changes in facial expressions. Because of its high stretchability, it can even monitor larger body movements of legs and fingers. A skin patchable strain sensor from Au micromesh, which is partially incorporated in a flexible polydimethylsiloxane (PDMS) support by the crackle templating method (Figure 4a) was developed.[76] The PDMS support provided robustness to the Au microwire network and the sensor had a high optical transmittance of about 85% with an effective stretching strain in the range of 0.02−4.5% in both tension and compression cycles for a gauge factor of 10.[8] This sensor was very sensitive to both high and low strains with an ultrafast response.

Apart from body movements, the pressure sensing is also an important factor to monitor blood pressure, heart beat, and blood flow rate. There is a great need for wearable pressure sensors with a broad pressure-sensing range, high sensitivity, temperature-independent sensing, and rapid response with relaxation times. Yu et al.[123] fabricated a high-performance pressure sensor based on microstructured carbon nanotube/polydimethylsiloxane (PDMS) arrays (Figure 4b) by an ultraviolet/ozone (UV/O_3) microengineered method, which is cost-effective, efficient, and can be used at room temperature. This pressure sensor has a broad sensing range of 7 Pa to 50 kPa with a sensitivity of around −0.101 ± 0.005 kPa, fast relaxation speed of 10 ms, and a good cycling stability.

3.2.1. Working Mechanisms of Pressure/Strain-Based Sensors. 3.2.1.1. Dimensional Effects in Resistive and Capacitive Sensors. In order to detect epidermal vibrations and the movement of the human body parts, sensors work on two distinct mechanisms, which solely depend upon the material characteristics, morphology, and fabrication procedure. These can be either resistive or capacitive type. In case of resistive sensors, resistance to mechanical strain is due to geometrical effects and peizo-resistivity.[39] These are quite different from the traditional strain-based sensors, which work upon the disconnection between the sensing constituents, propagation of cracks, and tunnelling effects. After countering strain, the sensor tends to contract in a transverse direction. If the sensor is resistive type, then the resistance is given by $=\rho\left(\frac{L}{A}\right)$, where ρ is resistivity, L is length, and A is area of the cross-section.[124] There is an increment in resistance upon increase of length and decrease in the area of cross-section.

On the other hand, the capacitive sensor works by change in capacitance, which relies on changes in thickness of the dielectric material and the capacitive area. The change in capacitance is expressed as $C_0 = (\varepsilon_0\varepsilon_r)\left(\frac{l}{wd}\right)$, where ε_0 and ε_r are permittivity in vacuum and dielectric medium. When the sensor undergoes a strain S, then its length and capacitance can be increased by $(1 + S)l$ and $(1{+}S)C_0$. Thus, the capacitance of a capacitor sensor increased linearly by $1 + S$ times the initial value and this linear relationship is valid only up to certain strain values, but not at larger strains.[125,126]

3.2.1.2. Piezo-resistive Mechanism. Piezo-resistivity is defined as the change in resistivity upon mechanical deformation of the material. For a piezo-resistive sensor, the change in resistance can be mathematically defined as $\Delta R = (1 + 2\nu)S + \left(\frac{\Delta\rho}{\rho}\right)$, where ν is poison's ratio of the sensor material. In this expression, the first term $(1 + 2\nu)S$ denotes the impacts of structural deformations and $\frac{\Delta\rho}{\rho}$ is change in resistivity upon deformation. The peizo-resistivity of semiconductors depend upon the change in band gap and interatomic distances.[124,127] This is considered as the most common sensing mechanism due to its simplicity in design and readout by pressure variation into resistance changes.[128]

Numerous efforts have been made to fabricate state of the art peizo-resistive skin-patchable sensors. It is, however, difficult to accurately monitor the pressure under mechanical deformation as a result of variable sensing performance.[129] This issue was overcome by developing bending insensitive ultraflexible and resistive type pressure sensors with the composite nanofibers.[130] This has resulted in no significant change in sensor properties even at low bending radius because of the thin support. This also allowed for accurate and precise measuring of pressure distribution on the sensor surface.

3.2.1.3. Mechanism of Disconnection and Crack Propagation. The stretching of skin-patchable sensors causes loosening of electrical connection between the conductive nanomaterials causing an increase in electrical resistance. This normally occurs as a result of weakening of interfacial binding and mismatch in stiffness among the polymers and nanomaterials.[131] On the other hand, cracks form and propagate in thin film polymer substrate containing brittle materials upon stretching. These cracks are usually formed in regions where there is more stress. Enlargement of cracks and separation between the cracks limit the electrical conductivity of thin films.[132] It was found that the sensor material underwent increment in crack size and upon stretching; it was restored to its initial state upon release because of reconnection between the crack edges. Also, this will lead to drastic increase in electrical resistance, which was used in the development of highly sensitive sensors.

3.3. Wearable Temperature Sensor. Body temperature is an important symptom of insomnia, fever, depression, and malfunctioning of metabolic processes[128] and is useful to gathering of information, which can be useful in medical diagnosis. Although conventional means of measuring body temperature is by the use of a mercury-containing thermometer, skin-patchable sensors can also be fabricated for this purpose of measuring the human body temperature.[133] Rapid response, better reliability, higher sensitivity, wider temperature measurement range and low weight are the most desirable characteristics of a flexible temperature sensor. The mechanism of working of the skin-patchable temperature sensor is based on the changes in resistance, which can be achieved by spreading the conductive fillers on the insulator polymer matrix or by heterogeneously spreading the temperature-sensitive conductors onto the flexible substrate.

Single-walled CNTs containing carbonyl groups and hydrogen-bond-based polymers were used to prepare a soft thermal sensor having an excellent mechanical adaptability because of noncovalent hydrogen bonds in the polymers.[134] It was observed that materials with positive temperature coefficient can be good candidates for the fabrication of flexible skin-patchable temperature sensors with increased sensitivity and better response time.[135] Recently, Oh et al.[136] fabricated a highly

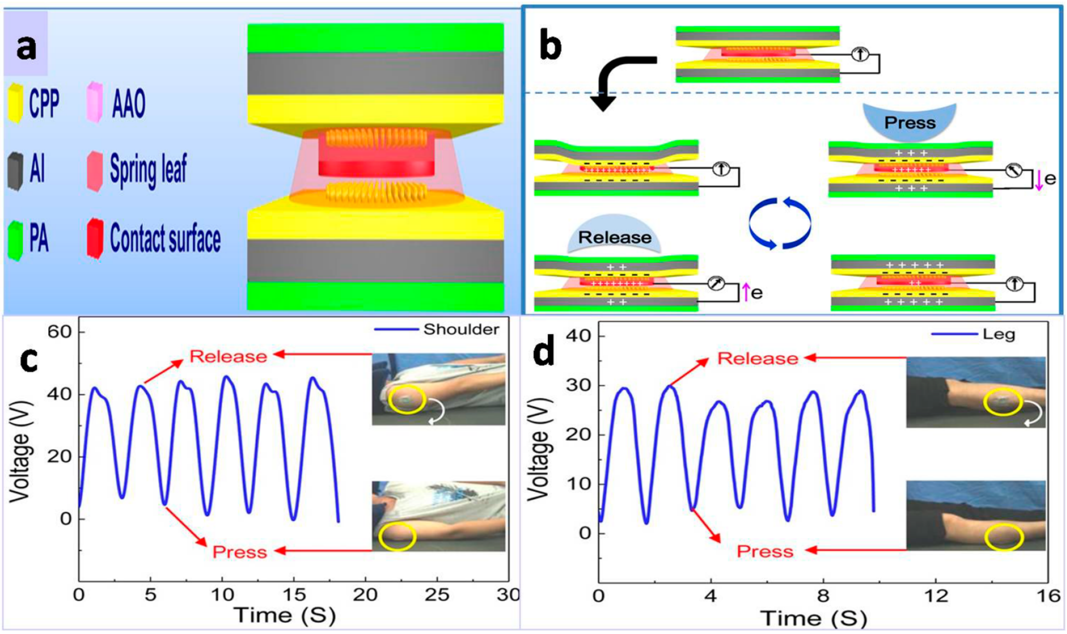

Figure 5. (a) Sandwiched structure of sleep monitoring sensor and (b−d) its working mechanism. (a−d) Reproduced with the permission from ref 99. Copyright 2016 American Chemical Society.

sensitive, flexible, wearable resistor-type temperature sensor using an octopus biomimicked adhesive. The sensor was fabricated using the composite of poly(*N*-isopropylacrylamide) (pNI-PAM) temperature-sensitive hydrogel, poly(3,4-ethylenedioxythiopene) polystyrenesulfonate, and CNT; the device showed a high temperature sensitivity of about 2.6% between 25 and 40 °C in order to accurately detect small changes (0.5 °C) in the body temperature. The sensor was fabricated by coating octopus mimicked rim structure of adhesive polydimethylsiloxane (PDMS) layer with pNI-PAM by a single mold formation via an undercut process of photolithography. This sensor performance remained unaffected even after repeated attachment/deattachment cycles on the skin epidermis without producing any long-term irritation.

In a recent report, Zhu et al.[137] reported some circuit design strategies based on stretchable CNT-based transistors that have led to increased sensor accuracy and robustness. Another temperature monitoring sensor was fabricated by Trung et al.[138] using freestanding single reduction graphene oxide (rGO). This fiber-based sensor was incorporated with textiles that could be worn as shocks or undershirts. The sensor showed a fast response time of 7 s with a good recovery time of 20 s. Its performance did not change even under applied mechanical deformation. The conventional skin-patchable temperature sensors could measure the skin temperature, which varies significantly from the core body temperature. In order to overcome this issue, Ota et al.[139] demonstrated a 3D printed wearable "earable" smart temperature sensor designed for wearing in the ear for measuring the core body temperature via tympanic membrane or ear drum-based infrared sensor. This sensor can be successfully interfaced with a wireless module for proper monitoring. The 3D printing fabrication method allowed easy customization of the device for personalized healthcare.

3.4. Skin-Patchable Sleep-Monitoring Sensor.

Irregular sleep is another major health disorder that can be diagnosed by the effective use of sensors that can measure the airflow breathing, movements of thorax and other body movements.[140] However, the conventional monitoring process requires bulky equipment that consume much energy for obtaining precise and sensitive measurements. It is also not very easy to sense body movements in a state of sleep apenea. A compact, flexible and smart sensor could accurately monitor the sleep disorders. Recently, Song et al.[102] developed a flexible and low-cost TENG device based on the patterned aluminum-plastic laminated film (APLF) and an entrapped cantilever spring leaf forming the sandwiched structure (Figure 5a). This acted as an effective sensitive sensor for sleep monitoring of the body, which rapidly responds to external pressure.

The sensing phenomenon (Figure 5b−d) involves pressure from an external environment and release by self-recovering due to rebound, leading to tribo-electric effect and charge separation between APLF and entrapped spring leaf. The open circuit voltage arising from APLF with the nanopillars of dia 600 nm and length of 1.5 μm is more than two-times 55 V. On the other hand, the patterned nanostructure plays an important role in enhancing the output voltage and current, resulting in a significant improvement of the device sensitivity.

3.5. Wearable Multisensing Electrodes.

Multisensing electrodes often produce a variety of human health monitoring applications. Collection of data from daily human activity and some critical parameters such as heart rate, body temperature, pulse rate and blood pressure is of much significance as these are highly affected by the day-to-day activities. Hence, simultaneous monitoring of these important parameters is highly desirable.[141] Incorporation of two or more sensors on a single substrate has gained immense attention, as it enables simultaneous detection and diagnosis of various diseases.[142] The most common strategy to integrate two or more different sensors is stacking of two active layers for creating a bimodal sensor.[143,144] In particular, integration of pressure and temperature sensors has opened doors for the fabrication of devices that can measure two different signals without any interfacing from the external devices.[145]

In the eariler literature, few attempts regarding multisensing devices have been reported.[146,147] In this respect, Yamamoto et al.[148] fabricated a planar sensor sheet that was incorporated with

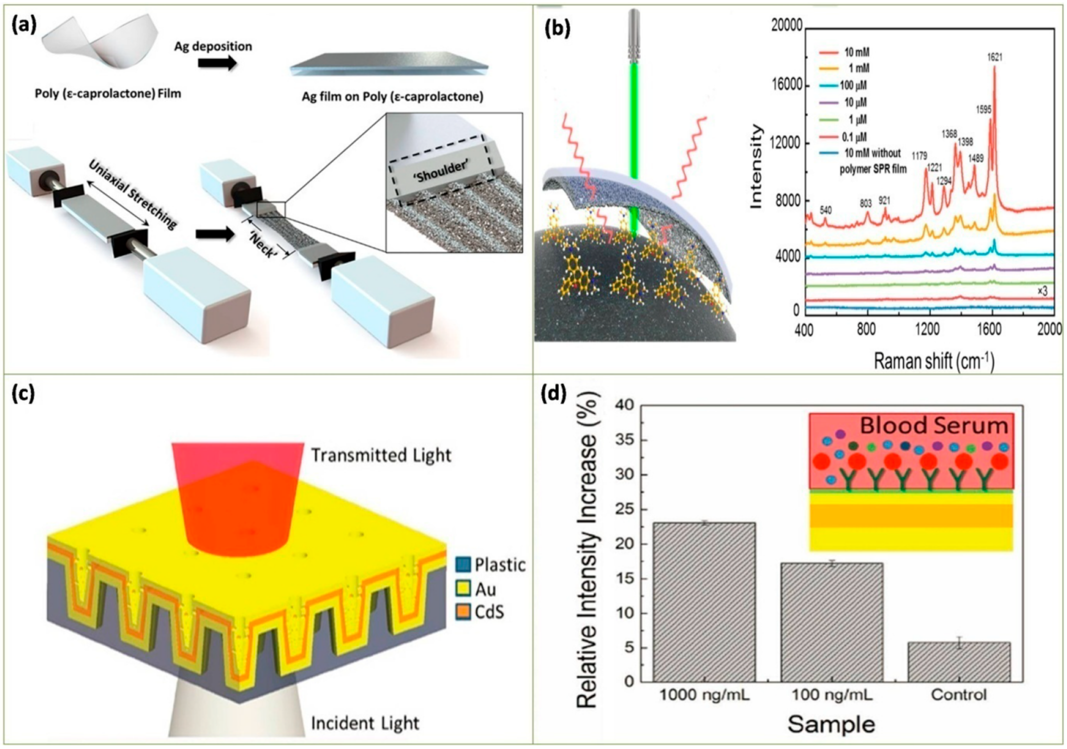

Figure 6. (a) Schematic representation of stretching of SPR film under external force. (b) Schematic diagram showing contacting polymer SPR film onto the green mussel and gathering of SERS signals from the back surface. (c) Schematic representation of the ML-nanoLCA depicting the multilayer structure and direction of illumination. (d) Schematic representation of surface fictionalization for CEA detection. (a, b) Reproduced with permission from ref 152. Copyright 2017 American Chemical Society. (c, d) Reproduced with the permission from ref 153. Copyright 2017 Wiley Online Library.

the sensors to detect human body movements and temperature. This sensor has a unique kirigami-type electrode architecture, which enables it to conveniently record the acceleration without any effect on the resistance. The sensor was also found to be mechanically reliable and can be placed in direct contact with the skin, and as a result, real-time measurements related to motion, temperature and even ECG signals for successful recording was possible. Nakata et al.[1] developed a wearable sweat chemical sensor sheet for pH measurement containing an ion-sensitive field-effect transistor (ISFET) and temperature sensor incorporated into it. The sensor enabled simultaneous measurement of sweat pH and skin temperature when the device was attached to the human neck during an exercise routine. This has led to the precise measurement of both these parameters, which was confirmed from the commercially available sensor devices.

3.6. Skin-Patchable Optical Sensors. Optical sensors are capable of detecting a wide variety of optical signals such as wavelength, intensity, frequency and polarization. Their working performance was evaluated on the basis of their selectivity, sensitivity, and response time.[149] In most optical sensors, photodetector is an important component in addition to pulse oximeters containing two light-emitting diodes (LEDs) having different emission wavelengths, which are placed on the human body and the light reflected or transmitted from the internal tissue was detected by the photo detector.[149,150]

The commercial oximeters are bulky, which hampers their practical applications. To overcome this issue, a sensor with ultrathin, flexible, and reflective pulse oximeters were fabricated[151] comprising polymeric LEDs and a near IR photodetector composed of regiro-regular narrow band gap poly(decanodithiopene-pyridyl [2,1,3] thiadiazole-cyclopentadithiopene) (PIPCP) polymer. These devices have shown fast and more precise on—off switching behavior with a high device yield, which is enabled by the deliberate optimization of physical dimensions of the active layer. The sensor showed better sensitivity in the near-IR region because of the balance between good responsively and mechanical conformability,

In a recent study, Xu et al.[152] fabricated a wearable photoplethysmogram sensor that has provided measurements to evaluate day-to-day health monitoring. The flexible near-infrared photoplethysmogram (NIR PPG) sensor was integrated to a low power and highly sensitive organic phototransistor (OPT) with an efficient inorganic LED. It was demonstrated that skin patchable and flexible PPG sensors were capable of monitoring the variation in heart rate and successful tracking of pulse pressures with a high precision at different postures of the human body and these exhibited a more reliable performance than the commercially available PPG sensors, and they also consumed less power.

Apart from NIR-based patchable sensors, there are sensing devices that work upon Surface Enhanced Raman Scattering (SERS), which provides a more rapid, sensitive and non-destructive strategy for label-free fingerprint diagnosis. Xu et al.[153] demonstrated a biodegradable and flexible SERS film by inversely and longitudinally stretching Ag-deposited biocompatible poly(e-caprolac-tone) film (Figure 6a). The composite film exhibited an exciting phenomenon upon stretching in which surface plasmon resonance of stretched polymer film offered 10-times more signal enhancement compared to unstretched polymer film. The uniform SERS signals also showed a good temperature stability. The flexible and transparent polymer film showing surface plasmon resonance (SPR) effect was effectively used to detect various chemicals. Ameen et al.[154] devised a sensor and sensing method based on plasmonic-photonic interactions that occurred when a nanocavity array was incorporated in a 3D tapered nanocup plasmonic substrate. Thus, prepared sensor allowed very sensitive sensing of changes in refractive index with respect to changes in transmission peak intensity without any shift in the resonance peak wavelength. Unlike the conventional plasmonic sensors, there is a consistent and selective change in the transmission peak intensity at the resonance peak wavelength without any spectral shift. The as-fabricated sensor was used as a biomarker to detect cancer, called carcino-embryonic antigen (CEA), which was found to have a detection limit of around 1.0 ng/mL or 5×10^{-12} M.

4. FUTURE PROSPECTS AND OUTLOOK

Wearable skin-patchable sensors are a step forward toward the development of health monitoring and diagnostic technologies. A variety of health related parameters can be observed via skin-patchable sensors like body temperature, heartbeat, respiration rate, movements of different body parts and sweat composition after interfacing them with the skin. Day-to-day advances in the field of thin film and flexible electronics as well as a number of efforts to integrate two or more sensors in a single substrate for the development of multisensor devices have contributed much toward the development of skin-patchable sensors.

When compared to conventional diagnostic methods, skin-patchable devices are promising for easy and rapid detection of vital disease symptoms to monitor routine health-related parameters like heart beat, blood pressure, pulse rate, and body temperature. However, the materials used for device fabrication and the current fabrication methods seem to increase the overall cost. The overall device cost can be successfully reduced by the use of carbon-based substrates and sensing materials such as graphene, CNTs, and polymers as well as by simplifying the fabrication methods. However, there still seem to be some challenges.

Overall, skin-patchable sensing materials can be the useful tools in the future for quick diagnosis and monitoring of health-related issues like blood pressure and symptoms of different diseases like malaria. The early diagnosis can be of much help for diabetic patients to monitor their sugar levels. This can be also helpful for researchers working medical science area because it can lead to further developments of medicines for treating different diseases.

5. CONCLUSIONS

Flexible and wearable skin-patchable sensors can enable the monitoring of human health and also help for quick diagnosis of some critical symptoms of the diseases. There have been a number of skin-patchable sensors developed that aim at measuring the blood pressure, heart beat, pulse rate, and body temperature. Apart from this, there are sensors that can detect sleep and motion of the human limbs. There are also sweat sensors that can measure the sweat pH and detect the presence of different ions in sweat fluids and sensors to test other body fluids such as saliva, blood, tears, and exhaled air. On the basis of their working pattern, these sensors fall into categories such as skin patchable chemical sensors, which are meant for sweat, blood, and saliva analysis, mechanosensors for measuring the strain, blood pressure, heart beat, and human motion, temperature sensors, and sleep monitoring sensors.

Efforts are underway to integrate one or more sensors into a single chip to create a multisensor. Stacking of two sensor materials has been achieved to create a bimodal sensor capable of sensing two different variables by a single chip. In this regard, the temperature and pressure sensors have been integrated as the bimodal sensor to measure simultaneously blood pressure and body temperature. Optical sensors, which work on the basis of wavelength, intensity, and polarization of light from the tissues, can be very promising to provide symptoms of deadly cancer, presence of harmful chemicals, and malfunctioning of important organs such as the heart. Overall, the development of skin-patchable sensors can be a revolution to healthcare and allied industries.

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Notes

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