Physical and Chemical Sensing with Electronic Skin

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Abstract— **This paper reviews current progress on flexible and stretchable transistors and sensors for the next generation multiplex electronics (commonly referred to as "electronics skin" or "e-skin") that is capable of simultaneous detection of multiple information from a variety of surfaces including human skin. Flexible chemical sensors for sweat analysis as well as physical sensors for detecting tactile force, bending, and temperature will be discussed, with emphasis on materials, detection mechanisms, and device demonstration to realize multiplex human-interactive devices. Next, system integration enabling the real-time monitoring of health conditions is also demonstrated as a proof-of-concept. Finally, perspectives on e-skin for moving towards realizing practical wearable electronics in the market are discussed. This review targets the translation of the nano- and flexible-technologies from academic innovations to industrial practical applications with high impact breakthroughs.**

***Index Terms*—electronic skin, physical sensors, chemical sensors, flexible electronics**

I. Introduction

Dynamic physiological monitoring is essential to the realization of personalized medicine through continuously collecting large sets of data from people's daily activities and capturing meaningful health status changes in time for preventive intervention. There is an urgent need for wearable devices that can perform personalized physical and molecular monitoring to provide in-depth understanding of the physiological status of an individual. In addition, the motion artifacts and mechanical mismatches between conventional rigid electronic materials and soft skin often lead to substantial errors in collected data. Thus, it will be of great importance to develop wearable and flexible electronic systems that could be conformally in contact with the skin and integrate the sensing capabilities for accurate dynamic health assessment.

Recent years, tremendous progress has been made in flexible electronic skin (commonly referred to as "e-skin") which has unique advantages including high transparency, light weight, low cost, high flexibility, and strechability[1-6]. The innovations in materials, fabrication processes, and sensing strategies have played a significant role in the development of the next generation e-skin based devices toward physical and chemical sensing[1-10]. Many applications that utilize the e-skin concept have been proposed such as healthcare[7, 8, 11, 12], robotics[13-15], and "Internet of Things (IoT)" [16, 17]. The e-skin concepts include not only multi-modality physical and chemical sensing, but also interaction with human and other objects based on the real time analysis of sensing data [14]. Another important consideration for wearable or biomedical application is comfort and flexibility due to direct contact to human body. To address many challenges for the next class of electronics, macro-scale and multi-functional flexible and/or stretchable devices need to be developed by applying a variety of materials, structures, fabrication technologies, and concepts.

In this paper, we will review current progress on e-skin starting with flexible and stretchable transistors and sensors for the next generation wearable and flexible electronics that are capable of simultaneous detection of multiple information from a variety of surfaces including human skin. Flexible chemical sensors for sweat analysis as well as physical sensors for detecting tactile force, bending, and temperature will be discussed, with emphasis on materials, detection mechanism, and device demonstration to realize multiplex human-interactive devices. Perspectives on e-skin for moving towards realizing practical wearable electronics in the market will also be discussed.

II. Physical Tactile Pressure Sensing E-skin

A. Flexible E-skin

Flexible e-skins consisted of the arrays of tactile pressure sensors and/or temperature sensors have been widely studied by using organic and inorganic materials [1, 2, 18-23]. Two types of the device backplanes, active matrix and passive matrix, are mainly used depending on the applications and requirements. Active matrix backplane integrated with switching transistors to select a pixel has better resolution compared to the passive one. This is because the cross-talk from the other sensors can be suppressed drastically. For passive matrix, the advantage is that the device structure is simple. To achieve macro-scale and low-cost processes for the active matrix system, organic-based transistors are often used [1, 4, 24]. Organic semiconducting materials can be formed by solution-based processes resulting

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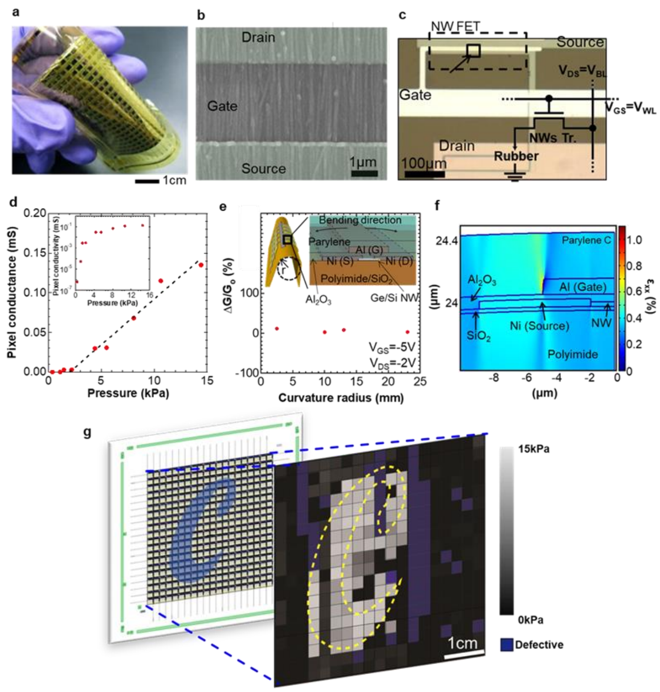

Fig. 1. Nanowire-based active matrix backplane e-skin. (a) Photo of 18×19 pixel array e-skin. (b) SEM image of Ge/Si core/shell NW array transistor. (c) Optical microscope image of a pixel. (d) Pixel conductance as a function of applied tactile pressure. (e) On-state conductance change ratio under mechanical bending of the transistors up to 2.5 mm radius. (f) FEM simulation of strain distribution under 2.5 mm bending radius. (g) Pressure distribution measured from the nanowire-based active matrix backplane e-skin. Reproduced with permission from ref.2. Copyright 2010 Nature Publishing Group.

in good scalability. However, the mobility of organic transistors still needs to be improved to realize low power operation. In contrast, macro-scale printing and transferring processes of inorganic nanomaterials have been proposed to realize high mobility and stable transistors [2, 25, 26]. Due to nanosize of the materials, the inorganic-based transistors can be mechanically flexible for the purpose of e-skin applications [27]. In fact, using nanowires [2], nanotubes [25], and two-dimensional material systems [26] have been used to develop the active matrix backplane e-skin. In this review, NW-based e-skin is discussed in detail.

Flexible e-skin capable of tactile pressure sensing has been demonstrated based on active matrix backplane using Ge/Si core/shell nanowire (NW) arrays on a polyimide film[2]. The nanowire arrays can be used to form p-type semiconductor thin-film transistors that act as switches in an active matrix for pixel selection. Pressure detection is realized using a commercially available pressure sensitive rubber (PSR). A 18 × 19 array of pressure sensors with active matrix transistors was fabricated by optimizing device process and materials (Figure 1a). Ge/Si NW arrays were printed between source (S) and drain (D) electrodes as the transistor channel materials with a density of ~5 NWs/µm as shown in Figure 1b. Due to strong van der Waals interaction between NWs and SiO_x layer formed on the polyimide film, the NW array is stably formed during device fabrication process. Each pixel comprises a transistor and PSR (Figure 1c). Field-effect mobility of nanowire-based flexible transistor is ~20 cm^2/Vs. It should be noted that the on-state conductance of the transistor should be much higher than that of the PSR in order to monitor precise tactile pressure change. To achieve this, transistors with relatively wide channel (~250 µm) was designed, resulting in transistor with on-state conductance of ~0.34 mS that is higher than the one of the PSR.

Tactile pressure detection was achieved by measuring the conductance change of the PSR through the transistor. The PSR consists of conductive nanocarbon material in an elastomer matrix. The matrix conductance determined by the tunneling current between the nanocarbon filler varies as a function of tactile pressure applied on the PSR. The pixel conductance increased drastically after the pressure exceeded 3 kPa, suggesting that the distance between nanocarbon in the rubber was small enough to cause tunnel current. At >3 kPa, the conductance increased linearly with increasing pressure as shown in Figure 1d. The sensor sensitivity extracted from the linear fitting above 3 kPa is ~11.5 µS/kPa. Because this e-skin can detect relatively small tactile pressure of a few kPa that is similar to the pressure exerted by fingers when typing a keyboard, it can potentially be used to imitate human skin for robotics applications. The response time of the e-skin is ~0.1 s, which is almost the same as the speed of the pressure input from the experimental setup used in this study. However, this time could be much faster because the response time is limited by the relaxation time of the elastomer in the PSR.

For many e-skin applications, mechanical flexibility and robustness of the devices are also important factors besides the sensing sensitivity. Conductance change of the NW flexible transistor was characterized under mechanical bending as a function of curvature radius down to 2.5 mm (Figure 1e). The bending direction was along the channel length direction between the S/D electrodes. The maximum conductance change ($\Delta G/G_0$, where $\Delta G=G_0-G$, G and G_0 are the conductance at bending and flat states, respectively) was ~6 % at a radius of 2.5 mm. This small change was most likely caused by a strain in the NW channel of ~0.35 % extracted from a finite-element method (FEM) simulation (Figure 1f). Because the applied pressure induced much larger conductance change in the PSR, this strain effect on the transistor conductance became negligible. The robustness of the device was also confirmed without having delamination or breakage after at least 2,000 bending cycles.

As a proof-of-concept demonstration of using the NW transistor-based active matrix e-skin for spatial mapping of tactile pressure distribution, an 18 × 19 array with a size of 7 × 7 cm^2 on polyimide film was used to measure the pressure from a "C"-shaped object with applied normal pressure of around 15 kPa. As expected, the pressure distribution measured by the active matrix e-skin resembled the object placed on top. Additionally, owing to the use of active matrix backplane design, cross-talk effects from adjacent pixels were minimized, resulting in high spatial resolution for tactile pressure monitoring.

B. *Stretchable E-skin*

Mechanical stretchability is another important function to cover three-dimensional objects and also movable objects like human bodies. Two strategies can be used to realize mechanical stretchability in the e-skin. The first one is to make all materials used in the sensors and transistors intrinsically stretchable. To achieve stable and reliable sensing results, the material formation is challenging but good progress has been made as will be reviewed in latter sections of this paper. The second approach is to use structural design to achieve stretchability for only the substrate. In this case, active materials need to be strain free under mechanical deformation based on the strain engineering. This section focuses on the second approach due to simple and reliable approaches in this present technology.

For this approach, serpentine metal structure is used to be stretchable without causing delamination and cracks in the devices [3, 15]. Other approach is to arrange the film structures such as kirigami and honeycomb shape of unstretched films. In particular, the polyimide substrate is designed to have a honeycomb shape formed by a laser cutter, and the transistors using organic and inorganic materials are formed on the place where is almost strain free under stretching conditions [1, 25]. As an example, carbon nanotube (CNT)-based stretchable e-skin with the honeycomb shape of polyimide film is discussed. CNT-based transistors were formed on the vertices of the honeycomb to reduce the strain under stretching condition as displayed in Figure 2a-b. Using this structure, the polyimide films with transistors can be covered conformably over a baseball without having cracking and delamination (Figure 2a). To understand strain distribution under stretching, two types of honeycomb structures were fabricated and simulated using finite element method (FEM) simulations for stress observations. Figure 2c shows that the film is stretchable which is enabled by the structure change of the honeycomb structure. Smaller width of the honeycomb structure in the polyimide film leads to more flexible and stretchable film. Strain distribution of both samples under 2 mm stretch is shown in Figure 2d. Firstly, the polyimide with honeycombs of smaller width has much less stress compared to the one with larger width. Secondly, the vertices of the honeycomb pattern highlighted with a white circle in Figure 2d exhibits almost zero stress for honeycomb structure with both widths. Utilizing this strain engineering, flexible thin-film transistors can be strategically fabricated at the vertices on the honeycomb polyimide substrate to prevent them from being affected by large tensile stress when the sample is stretched.

To achieve uniform and high-performance flexible transistors, semiconductor-enriched CNT network film was developed. The density of CNT network can be controlled by the deposition time as shown in Figure 2e[25]. This density difference affects the on-current and transconductance (corresponding to the field-effect mobility), as well as I_{on}/I_{off} ratio of the devices. For higher density CNTs, on-current is increased while I_{on}/I_{off} ratio is decreased due to high off-current caused by the bundling of CNTs. The average mobility, Log (I_{on}/I_{off}), and threshold voltage change from 16 cm^2/Vs to 26 cm^2/Vs, 6 to 2.4, and -4.6 V to -5.0 V, respectively, when the CNT deposition time is increased from 5 min to 90 min. By integrating a PSR with a CNT transistor in series, output conductance changes as a function of applied pressure due to the resistance change of the PSR (Figure 2f-g). Briefly, the conductance increases almost linearly with increasing pressure from 0 to 6 kPa and then saturates at above 6kPa, which is limited by the dynamic range of pressure detection of the PSR.

After confirming the fundamental pixel operation, the stretchability for the integrated CNT transistor active matrix

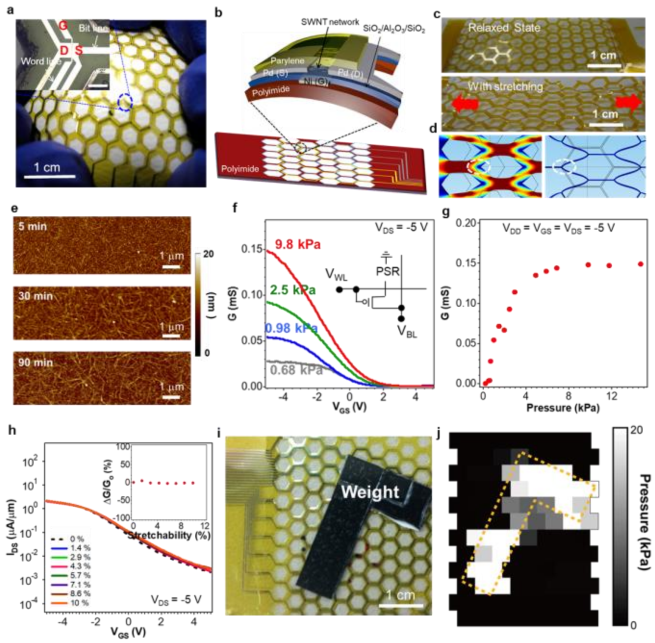

Fig. 2. Stretchable e-skin with CNT-based active matrix circuits. (a) Photo of the stretchable e-skin covered over a baseball. (b) Schematic of the honeycomb polyimide structure and CNT transistor. (c) Photo of the honeycomb structure polyimide at relaxed state (top) and stretching state (bottom). (d) FEM simulation of stress under 2 mm stretch with wider (left) and shallower (right) honeycomb width. White dashed circles indicate almost zero stress region under stretching deformation. Reproduced with permission from ref.25. Copyright 2011 American Chemical Society.

backplane was investigated. Under tensile strain of up to 10 %, the transistor characteristics remained almost unchanged as shown in Figure 2h. As described above, the stable and robust operation of the transistors was realized by placing the transistor at almost stress-free regions under stretch. However, for this honeycomb structure, strain of above 10 % causes damage to the polyimide film. If more stretchability is required, the honeycomb width should be smaller.

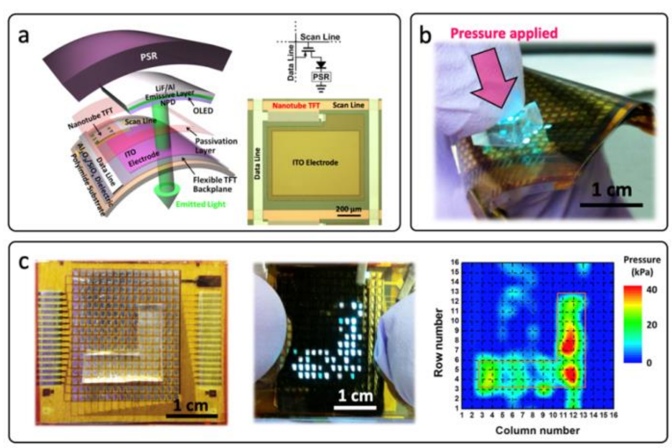

Fig. 3. User interactive e-skin. (a) Schematic and optical micrograph showing one pixel of the user-interactive e-skin, comprising a TFT, a pressure sensor, and an OLED. (b) Photograph illustrating the operation of the user-interactive e-skin - the OLED in the pixels are turned on locally when pressure is applied. (c) Mapping of applied pressure using the interactive e-skin with both optical and electrical output. The data show high spatial resolution and good agreement between the two sets of outputs. Reproduced with permission from ref.14. Copyright 2013 Nature Publishing Group.

Finally, the stretchable e-skin was used to monitor pressure distribution by applying a pressure of up to 20 kPa as shown in Figure 2i. The stretchable e-skin is comprised of 12×8 pixels and all pixels worked without any malfunction. To obtain the pressure mapping, each pixel was addressed by turning the pixel transistor on, and the result is shown in Figure 2j. Due to the large size of pixel limited by the honeycomb structure of the polyimide film, the resolution is not high enough to accurately reflect the shape object. However, pressure distribution was clearly obtained. To improve the resolution, transistor size and honeycomb structure needs to be shrunk down.

C. Human-Interactive Responsive E-skin

In additional to combining a variety of sensing modalities, it is also possible to integrate human-interactive functionalities such as display or haptic feedback to the e-skin to expand its potential wearable applications. For example, light emission [14] or color changing [28] are effective ways to instantaneously display information about the sensor response to the users. Figure 3 shows an example of a user-interactive e-skin, in which the pressure sensors are monolithically integrated with organic light-emitted diodes (OLEDs) in an active matrix backplane individually addressed by carbon nanotube thin-film transistors (TFTs) [14]. As shown in the schematic in Figure 3a, each pixel is comprised of a pressure sensor (PSR) that is connected in series with an OLED. In such a configuration, the externally applied pressure changes the resistance of the PSR and thus the current flowing through the OLED, allowing the applied pressure to be visualized as the emitted light from the OLED. The light intensity qualitatively represents the magnitude of the applied pressure. Figure 3b gives another example where only pixels with pressure applied are turned on. Such human-interactive e-skin enables both electrical readout (by measuring the current in each pixel) and visual feedback of the applied pressure with a high spatial resolution (Figure 3c). While this example uses pressure sensor and OLED as a proof-of-concept demonstration, one can easily replace or add other types of sensors such as temperature sensor, photodetector, or sweat sensor, to enable more complex functionalities that may find a wide range of applications in wearable health-monitoring devices.

III. E-SKIN FABRICATED BY PRINTING PROCESS

Among the various potential applications of e-skin, many of them have less demanding requirements on device performance and speed compared to modern integrated circuits. The main consideration is instead focused on large area manufacturability and cost. For example, considering an e-skin is designed to be used as a wearable patch for biomedical applications, if the cost is sufficiently low, it will be feasible to use it as a disposable device, which is ideal. Conventional processes adapted from the semiconductor industry usually involve photolithographic patterning and vacuum-based deposition and etching processes that are costly. Alternatively, recent work has demonstrated that printing – a widely used additive manufacturing method, could also be adopted for large-scale and low-cost fabrication of electronic devices and sensors [29] [30]. Additionally, printing also enables fabrication on many unconventional substrates including plastics, elastomers, papers and textiles, which could enable numerous applications ranging from smart packaging to electronic wallpaper and ubiquitous wearable electronics [31]. In this section, we will review progress made on printed TFTs used as switching devices in the active-matrix backplane of the e-skin. We will also cover a variety of printed sensors in the next section.

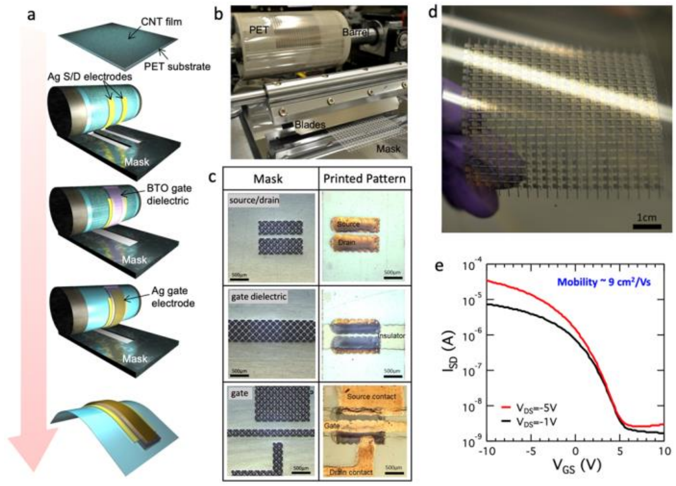

Fig. 4. Gravure printed carbon nanotube TFT on plastic substrate. (a) Schematic diagram illustrating the printing process. (b) Photograph of the inverse gravure printer. (c) Mask design and the printed pattern after each printing step. (d) Photograph of a fully printed 20×20 TFT array. (e) Transfer characteristics of a printed TFT. Reproduced with permission from ref.54. Copyright 2013 American Chemical Society.

A. Printed CNT-based Flexible Transistors

Printing requires the various types of materials used in a transistor – metal, insulation, and most importantly semiconductor, to be in solution form and formulated as electronic inks with appropriate concentration, viscosity, vapor pressure, and wetting on the target substrates. A variety of materials have been studied in the printed electronics community. For metal ink, metallic nanoparticles (e.g. silver), one-dimensional metallic nanowires or carbon nanotubes [32-35], two-dimensional graphene [36], conductive polymer [37, 38], or nanomaterial/polymer composites [39, 40] can be used. Depending on the material, the sheet resistance of the printed film typically ranges from ~10 Ω/sq for metal-based nanomaterials to above 100 Ω/sq for carbon-based materials and conductive polymers. The most widely studied dielectric inks are ion gels [41] and polymer-inorganic nanoparticle hybrid dielectrics [42]. Due to the formation of electrical double layers, ion gel offers very large gate capacitance, which is in principle independent of the film thickness. Hybrid dielectrics consisting of polymers and inorganic nanoparticles also offer dielectric constants that are significantly better than pure polymer and are also excellent choices for printed transistors. Lastly, for the channel semiconductor, organic semiconductors have been widely studied in printed electronics for a long time [43, 44]. More recently, inorganic 1D [45, 46] or 2D nanomaterials [47] , and metal oxides [48] have also been widely explored for printed electronics, many of which have significantly outperformed organic semiconductor in terms of overall device performance and long-term stability. In particular, solution-processed semiconducting carbon nanotubes have shown great promise for high performance printed TFTs and circuits on flexible substrates. Regarding printing methods, there are also several options available, with inkjet printing [49, 50], gravure printing [51-54], aerosol jet printing [41, 55] and screen printing [56] being the most commonly used. Gravure and screen printing are particularly suitable for large area fabrication, while inkjet and aerosol jet printing are maskless and more flexible for changing the layout design, which is important in a research and development setting. On the other hand, for inkjet and aerosol jet printing, the patterns need to be printed line-by-line so the process can be relatively slow. Gravure and screen printing are much faster but they instead require multiple set of masks that may be costly and also make modifying the design more difficult. Lastly, inkjet, aerosol jet, and gravure printing are all additive manufacturing processes while the screen printing is subtractive meaning that a lot of material could potentially be wasted. While all printing methods above have been used to fabricate printed carbon nanotubes TFTs, it is important to consider the pros and cons of each method above and choose the most suitable printing method for a particular application.

Figure 4 gives an example of high-performance flexible carbon nanotube TFTs made by gravure printing [54]. The detailed fabrication process is illustrated in Figure 4a-c, in which the source/drain electrodes, gate dielectric layer, and gate electrode are sequentially printed onto flexible polyethylene terephthalate (PET) substrates in a roll-to-roll or roll-to-plate manner. For such gravure printing processes, the viscosity and surface tension of the inks and surface chemistry of the substrates are critical and need to be optimized to achieve uniform and reproducible device performance. Silver nanoparticle ink are widely used as printable conductor for making the electrodes. The printed gate dielectric layer plays a more crucial role as it directly affects the device performance. Because conventional polymers have low dielectric constant and it is also very difficult to achieve ultrathin pinhole-free printed dielectric layer with good uniformity and clean interface, the transistors inevitably require large voltage to operate. In order to address this issue, hybrid dielectrics consisting of polymers and inorganic nanoparticles such as $BaTiO_3$/PMMA hybrid dielectric can be used [42]. The high dielectric constant of $BaTiO_3$ nanoparticles greatly enhances the dielectric constant of the composite film and thus gate strength, which can lead to high performance printed transistors. Figure 4d shows the above gravure printed TFTs being integrated into a 20×20 pixel active-matrix backplane, which can be used to drive displays and e-skin. The printed transistors in the active-matrix backplane exhibit high performance with small operating voltage (< 10 V), large on/off ratio (> 10^4) and a good field-effect mobility of ~9 cm^2/Vs as shown in Figure 4e. In addition to the printed TFT array, integrated arithmetic logic circuits such as logic gates, half adder, D-flip-flop and one-bit radio-frequency identification (FRID) tags have all been successfully demonstrated using the gravure printing process [51-53].

B. Printed Intrinsically-Stretchable Transistors

For potential applications in wearable/implantable health monitoring and diagnostic devices, it is desirable to have stretchable e-skin built on soft elastic substrates instead of plastic substrates in order to achieve conformal body surface coverage and minimize motion artifact [3, 5, 6, 57]. Through the use of intrinsically stretchable elastomeric or composite electronic materials that are solution processible, the printing process can also enable large-area and low-cost fabrication of high-performance stretchable electronic devices on elastic substrates such as polydimethylsiloxane (PDMS) [35, 58, 59]. Compared with the silver nanoparticle ink used above for the electrodes, carbon nanotube thin film is a perfect choice for stretchable electrodes because of its ultrahigh aspect ratio and the formation of highly deformable mesh structures in macroscale assemblies [60]. As shown in Figure 5a, dense unsorted carbon nanotubes can be printed to form the source/drain/gate electrodes, and monolayer of high purity semiconducting single-walled carbon nanotubes (sSWCNTs) can be printed as the channel semiconductor in the TFT [61]. Similar to the concept of the $BaTiO_3$/PMMA hybrid dielectric used in the printed flexible TFT above, the gate dielectric for the stretchable TFT is a composite consisting of PDMS, the same material as the stretchable substrate, and $BaTiO_3$ that offers high dielectric constant. The dielectric constant of the composite PDMS/$BaTiO_3$ gate dielectric increases

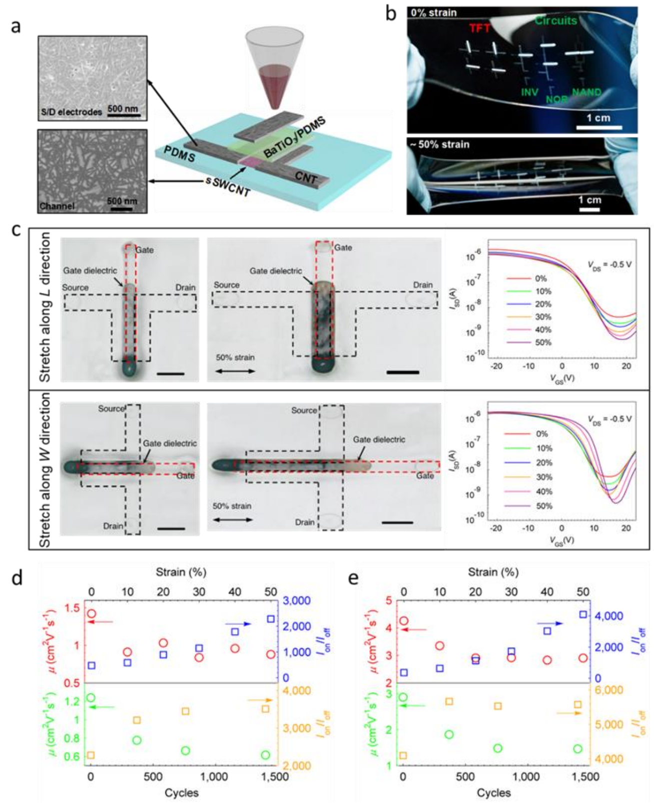

Fig. 5. Printed intrinsically-stretchable carbon nanotube TFT. (a) Schematic illustrating the structure of a printed stretchable TFT. (b) Optical photograph of a representative sample consisting of printed TFTs and logic circuits in relaxed state (top) and under a tensile strain of ~50% (bottom). (c) Optical micrographs and transfer characteristics of two printed TFTs when stretched along channel length and channel width directions to a tensile strain of up to 50%. Scale bar: 1 mm. (d, e) Carrier mobility and on/off current ratio of the printed stretchable TFT as functions of strain and number of stretching cycles when the device is stretched along the channel length (d) and channel width (e) directions, respectively. Reproduced with permission from ref.61. Copyright 2016 American Chemical Society.

monotonically with increasing volume ratio of $BaTiO_3$ dispersed in PDMS and it goes up to ~9 at 26%. This is significantly better than the dielectric constant of ~ 2 in pure PDMS and will help increase the gate strength and reduce the operating voltage in printed transistors. Figure 5b shows the photographs of a representative sample with printed stretchable TFTs and integrated logic circuits that can be stretched by up to 50%. The transistors are able deliver excellent performance and mechanical robustness, with minimal change in transfer characteristics in both relaxed and stretched states as shown in Figure 5c. Key figures of merit of the transistors including carrier mobility and on/off current ratio can be extracted from the transfer characteristics and the results are shown in Figure 5d and 5e. Such printed stretchable transistors exhibit average mobility of around 4 $cm^2V^{-1}s^{-1}$ and on/off ratio greater than 500 with maximum values of 7 $cm^2V^{-1}s^{-1}$ and 3000, respectively, which is respectable for all-printing process. Regardless of the stretching direction (along the channel length or channel width directions), the mobility decreases by about 1/3 of its original value when the device is under 50% tensile strain, which can be attributed to the microscopic structural changes in the sSWCNT networks. On the other hand, the on/off ratio increases by almost 1 order of magnitude at 50% strain due to the significantly suppressed off-state current. The devices have been tested for up to 1500 stretching cycles and the device characteristics remain relatively stable.

C. Printed E-skin with Active Matrix Circuit for Tactile Sensing

The printed carbon nanotube TFTs have also been incorporated in functional electronic systems such as e-skin with monolithically integrated pressure sensor arrays. Using the gravure printed carbon nanotube TFT active-matrix backplane described above, a large-area compliant tactile sensor array has been successfully demonstrated by integrating the printed TFT backplane with pressure sensitive rubber (Figure 6a) [62]. The active-matrix backplane consists of up to 400 TFTs (Figure 6b) with high yield (97%) and excellent uniformity. The system is capable of detecting pressures ranging from 1 kPa to 20 kPa with a linear sensitivity of 800 %/kPa. In addition, owing to the mechanical compliance of the hybrid dielectrics, the active-matrix backplane is highly flexible with invariant electrical characteristics when bent to a curvature radius of 1.85 cm. As demonstrated in Figure 6c, spatial mapping of the pressure profile can be achieved (Figure 6d).

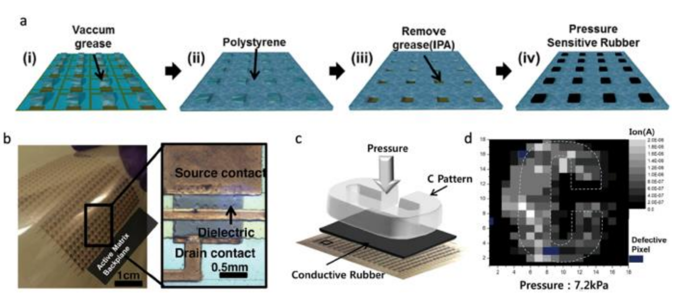

Fig. 6. Gravure printed carbon nanotube TFT backplane for applications in e-skin. (a) Schematics illustrating the fabrication process where the pressure sensitive rubber is monolithically integrated with the TFT backplane. (b) Printed flexible active-matrix backplane with 20×20 pixels. Inset: Enlarged view showing a single printed TFT. (c, d) E-skin with printed active-matrix backplane for spatial mapping of the applied pressure profile. The measured current in each pixel corresponds to the magnitude of the applied pressure. Reproduced with permission from ref.62. Copyright 2015 John Wiley and Sons.

IV. Printed Physical Sensing E-Skin

A. Printed Strain and Temperature Sensor

Similar to the printed flexible and stretchable transistors discussed above, printed flexible sensors are also an important technology for low-cost, macroscale electronics. To address the challenge of replacing conventional vacuum-based semiconductor fabrication processes, printed sensor made with inks of organic and/or inorganic materials have been reported[13, 20, 63-66].

Nanocarbon-based strain and tactile force sensors are widely studied due to their mechanical flexibility and network structure on flexible films[67]. These sensors have mainly three types of detection mechanism of resistive[68, 69], capacitive[70, 71], and piezoelectric[72] changes. In addition to the mechanical stretchability, by choosing the materials, biodegradable strain and pressure sensors have been reported[73].

For the temperature sensor, transistor-based sensor and resistive type sensor are often developed[18, 74]. Other important factor is selectivity to measure temperature precisely without affecting the strain caused by bending and stretching the films. One of the approaches has been studied by utilizing the stretchable circuits with strain suppression functionality[75]. Although many types of flexible/stretchable sensors have been proposed, specific examples of inorganic nanomaterial-based printed strain and temperature sensors are introduced in this section.

For printed strain sensor, silver (Ag) nanoparticles (NPs) and CNTs inks were mixed together and used to monitor the resistance change as a function of applied strain (Figure 7a) [66].

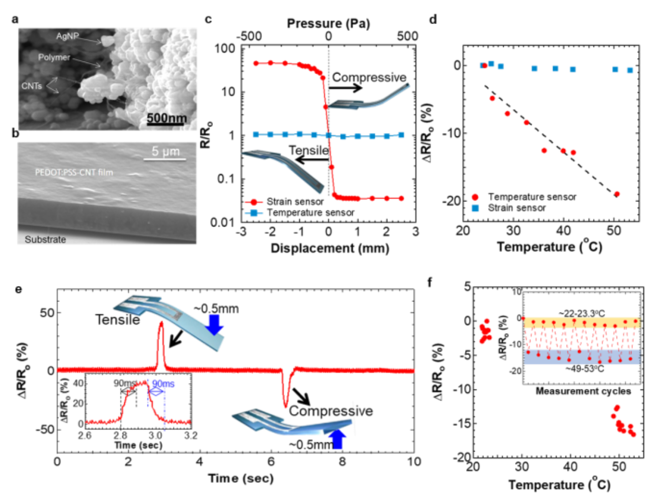

Fig. 7. Printed flexible sensors. SEM images of (a) the strain sensor and (b) the temperature sensor. (c) Resistance change by bending the film for both strain and temperature sensors. (d) Resistance change ratio at different temperature for each sensor. (e) Real-time resistance change monitoring by applying tensile and compressive strain. Inset shows the response speed of response and relaxing time. (f) Repeatable resistance change ratio as a function of temperature. Reproduced with permission from ref.66. Copyright 2014 American Chemical Society.

The underlying mechanism of the observed resistance change under tensile strain is due to the change in tunneling current between the AgNPs. The role of the CNT is to electrically bridge the AgNPs under high tensile strain in order to extend the dynamic range of the sensor. The CNT/AgNP ink was screen-printed and cured at 70 ºC to form the strain sensor. For temperature sensor, the CNT ink was mixed with conductive polymer solution of poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) and printed (Figure 7b) [76]. The sensing mechanism for temperature relies on electron hopping at the junction between two different materials (CNT and PEDOT:PSS).

Figure 7c shows the fundamental electrical property of the strain sensor on 0.5 mm-thick silicone rubber with 8 mm length. The resistance of the sensor increases when tensile strain is applied, which is resulted from the enlarged distance between the AgNPs than that of the device in relaxed state. On the other hand, under compressive strain, the distance between the AgNPs becomes smaller, resulting in decreased resistance. The observed sensor response is in good agreement with the trend of probability of tunnel current flow between AgNPs depending on the AgNP distance. The resistance change sensitivity of this structure with 4:5 wt% ratio of PEDOT:PSS and CNT inks is ~59 %/Pa. In the case of the temperature sensor, which does not contain AgNPs in the printed film, the resistance change is very small under strain conditions and the corresponding sensitivity is ~0.02 %/Pa (Figure 7c). On the other hand, the temperature sensor shows relatively high sensitivity of up to ~0.63 %/ºC for the temperature change ranging from 0 ºC to 60 ºC (Figure 7d) while the strain sensor has low sensitivity of 0.03 %/ ºC for temperature. Based on the results above, the high selectivity allows the strain and temperature sensors to be integrated together without interfering each other. Real-time monitoring and repeatability of the strain sensor were analyzed by applying tensile and compressive strain. Figure 7e indicates that the response and release time of strain sensor were less than 90 ms, which is limited by the speed of bending modulated by a hand manually. In addition, the sensor responses were robust and repeatable with stable output for at least 200 cycles of tensile strain. For printed temperature sensor, the response time depends on the film material but is usually over several seconds due to the low thermal conductivity of the polymer film used. The temperature sensor also exhibited repeatable and stable response repeated temperature changes are confirmed by the experimental results in Figure 7f. However, it should be noted that both strain and temperature sensors also react to moisture. For practical applications, passivation layer is required to protect the sensors from moisture.

B. Printed Multifunctional E-skin

Using the printed strain and temperature sensors, multi-functional printed e-skin were demonstrated, which enables simultaneous detection of tactile force, friction force, and temperature distributions in a single system[18, 20]. For printed multifunctional e-skin, nanocarbon and polymer materials including nanotubes [20, 60, 62], nanowires [77], and piezoelectric polymer [18, 78] are dispersed into printed solutions. To realize scalability and multi-functionality especially adding friction force detection, multi-layer structures with a suspension sheet, strain sensor sheet, three-dimensional fingerprint-like structure sheet, and temperature sensor sheet are fabricated as shown in Figure 8a-b. Each pixel has four strain sensors, one or two temperature sensors, and a fingerprint like structure (Figure 8c). By having the fingerprint-like structure, depending on the force directions of tactile and friction forces, strain distributions are completely different as depicted by the FEM simulation shown in Figure 8d-e. By recording the strain difference using the strain sensor, tactile force and friction force can be estimated. Furthermore, by monitoring the amplitude of strain, the force amplitude can also be extracted.

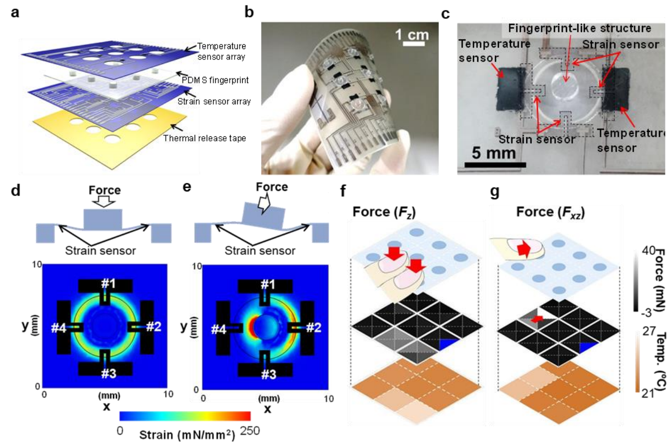

Fig. 8. Fully printed e-skin. (a) Schematic and (b) photo of the printed e-skin with multi-functionality of tactile force, friction force, and temperature monitoring. (c) Zoom-up image of a pixel integrated with 4 strain sensors, 2 temperature sensors, and a fingerprint-like structure. (d) FEM simulations of strain distribution at (d) tactile force and (e) friction force. (f) Tactile and (g) friction force detections using 3×3 pixel array with temperature distributions. Reproduced with permission from ref.20. Copyright 2014 American Chemical Society.

Finally, as a proof-of-concept of the fully printed e-skin with multi-functionality, a 3 × 3 array was demonstrated. To confirm its capability of simultaneously detecting tactile, friction, and temperature distribution, two types of stimuli F_Z and F_{XZ}, representing tactile and friction forces from fingers, respectively, were applied over the fingerprint-like structure. As shown in Figure 8f-g, when F_Z force was applied, all four strain sensors for each pixel indicated almost same value. In contrast, when friction force F_{XZ} was applied, asymmetric responses were observed in the four strain sensors in the pixel. These results are in good agreement with the trend of stress difference at each force direction extracted from the FEM simulations (Figure 8d-e). In addition, temperature distributions were also successfully monitored simultaneously due to temperature difference between room environment and fingers.

V. Chemical Sensing E-Skin

Most wearable sensors reported so far focus on monitoring of the vital signs and physical activities, and fail to provide the individual's health state at molecular levels. Continuous and real-time monitoring of molecular information is strongly desired for accurately evaluating the user's health conditions. The blood analysis in traditional clinical settings relies on invasive blood draws and cannot provide real-time and

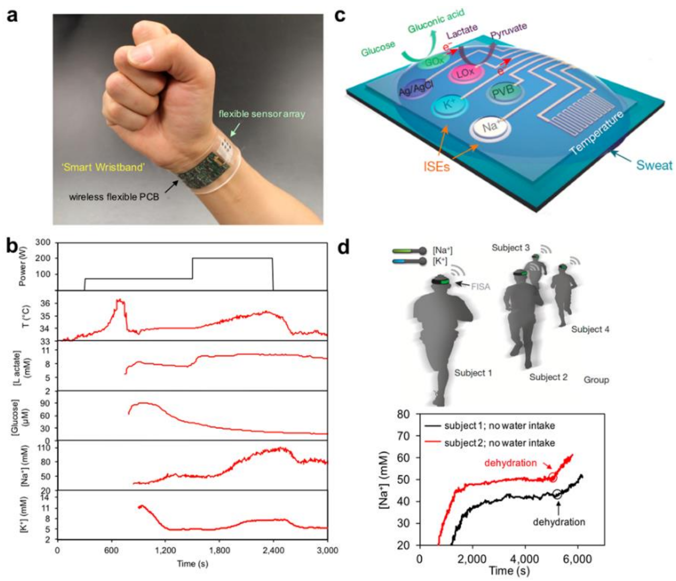

Fig. 9. Wearable sensor for multiplexed *in situ* perspiration analysis. (a) A fully integrated flexible sweat sensor array. (b) Schematic of a sensor array for monitoring of sweat metabolites, electrolytes as well as skin temperature. (c) Real-time sweat analysis using during a cycling exercise. Reproduced with permission from ref. 7. Copyright 2016 Nature Publishing Group.

continuous information. Human sweat is an important body fluid that can be retrieved conveniently, continuously, and non-invasively. It contains a wealth of chemicals including metabolites, electrolytes, hormones, peptides, and proteins that can reflect the body's physiological state [10, 79, 80]. For example, sweat glucose is currently being intensively explored for non-invasive glucose monitoring [81]; sweat sodium could be an attractive candidate for monitoring dehydration [82]; sweat chloride is the gold standard for cystic fibrosis diagnosis[83]; sweat test holds great promise in substance abuse monitoring [84, 85]. The transition from invasive blood analysis to continuous sweat analysis could provide a noninvasive and more attractive means of continuous health monitoring in daily life. In the past three years, tremendous progress has been made toward real time and continuous monitoring of a broad spectrum of analytes metabolites (e.g., glucose, lactate, creatinine), electrolytes (e.g., Na^+, K^+, NH_4^+, Cl^-, pH, Ca^{2+},), heavy metals (e.g., Cu, Zn, Pb, Hg, Cd), and small substances (e.g., alcohol and caffeine) [7][9-13][58-61]. The main detection approaches are primarily based on either electrochemical sensing or colorimetric sensing [9][11]. Bioaffinity sensors can be used for detecting a wide range of proteins, peptides, and hormones, but require further development for deployment in wearable platforms due to the challenges of in situ regeneration [53][54].

A. Chemical Sensor for in situ Multiplexed Perspiration Analysis

Given the complexity of sweat secretion, multiplexed detection of target analytes of interest is in urgent need. A fully integrated wearable sensor array was recently developed for *in situ* perspiration analysis (Figure 9a) [7]. Packaged in a wristband or a headband, this mechanically flexible system has conformal contact with the skin and can perform on site signal conditioning, processing and wireless transmission. The flexible system simultaneously and selectively measures sweat electrolytes (e.g., Na^+ and K^+) and metabolites (e.g., glucose and lactate), as well as the skin temperature for real time signal calibration (Figure 9b-c). The fully-integrated wearable sensor platform was successfully used to measure the detailed and dynamic sweat profile of the users in prolonged physical activities, and to make a real-time assessment of the physiological state of the individuals. To evaluate the use of this platform for non-invasive monitoring of dehydration, real-time sweat electrolyte (*e.g.*, Na^+) measurements were conducted on the subjects engaged in outdoor running trials (Figure 9d). Sweat Na^+ levels increased substantially during the exercise when the subjects lost a significant amount of water, indicating that sweat sodium can potentially be used as a key biomarker for dehydration monitoring.

B. Iontophoresis based Sweat Sensor

The inherent inaccessibility of large amount of sweat beyond physical exercise (particularly in sedentary individuals) remains to limit our ability to capitalize on the attractive rich source of physiological information for broad population monitoring. To address this problem, a very promising sweat on demand extraction method is iontophoresis, which could be used to induce sweat excretion in a small area continuously. The iontophoresis process involves the use of a small electrical current to deliver stimulating agonists (e.g., pilocarpine) to the sweat glands. A wearable sweat extraction and sensing platform has recently been demonstrated by our group which contains an integrated iontophoresis module able to induce sweat with different profiles at periodic intervals (Figure 10a) [12]. The electrochemical sensing electrodes between the two iontophoresis electrodes allows *in situ* real time analysis of extracted sweat samples (Figure 10b). The use of the sweat sensor for the diagnosis of a lung disease - cystic fibrosis - was successfully demonstrated through simultaneous detection of the elevated sweat Na^+ and Cl^- levels (Figure 10c). The wearable device was also utilized to investigate the correlation between sweat and blood analyte levels (*i.e.*, glucose). It was shown that oral glucose intake in fasting subjects resulted in a significant increase of glucose level in both blood and sweat (Figure 10d).

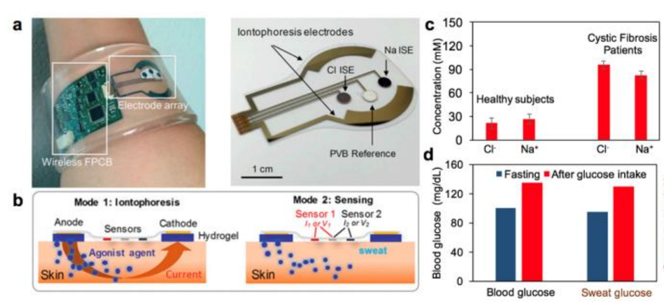

Fig. 10. Wearable sweat sensor with an integrated iontophoresis module toward autonomous sweat extraction. (a) Images of the autonomous sweat extraction and sensing platform. (b) Schematic illustrations of the iontophoresis and sensing modes. (c) Screening and diagnosis of cystic fibrosis. (d) Correlation study of sweat and blood glucose levels toward non-invasive glucose monitoring. Reproduced with permission from ref. 12. Copyright 2017 National Academy of Sciences USA.

C. Sweat Sensor for Drug Analysis

Wearable sweat sensors have great promise in real time and continuous drug monitoring for doping control and precision medicine. Drug analysis is commonly implemented for drug abuse testing, doping control, and chemotherapy. Conventional drug monitoring relies on invasive blood test and time consuming instrumental analysis. Sweat is an attractive alternative biofluid which could provide real-time information of the drug dosage and drug metabolism. A wearable sweat band was developed for noninvasive and dynamic monitoring of drug levels (Figure 11a) [88]. Caffeine was selected as a model methylxanthine drug to demonstrate the sensor's capabilities. The detection of caffeine is achieved by measuring the oxidation of caffeine through differential pulse voltammetry (DPV). Sweat caffeine levels increased with the increase of drug dosage and the confirmable caffeine physiological trends over time after the intake were observed (Figure 11b). Such wearable sweat sensor was capable of monitoring the dynamic drug levels over time (Figure 11c).

D. In Situ Sweat Sampling and Sweat Rate Sensing

Proper sweat sampling is critical to improve the temporal resolution of the sweat analysis and to minimize the errors caused by sweat evaporation and skin contamination. In addition, sweat compositions and sweat rate are found to be inextricably linked. In order to achieve enhanced sweat sampling and dynamic sweat rate monitoring, a flexible and wearable microfluidic sweat sensing patch has been developed recently [89]. As sweat travels along the microchannel, the electrical impedance measured between two parallel electrodes in the microchannel drops as the sweat volume increases due to the increase in the capacitance and the decrease in the effective resistance. Sweat rate was calculated as the sweat volume change in the microchannel divided by the time interval. System integration of chemical sensing and electro impedance measurement in a single device greatly facilitates the in situ sweat analysis. Such sweat patch could provide a comprehensive sweat secretion information for a number of fundamental and clinical investigations.

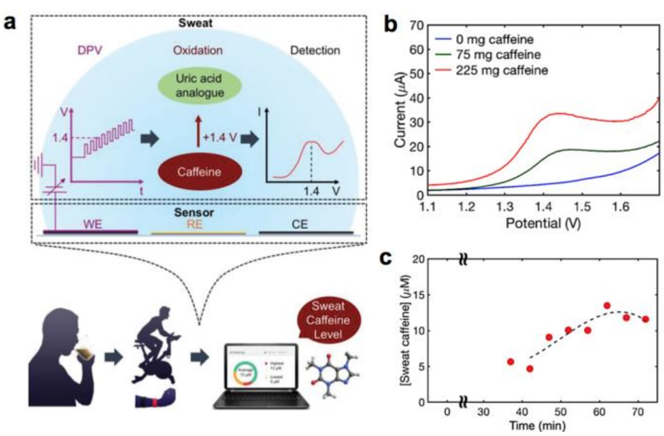

Fig. 11. Methylxanthine drug monitoring with wearable sweat sensors. (a) Electrochemical detection of caffeine through differential pulse voltammetry (DPV). Oxidation of caffeine leads to an observable oxidation peak around 1.4 V. (b) Sensor response in human sweat samples for all caffeine intake conditions. (c) A summary plot of the sweat caffeine levels over time during the exercise after caffeine intake. Reproduced with permission from ref. 87. Copyright 2018 John Wiley and Sons.

E. Fusion of Sweat Chemical Sensor and Physical Sensor

In practical biomedical applications, acquiring both molecular information of the human body, alongside physical characteristics such as vital signs, are extremely critical for accurately identifying health conditions [7]. Moreover, the measured information from one sensor could potentially be used to calibrate the readings of other sensors. For example, a flexible ion-sensitive field-effect transistor (ISFET)-based pH sensor was developed and integrated with a flexible skin temperature sensor to monitor both skin temperature to compensate the temperature effect of the ISFET for accurate measurement (Figure 12) [90]. For the ISFET, an Al_2O_3 layer was used for the pH sensing membrane while an InGaZnO thin-film was used as the n-type FET material. In moving toward the realization of personalized medicine, the integration of more functionality and sensing modality into the flexible e-skin system such as accelerometers, other vital sign and chemical sensors, and signal processing circuits, is needed.

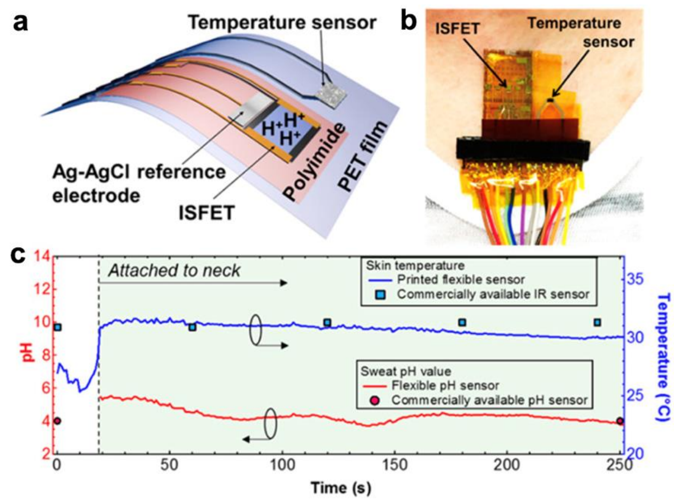

Fig. 12. Flexible and multifunctional healthcare device with an ISFET chemical sensor for simultaneous sweat pH and skin temperature monitoring. (a) Schematic of a wearable device integrating flexible pH and temperature sensors. (b) Photograph showing attachment of the flexible pH and temperature sensors to the test subject's neck. (c) Real-time pH and skin temperature information acquired by the wearable device. Reproduced with permission from ref. 90. Copyright 2017 American Chemical Society.

VI. Summary and Perspectives

In this review, we have explained and highlighted the recent progress on flexible and stretchable physical and chemical sensors for e-skin applications, with the emphasis on design and selection of materials and structures, and the integration of various types of sensor and circuits. This review also introduced a few key potential applications of the e-skin in robotics and healthcare. With its scalability, low-cost, multi-functionality, and the desirable form factor of a flexible or stretchable sheet, such physical and chemical sensing e-skin can be applied to many different applications especially for "Internet of Things (IoT)" to collect information from any surfaces or objects.

For practical application of this e-skin concept, many issues such as integrating more sensing modalities, improving device reliability and reducing motion artifacts, feedback and interaction between human and e-skin through artificial intelligence and deep learning still need to be addressed. In particular, e-skin developments are mainly conducted by academia. However, to achieve the practical platform for e-skin applied to robotics, healthcare, and industry, long-time stability and reliability including the uniformity of the fabrication should be addressed while more functionalities, power generation, and circuit systems are in parallel developed with consideration of their integrations on flexible and stretchable films. To open the technologies to the market, deep collaboration works between academia and industries are required to support each weakness of the developments. Furthermore, key applications should be proposed to use the flexible electronic systems by building new concepts or replacing conventional electronic systems[91]. Without showing this concept, it is hard to move forward to developing the flexible electronic systems. Although a lot of developments are required, we believe that the described technologies and concepts may lead us toward realizing the next class of low-cost, multi-functional, macroscale, and stretchable e-skin.

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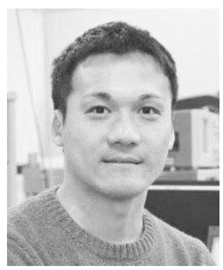

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