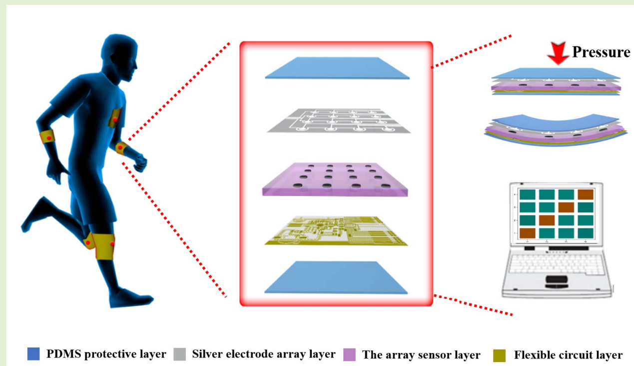
A Fully Integrated Flexible Electronic System With Highly Sensitive MWCNTs Piezoresistive Array Sensors for Pressure Monitoring

Jiagen Cheng, Yuanbin Mu, Chaoran Liu, Weihuang Yang, Wenjing Liu, Huanze Wang, Jiashu Wu, Fujing Hou, Dandan Hao, Lian Cheng, Binhui Weng, Ying Chen, and Linxi Dong

***Abstract*—Pressure ulcers are a common and costly health problem in clinical medicine, caused primarily by continuous vertical pressure on local tissues of the body. Recently, flexible devices have been used to alert pressure ulcers by continuously monitoring the pressure applied to the human skin. Although many pressure sensors provide excellent sensing performance, they still rely on uncomfortable, unstretchable systems with rigid electronics. These devices are mounted on the human body, which hinders continuous pressure monitoring. Herein, we propose a fully integrated flexible electronic system (FIFES) with highly sensitive multi-walled carbon nanotubes (MWNTs) piezoresistive array sensors for pressure monitoring, which accurately achieves external pressure signals acquisition and independently performs processing and wireless transmission. We develop a heat seal connecting technology for integrating the piezoresistive array sensors and its flexible processing circuit, which overcomes the compatibility defect of a rigid circuit board and the flexible sensors. Furthermore, the Cu–Ge metal sputtering and transferring technologies are employed to achieve an extensible circuit. The as-fabricated fully integrated electronic system has excellent flexibility, a rapid response time (the response/recovery time is 0.11/0.08 s, respectively), a wide dynamic range (6–50 kPa), and an excellent sensitivity (1.61 kPa^{-1} for the range under the pressure of 18 kPa, 0.47 kPa^{-1} for the range of 18–50 kPa) attributes to the optimized conical microstructures number. Owing to its excellent performance, the FIFES has extensive applications for pressure monitoring in different human epidermises, such as elbow bending and knee-bending. Also, it has immense potential applications in sleep posture monitoring and pressure ulcer warning.**



***Index Terms*— Flexible electronic, fully integrated, high sensitivity, pressure monitoring.**

I. Introduction

PRESSURE ulcers are one of the most common complications in patients who are bedridden; without timely treatment, the ulcer will expand followed by serious infections, sepsis, septicemia, and other major diseases [1]–[3]. Pressure ulcers are caused primarily by continuous vertical pressure on the local tissues, especially at the bony projections of the body [4]. The classical prevention of pressure ulcers requires frequent and artificial changes of body position, which help to redistribute pressure and avoid long-term localized pressure on the body [5]. However, changing postures frequently demands that the patient or caregiver remains awake at all times, which is difficult to do during such a long period of illness. Therefore, a real-time external pressure monitoring system with good flexibility, skin adaptability, and biocompatibility is urgently desired, which can be applied to the human body for pressure ulcer alarm.

Recently, flexible pressure sensors have attracted great interest due to their good flexibility, bendability, high sensitivity, and wide measurement range [6]–[9]. Compared with other flexible pressure sensors, the contact-type piezoresistive sensors are more widespread in pressure measurement systems in daily life because of their high sensitivity, wide measuring

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range, simple structure, and strong elastic recovery [10]–[13]. In general, such pressure measurement systems contain a number of pressure sensors and separate traditional rigid circuits with wires, which have many limitations, such as uncomfortable wear and inflexibility that can disrupt daily activities and bring in noise [14]–[18]. Nowadays, based on the rapid development of flexible electronic technology, numerous new wireless flexible wearable devices are mounted on various parts of the human body, including the nose, arms, wrists, and feet [19]–[26]. For example, Kim *et al.* [27] have proposed a fully flexible, battery-free, and completely wireless mode active optoelectronic system that can be tightly integrated with the epidermis to obtain various health information through the skin, with potential applications in hospital care and home diagnostics. In another instance, Chen *et al.* [28] demonstrate skin-like hybrid integrated circuits (SHICs) with stretchable sensors and commercial chips that can be easily applied to the nose for continuous respiratory monitoring owing to their excellent flexibility. However, such a fully integrated, biocompatible, scalable, wireless flexible skin-like electronic system for continuous pressure monitoring has not been reported.

Herein, unlike traditional rigid pressure monitoring instruments, we demonstrate a strategy to integrate flexible sensors with extensible circuits and fabricate a fully integrated flexible electronic system (FIFES) for external pressure monitoring. FIFES integrates signal acquisition, processing, wireless transmission, and power modules and is encapsulated with polydimethylsiloxane (PDMS), which provides good flexibility, biocompatibility, and low manufacturing cost and the overall size can be adjusted by arraying. With regard to piezoresistive sensors, we used a mixture of multiwalled carbon nanotubes (MWCNTs) and PDMS with good stretchability, biocompatibility, and mechanical tolerance as the sensitive material [29]. Early in 2008, Lu *et al.* [30] prepared carbon nanotube-PDMS composite materials with different concentration ratios and investigated their mechanical/electrical and piezoresistive properties in detail, showing good flexibility and a gauge factor (a measure of piezoresistive sensor performance) in the range of 1.38–12.4, indicating good piezoresistive properties. Kim *et al.* [31] based on this prepared a flexible pressure sensor based on a thin porous elastomeric sponge coated with a carbon nanotube network for use in human–machine interface devices. In this work, the sensitivity of MWCNTs/PDMS piezoresistive sensors has been improved by adding conical microstructures and optimizing their number and preparing piezoresistive sensor arrays by a simple injection molding process. The independently designed 3-D fully printed silver electrodes based on PDMS substrates provide the sensor with high sensitivity, fast response time, and flexibility. Benefiting from these excellent properties, FIFES can be worn on the human epidermis not only to better detect the pressure generated by finger and elbow bending but also to achieve accurate localization of external jolts. Generally, this work first introduces the structure of FIFES and preparation method and then carries out the device performance analysis and human pressure monitoring experiments, which show that the FIFES has great potential for applications in pressure ulcers warning and sleeping posture monitoring.

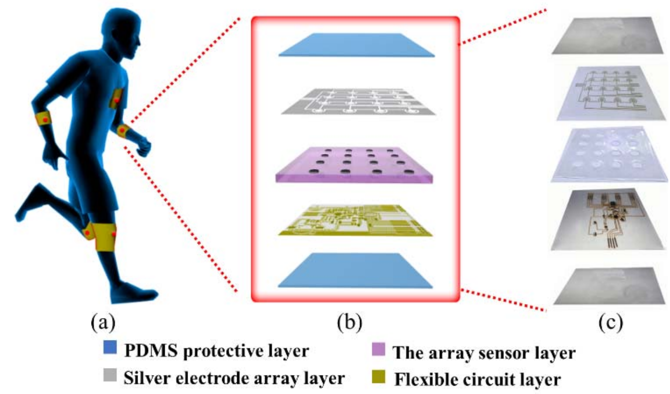

Fig. 1. (a) Illustration of the FIFES wearing on the human body to monitor epidermal pressure. (b) Five-layer structure schematic and (c) physical diagram of the FIFES.

II. Results and Discussion

A. System Structure

Fig. 1(a) shows the FIFES with a five-layer sandwich structure for monitoring external pressure. The total thickness of the manufactured FIFES is approximately 2.8 mm and uses flexible PDMS films as substrates, where the top and bottom PDMS layers with a thickness of 170 μm are used for system packaging, as shown in Fig. 1(b) and (c). Due to the excellent tensile properties, good biocompatibility, and physicochemical stability of PDMS, the system is highly resistant to stretching and comfortable to wear.

In addition, the middle three layers are the silver electrode layer, array sensors layer, and flexible circuit layer from top to bottom, respectively, where the silver electrode array layer consists of 4 $\times$ 4 silver electrode units, which are fabricated economically on 150-μm-thick PDMS substrate using 3-D microelectronic all-printing technology. It should be noted that the PDMS substrate needs to be treated with oxygen plasma several times in order to enhance its surface adhesion, and it cannot be contaminated during the whole process. Corresponding to the electrodes, the array sensors layer also has 4 $\times$ 4 circular holes for fixing 16 flexible piezoresistive sensors, which are fabricated by an injection molding process using a mixture of PDMS (PDMS main agent and the PDMS curing agent) and MWCNTs [32]. The flexible circuit layer was prepared on PDMS substrates (150 μm) by Cu–Ge metal sputtering and transfer techniques, and to improve flexibility and stretchability, serpentine interconnect wires are adopted to connect various components of the flexible circuit. Moreover, heat seal connectors are employed to connect the flexible circuit layer to the silver electrode layer, and then, the liquid adhesive is used to bond the different layers to form the final flexible electronic system, as displayed in Fig. 2(a). The front view of the flexible circuit layer and the silver electrode layer diagram [see Fig. 2(b) and (c)] illustrates that they have the same interface size, which is convenient for the integration of the system by using flexible heat seal connectors.

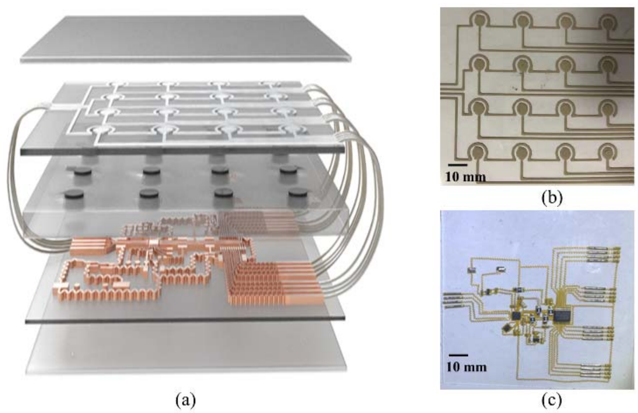

Fig. 2. (a) Structure and connection diagram of FIFES. (b) Flexible silver electrode array layer with 4 × 4 electrode units. (c) Flexible circuit physical diagram.

The size of the final integrated fully electronic system is 10 cm × 10 cm [see Fig. 3(a)], and the diameter of the piezoresistive MCNT sensitive unit with conical microstructures is 10 mm, and it is worth noting that the diameter and height of the conical microstructure are both approximately 1 mm as displayed in Fig. 3(b). Fig. 3(c) shows the excellent bendability and reliability of FIFES, which can still operate normally and perform pressure acquisition, conversion, and wireless transmission during bending and flattening. Fig. 3(d) presents the layout of the circuit, which is realized by integrating commercial chips into the flexible circuit layer using the reflow soldering technique. As marked in the block diagram [see Fig. 3(e)], the integrated circuit includes six modules. More specifically, the resistance of the piezoresistive sensor is first measured by a voltage divider and converted into an analog signal, which is sent to an analog-to-digital converter (ADC) through a multiplexed analog switch for further conversion and processed in a microcontroller (MCU). Then, the processed signal is transmitted to the mobile phone by an ultralow power Bluetooth system on a chip (BLE SOC). In terms of power supply, a button battery (3.7 V) is used as the energy source. It should be noted that the MCU works in ultralow power mode to save energy when no pressure signal is monitored (comprehensive circuit schematic is shown in Fig. S3 in the supplementary information).

B. Working Mechanism

The working mechanism of the piezoresistive sensor is shown in Fig. 4(a), in which the contact area between the conical microstructure and the electrode unit is changed by external pressure. Specifically, as the pressure increases, the particle clearance between the MWCNTs materials will decrease, while the contact area of the conical microstructure and conductive electrode unit will increase, which simultaneously leads to a corresponding decrease in the resistance of the flexible pressure sensor. By reading this changing resistance and transmitting it wirelessly to a cell phone and personal computer, the pressure information on the human epidermis can be observed graphically [see Fig. 4(b)].

C. Testing the Performance of Sensors

Accurate measurement of pressure depends on the excellent performance of the flexible pressure sensor, such as high sensitivity, rapid response time, and low hysteresis [33]. Therefore, the characteristics related to the prepared flexible piezoresistive sensors are studied, and the quantity of microstructures of the piezoresistive sensor is optimized with the aim of obtaining better performance. Fig. 5(a) and (b) displays the model diagram and the physical diagram of the test environment built in this study. Specifically, a piezoresistive sensor is placed on a microcomputer-controlled electronic universal testing machine iWDW-05j to perform the piezoresistive experiment, where the stretcher applies regulated force and the resistance is measured by a precision *LCR* meter (TH2838).

First, the response time and recovery time of the piezoresistive sensor were evaluated, and they were about 110 and 80 ms, respectively [see Fig. 5(c)]. This response speed is sufficient to meet the requirement to monitor the external pressure applied on the human skin to cause pressure ulcers, which require constant rather than rapid, brief pressure [34].

Sensor sensitivity constrains the accuracy of pressure monitoring. Fig. 5(d) plots the relationship between the resistance change of the designed sensor and the applied force, where the red curve and the blue curve correspond to the resistance response curves when the loading/unloading is applied, respectively. It is clearly shown in Fig. 5(g) that when the applied force is greater than 6 kPa, the sensor resistance begins to decline sharply, and the resistance changes relatively slowly when it exceeds 18 kPa until it is saturated. The main reason for this phenomenon is that the microstructure contact area and the particle clearance have varying degrees of effect on the resistance change when the same force is applied. People will feel uncomfortable when the pressure applied to the human skin exceeds 20 kPa [35]–[37]. Hence, pressure sores may occur when the pressure exceeds 20 kPa. The pressure sensor in this article has a measurement range of 6–50 kPa. So, it can meet the requirements. Therefore, by choosing a suitable working range, it is sufficient to be used to accurately monitor various external pressures from the human epidermis.

Stability and repeatability are important for the reliable operation of the sensor. To confirm the stability of the designed sensor, a cyclic test was performed by applying a force of 18 kPa on the sensor over 1000 times in 5000 s [see Fig. 5(e)]. It shows higher stability of the developed sensor even after a 5000 s continuously applied pressure (18 kPa), which has an accuracy of 96.4% (an absolute error is $\pm 20\ \Omega$ and the true predicted value is $560\ \Omega$) [38].

Furthermore, the consistency of multiple samples was studied, as displayed in Fig. 5(g), and S1–S8 represent eight flexible piezoresistance sensor samples of the same batch. Fig. 5(h) describes the resistance response curves of these eight sensors when loading and unloading under the same applied force, suggesting that they have a good consistency.

Finally, the sensitivity of piezoresistance sensors with different numbers of conical microstructures was studied. Fig. 5(f) and (i) corresponds to the resistance response curve and sensitivity fitting curve of the sensor, respectively, and each curve includes two working ranges [the slope of the curve

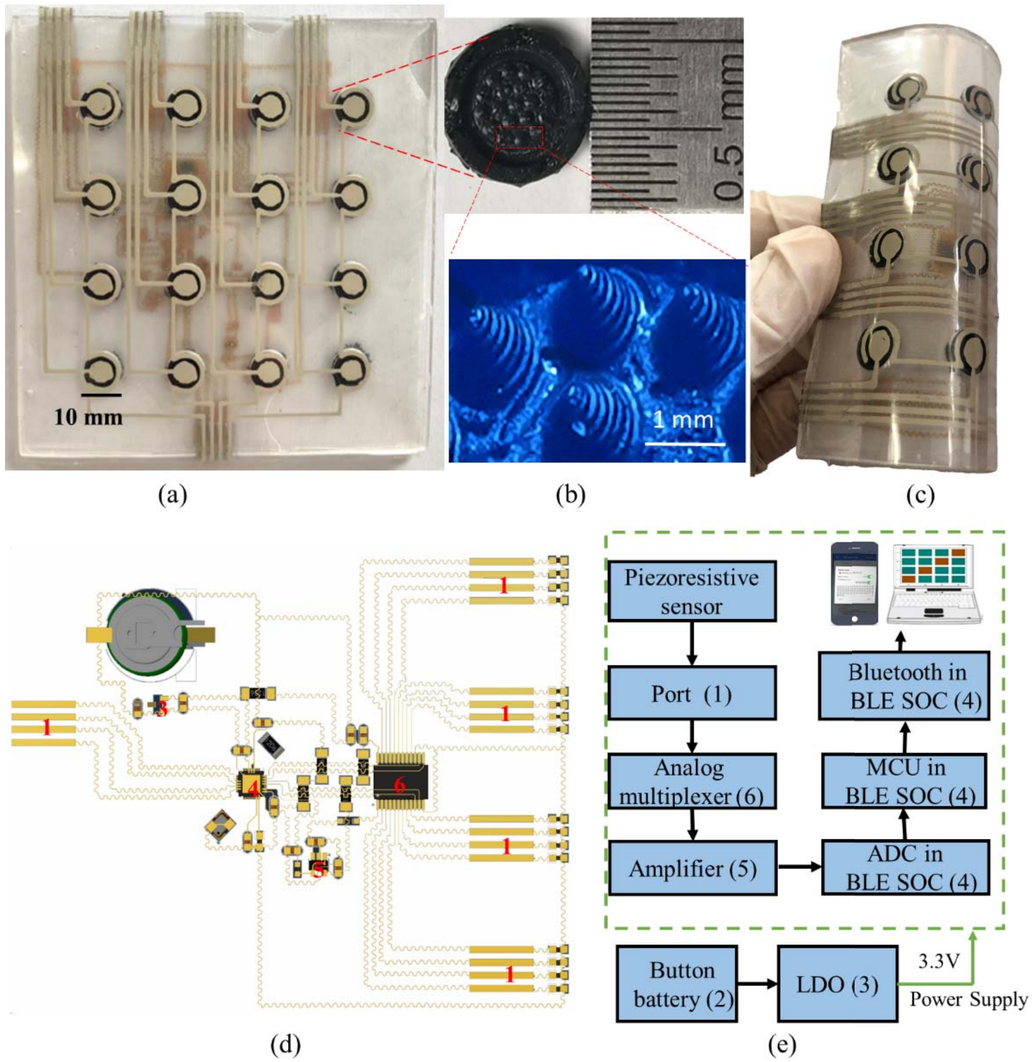

Fig. 3. (a) Physical drawing of FIFES (size: 10 cm × 10 cm) and (b) its built-in flexible piezoresistive sensor (diameter: 1 cm). (c) Bending the FIFES demonstrates its flexibility. (d) Flexible circuit physical diagram. (e) Circuit function block diagram.

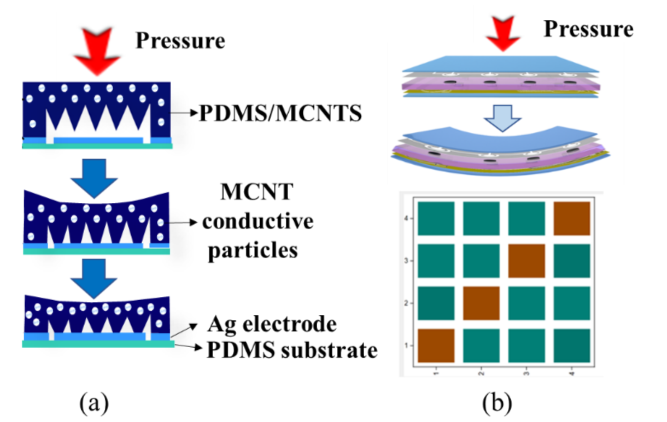

Fig. 4. (a) Working mechanism diagram of MWCNTs piezoresistive sensor and (b) FIFES.

in Fig. 5(i) represents the sensitivity in units of kPa^{-1}, the x-axis represents the pressure in units of kPa, and the y-axis represents the dimensionless current variation ratio, so take the 18 cone structures as an example, the fitted sensitivities are 1.61 kPa^{-1} for the range under the pressure of 18 kPa, 0.47 kPa^{-1} for the range of 18–50 kPa], which is in accordance with the working mechanisms of the piezoresistance sensor described above. As shown in Fig. 5(i), it can be concluded that the larger the number of conical structures, the higher the sensitivity of the sensor. Therefore, to obtain better performance, the number of conical microstructures should be as much as possible when preparing the sensor.

III. Experimental Section

A. Assembly and Fabrication

The assembly process of the FIFES consists of four parts, which are preparation and integration of the PDMS protective layer, silver electrode array layer, array sensor layer, and the flexible circuit layer, respectively. PDMS protective layer is prepared using the main agent and curing agent (Dow Corning DC184, USA) with a mass ratio of 10:1, which mainly includes the steps of magnet mixing, spin coating, vacuuming, and curing. To improve the comfortability and stretchability of the system, the silver electrode layer was prepared using PDMS film and composite nanosilver ink as

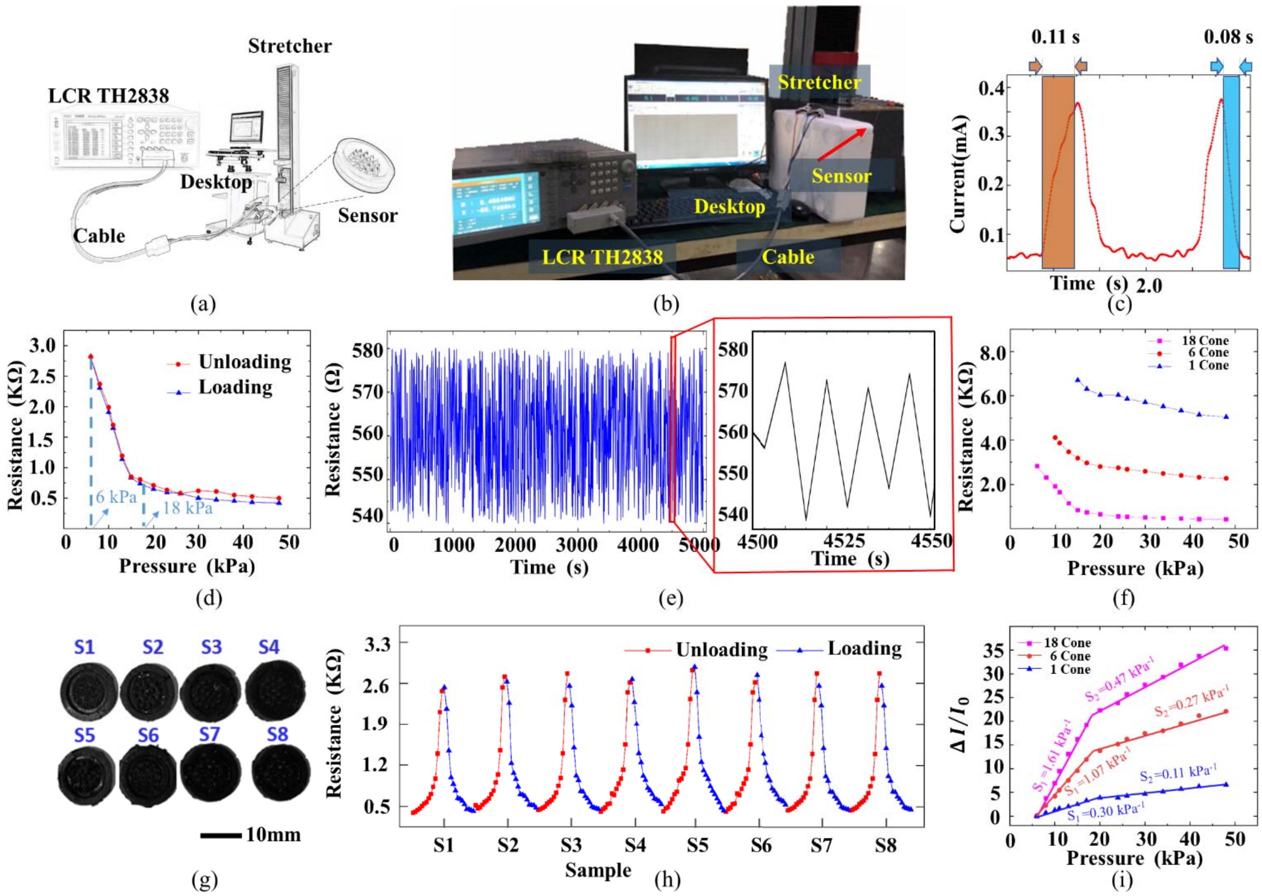

Fig. 5. (a) Flexible piezoresistive sensor model. (b) Physical diagram of the test environment. (c) Flexible piezoresistive sensor response time and recovery time. (d) Hysteresis test for flexible piezoresistive sensors. (e) Flexible piezoresistive sensor was cycled about 1000 times at 18 kPa. (f) Piezoresistive characteristics of the flexible piezoresistive sensors with conical microstructures are 1, 6, and 18, respectively. (g) Eight samples of flexible piezoresistive sensors and (h) their consistency experiments. (i) Sensitivity of the flexible piezoresistive sensors with conical microstructures is 1, 6, and 18, respectively, under different applied forces.

substrate and sensitive material, respectively (Fig. S1 in the supplementary information). It is important to emphasize that the preparation was carried out using an economical microelectronic all-printing technology. Considering the hydrophobicity of PDMS, the PDMS film needs to be first soaked in polyvinyl alcohol (PVA) solution (1% wt) for 1 min and dried with nitrogen, followed by 10-min treatment with an oxygen plasma processor (100-W power, 100-sccm oxygen flow rate).

The array sensor layer is formed with 16 flexible piezoresistive sensors, which are embedded into a porous PDMS film (2 mm), where the PDMS film is prepared with a 3-D printed mold and other steps are similar to the preparation process of the PDMS protective layer. The manufacturing process of the flexible piezoresistive sensor with conical microstructures is detailed in the following steps (Fig. S2 in the supplementary information). First, PDMS, MWCNTs, and curing agent with a mass ratio of 9:1:1 are stirred thoroughly with a magnetic stirrer at 180 r/min for 3 h to obtain a homogeneous mixture. Note that it is necessary to add some cosolvent, such as hexane, to ensure the homogeneity of the mixture. Then, the mixture was placed in a vacuum chamber for 30 min to eliminate the bubbles generated by stirring. Finally, it is injected into the mold of the piezoresistive sensor and cured in a thermostat at 90 °C for 2 h to obtain the final piezoresistive sensor with conical microstructures [32].

Fig. 6 describes the process of fabrication for flexible circuits. First, 1% poly (methyl methacrylate) (PMMA) solution was spin-coated at 500 r/min on a clean silicon wafer by a spin coater and then baked on a heating table for 25 min. The same method was employed to spin coat the 15% polyimide (PI) solution on the cured PMMA and cured it for 120 min using a heating table. Subsequently, 30-nm-thick Ge and 100-nm-thick Cu are sputtered in turn on the PI-PMMA-Si carrier using magnetron sputtering. Finally, the flexible circuit layer is obtained on the PDMS substrate by transferring the circuit from the silicon substrate to a temporary flexible film with transparent tape and removing the extra PI film with reactive-ion etching (RIE).

The final assembled FIFES was bonded by transparent adhesive, and the circuit layer was connected to the array electrode layers by heat-sealed connectors.

B. Function Verification

To verify the aforementioned sensor performance, some examples of FIFES applications are shown in Fig. 7. First, four cylindrical iron blocks of the same mass and shape are randomly placed on the sensors of FIFES, and the pressure

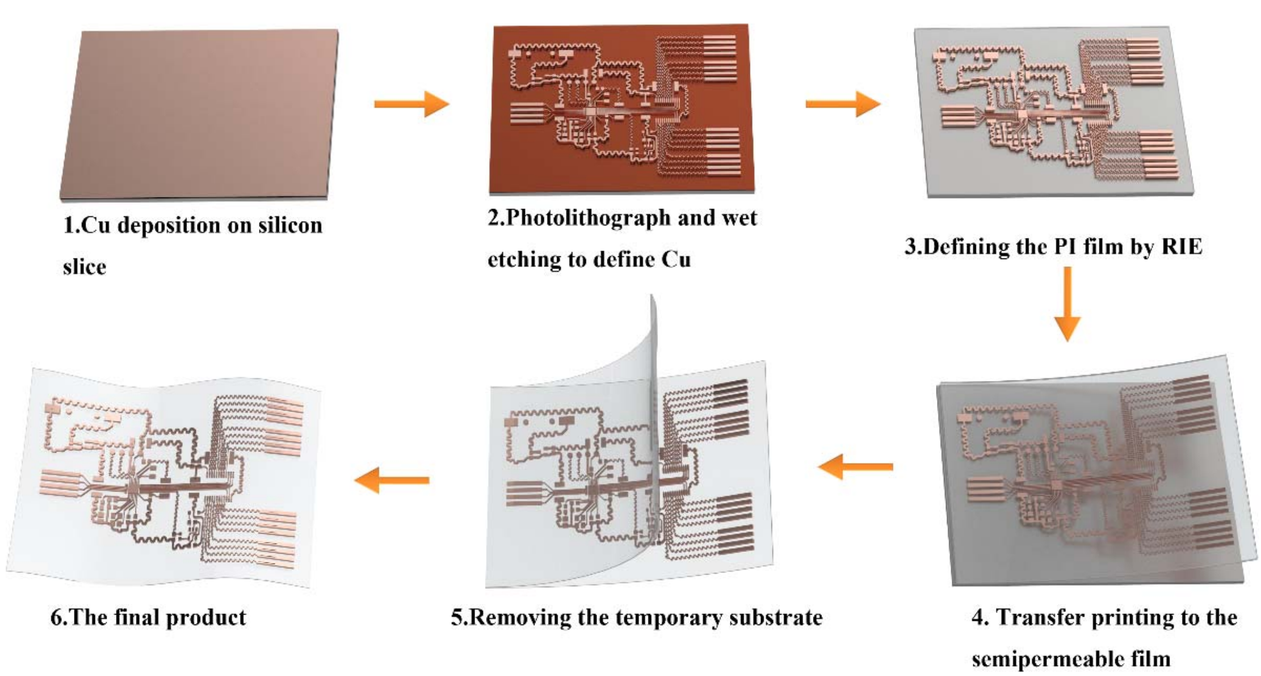

Fig. 6. Flowchart of flexible circuit preparation based on Cu–Ge sputtering process.

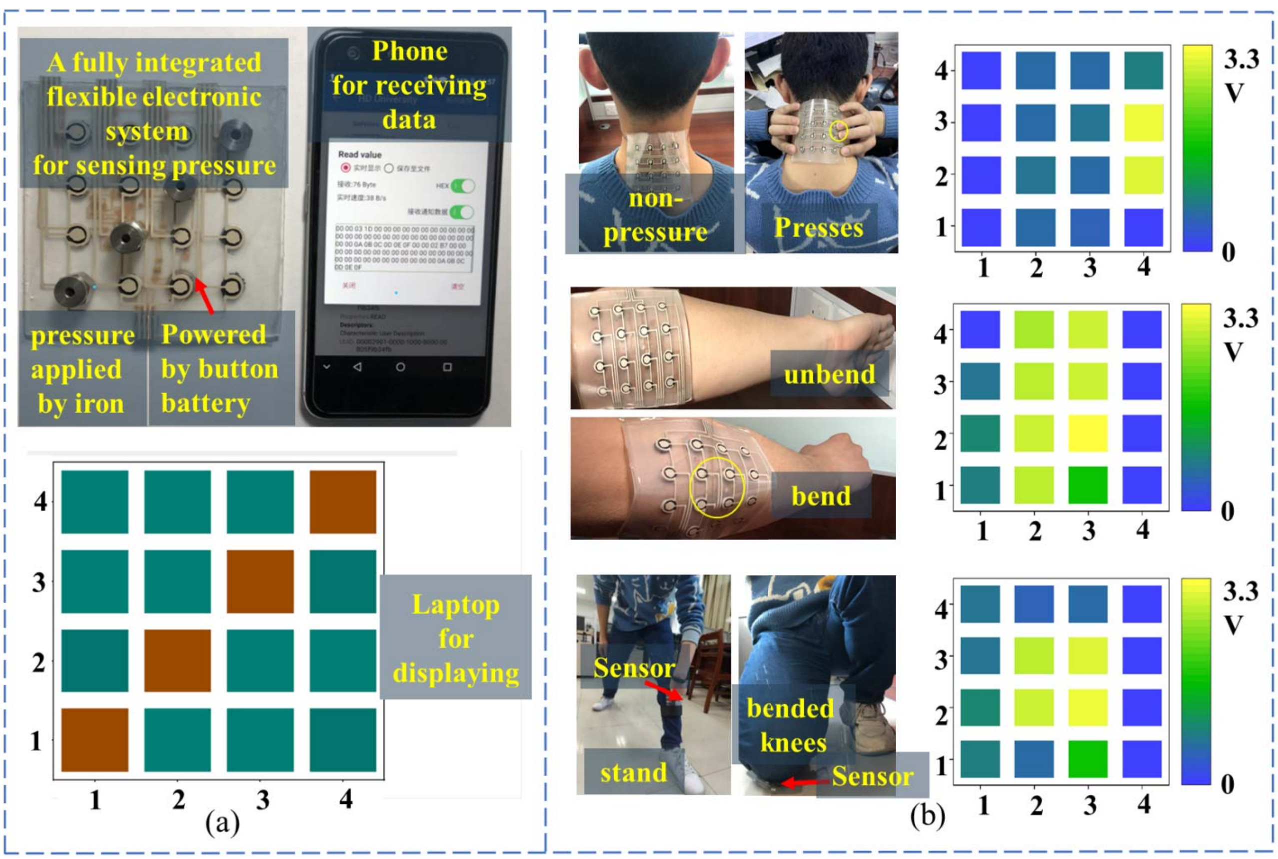

Fig. 7. (a) Applications of the FIFES for pressure monitoring. A monitoring and display for the load pressure of the same mass and shape by the FIFES system. (b) Monitoring and display for different sizes of external pressure from finger touching or joint bending.

sensing data from all sensors are transmitted wirelessly to self-customized software of cell phones and personal computers and displayed visually through algorithms processing [see Fig. 7(a)]. Eventually, the mapping gram shows equal pressure magnitude and the pressure mapping positions are consistent with the placement of the iron blocks, indicating that the designed sensors can accurately sense the force magnitude at different positions. In fact, by further optimizing the size of the sensors, the system can achieve more accurate pressure positioning. Then, the FIFES system was worn in a different part of the body and monitored for various pressures applied to the system, such as finger touching, elbow, and knee bending [see Fig. 7(b)]. Similarly, when external pressure is applied, the sensor resistance change is directly converted into a voltage of 0–3.3 V by a flexible circuit and displayed visually on the self-customed software, noting that the different color magnitudes in the mapping diagram indicate the degree of force applied. It can be seen from the mapping diagrams that the designed FIFES can better distinguish the various external pressure magnitudes due to the existence of the conical microstructures. The above experiments prove that FIFES effectively detects external pressure and has potential applications in the prevention of pressure ulcers caused by continuous external vertical pressure.

IV. Conclusion

In summary, we have developed and evaluated the characteristics of an FIFES based on a highly sensitivity MWCNTs piezoresistive array sensor, which can be used for external pressure monitoring. The FIFES contains MWCNTs flexible piezoresistive sensors based on conical microstructures and extensible circuits fabricated by sputtering and transfer technology. It effectively overcomes the limitations of pressure measurement systems with rigid circuit boards, such as unstretchability, unwearability, and uncomfortability. Owing to the robust electrochemical stability of MWCNTs/PDMS, the piezoresistive sensors exhibit excellent sensing performance, such as fast response time (0.11/0.08-s response/recovery time, respectively) and wide dynamic range (6–50 kPa). Moreover, the sensitivity of the flexible piezoresistive sensor is further improved by optimizing the number of cone microstructures, which is up to 1.61 kPa^{-1} for the range under the pressure of 18 kPa and 0.47 kPa^{-1} for the range of 18–50 kPa. External pressure experiments have proved that this FIFES can independently perform pressure signal acquisition, processing, and wireless transmission as well as showing its unlimited potential for pressure sore warning and sleeping posture monitoring.

Acknowledgment

Jiagen Cheng, Chaoran Liu, Weihuang Yang, Wenjing Liu, Huanze Wang, Jiashu Wu, Fujing Hou, Dandan Hao, Lian Cheng, and Binhui Weng are with the Smart Microsensors and Microsystems Engineering Research Center, Ministry of Education, College of Electronics and Information, Hangzhou Dianzi University, Hangzhou 310018, China (e-mail: liucr@hdu.edu.cn; yangwh@hdu.edu.cn).

Yuanbin Mu is with the Smart Microsensors and Microsystems Engineering Research Center, Ministry of Education, College of Electronics and Information, Hangzhou Dianzi University, Hangzhou 310018, China, and also with the Internet of Things Technology Research Institute, Hangzhou Polytechnic of Science and Technology, Hangzhou 311402, China.

Ying Chen is with the Zhejiang Tsinghua Institute of Flexible Electronics Technology, Nanhu, Jiaxing 314000, China (e-mail: chenying@ifet-tsinghua.org).

Linxi Dong is with the Smart Microsensors and Microsystems Engineering Research Center, Ministry of Education, College of Electronics and Information, Hangzhou Dianzi University, Hangzhou 310018, China (e-mail: donglinxi@hdu.edu.cn).

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Jiagen Cheng was born in Poyang, Jiangxi, China, in 1996. He is currently pursuing the master's degree with Hangzhou Dianzi University, Hangzhou, China.

His research interests include the integrated circuit, sensor systems, and wearable electronics device.

Yuanbin Mu was born in Tai'an, Shandong, China, in 1986. He received the M.S. degree in signal and information processing from the School of Electronic Information, Zhejiang Sci-Tech University, Hangzhou, China, in 2014. He is currently pursuing the Ph.D. degree with Hangzhou Dianzi University, Hangzhou.

Since 2014, he was a Research Assistant with the Institute of Internet of Things Technology, Hangzhou Polytechnic of Science and Technology, Hangzhou. His research interests include micro-electro-mechanical system (MEMS) sensors, flexible wearable electronics, and wireless sensor networks.

Chaoran Liu received the B.S. and M.S. degrees from Zhengzhou University, Zhengzhou, China, in 2011 and 2014, respectively, and the Ph.D. degree from the Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences, Shanghai, China, in 2017.

He became a Lecturer with the College of Electronics and Information, Hangzhou Dianzi University, Hangzhou, China, in 2017. His research interests include energy harvesting, and flexible self-powered and traditional micro-electromechanical system (MEMS) sensors.

Weihuang Yang received the Ph.D. degree in condensed matter physics from the College of Physical Science and Technology, Xiamen University, Xiamen, China, in June 2013.

He finished the Postdoctoral Research from Nanyang Technological University, Singapore, in 2016. His major research pursuits are growth and properties of 2-D materials, optoelectronic devices and flexible micro/nanosensors based on 2-D materials, and high Al-content AlGaN-based multiple quantum wells UV LEDs.

Wenjing Liu, photograph and biography not available at the time of publication.

Huanze Wang, photograph and biography not available at the time of publication.

Jiashu Wu, photograph and biography not available at the time of publication.

Fujing Hou, photograph and biography not available at the time of publication.

Dandan Hao, photograph and biography not available at the time of publication.

Lian Cheng, photograph and biography not available at the time of publication.

Binhui Weng, photograph and biography not available at the time of publication.

Ying Chen received the B.Sc. degree from the Department of Mechanics, Southwest Jiao tong University, Chengdu, China, in 2008 and 2012, respectively, and the Ph.D. degree from the Department of Engineering Mechanics, Tsinghua University, Beijing, China, in 2012 and 2017, respectively.

She works as a Research Assistant with the Institute of Flexible Electronics Technology of THU, a high-end research institution based in Zhejiang, China. Her research interests include the application of flexible electronic technology and its application in health care.

Linxi Dong received the Ph.D. degree in microelectronics and solid state electronics from Zhejiang University, Hangzhou, China, in 2004.

From 2015 to 2016, he was a Visiting Scholar with the Berkeley Sensor and Actuator Center, University of California at Berkeley, Berkeley, CA, USA. He is currently a Professor with the Smart Microsensors and Microsystems Engineering Research Center, Ministry of Education, College of Electronics and Information, Hangzhou Dianzi University, Hangzhou. His current research interests include industrial Internet of Things, sensor peripheral processing IC design, micro-electro-mechanical system (MEMS) sensors, and resonators, including design, modeling, and fabrication of microstructures.