iScience

CellPress
OPEN ACCESS

Review

Flexible thin film thermocouples: From structure, material, fabrication to application

Zhongkai Zhang,[1] Zhaojun Liu,[1,2,*] Jiaming Lei,[1] Luntao Chen,[1] Le Li,[1] Na Zhao,[1] Xudong Fang,[1,3] Yong Ruan,[4] Bian Tian,[1,3,5] and Libo Zhao[1,3,5]

SUMMARY

Flexible thin-film thermocouples (TFTCs) have been garnering interest as temperature sensors due to the advantages of being flexible, ultrathin, and ultralight. Additionally, they have fast response times and enable detection of temperature. These properties have made them suitable for applications such as wearable electronics, healthcare, portable personal devices, and smart detection systems. This review presents the progress in the development of flexible TFTCs. The mechanism, structural design, materials, fabrication methods, and related applications of flexible TFTCs are also elaborated. Finally, future development directions of flexible TFTCs are discussed such as wide-range temperature measurement, multiple sensor integration, and achieving reliable cold-end compensation systems.

INTRODUCTION

Over the past few decades, flexible sensors have gained popularity due to their versatile sensing ability across several fields such as optics, biology, chemistry, and mechanical engineering, among others.[1,2] Flexible sensors have many excellent performance characteristics, such as fast response times, high sensitivity, low cost, and can be manufactured with ease.[3–7] In addition, they exhibit high stability that enables them to be operated under various harsh environmental conditions.[8–12] The flexibility and wearability aspects of flexible sensors make them suitable for a wide range of monitoring applications in biomedicine, sports, engineering, and military.[13–24] In the medical field, flexible sensors are used for sensing body temperature, electrocardiography, and electromyography monitoring.[25–29] In the sports field, flexible sensors are used for monitoring human movements and physiological signals, providing data for sports training, and injury prevention. In engineering, flexible sensors are used for monitoring machinery and equipment performance, with the goal of improving efficiency and reducing downtime. Flexible sensors also have shown potential in the fields of wearable technology and Internet of Things (IoT).[30–33] For example, flexible sensors can be integrated into wearable devices such as smart watches and fitness trackers to monitor the health and physical activity of the wearer in real time. In the IoT field, flexible sensors can be used for remotely monitoring and controlling various devices and systems, which facilitates convenient and efficient management of devices.

Accurate measurement of temperature is necessary in many physical and chemical processes and plays an important role in many important practical applications such as industrial production, medical diagnosis, and weather forecasting, among others.[34–38] Thermocouples are temperature sensing devices that consist of two wires made of different metal alloys connected together at one end. These metal alloys have different electrical properties that vary with temperature, and the voltage they produce is proportional to the difference in temperature between the two wires. The junction temperature can then be determined by measuring this voltage. Thermocouples are widely used in temperature sensing due to their design simplicity, robustness, and their ability to measure a wide range of temperatures.[30,33,39–49] Due to these characteristics, they are used in several industrial, scientific, and consumer applications such as process control, ovens, furnaces, and automotive engines.[39,43,45,50–55] Based on the type of alloys used, several varieties of thermocouples are available for commercial use, such as Type K (nickel-chromium alloy and nickel-silicon alloy), Type J (iron and copper-nickel alloy), Type T (copper and constantan), Type B (platinum-rhodium alloy and platinum-rhodium alloy), Type S (platinum-rhodium alloy and platinum, Type R (platinum-rhodium alloy and platinum) and Type E (nickel-chromium alloy and constantan). Due to the distinct material characteristics of these thermocouple varieties, each of them has a different operation temperature range. In addition to traditional thermocouples, the rapid development of microelectromechanical system (MEMS) technology has enabled

[1]State Key Laboratory for Manufacturing Systems Engineering, School of Mechanical Engineering, Xi'an Jiaotong University, Xi'an 710049, China

[2]Department of Electrical & Computer Engineering, National University of Singapore, 4 Engineering Drive 3, Singapore 117576, Singapore

[3]State Key Laboratory for Manufacturing Systems Engineering, International Joint Laboratory for Micro/Nano Manufacturing and Measurement Technologies, Xi'an Jiaotong University, Xi'an Jiaotong University (Yantai) Research Institute for Intelligent Sensing Technology and System, Xi'an Jiaotong University, Xi'an 710049, China

[4]State Key Laboratory of Precision Measurement Technology and Instruments, Department of Precision Instrument, Tsinghua University, Beijing 100084, China

[5]Shandong Laboratory of Yantai Advanced Materials and Green Manufacturing, Yantai 265503, China

*Correspondence: lzj2018@stu.xjtu.edu.cn

https://doi.org/10.1016/j.isci.2023.107303

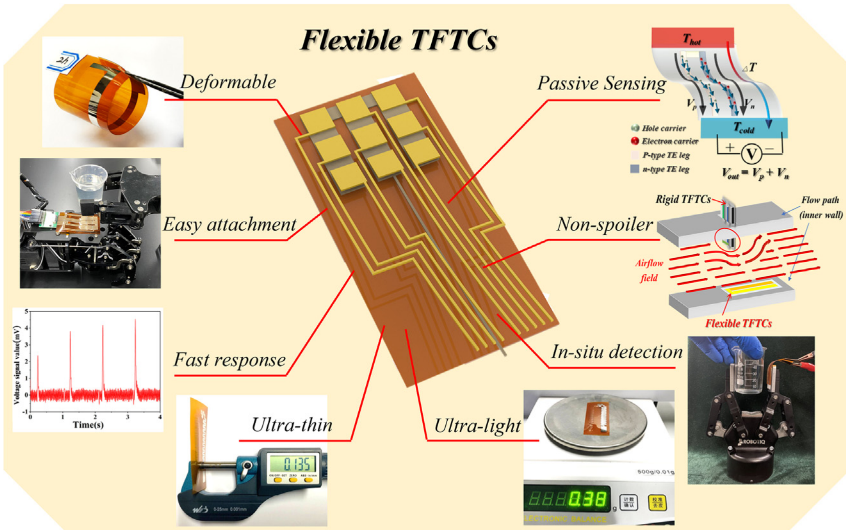

Figure 1. Advantages of flexible TFTCs: Deformable; Easy attachment; Fast response; Ultra-thin; Ultra-light; *in situ* detection; Non-spoiler (low interference r in flow filed); Passive sensing

thin-film thermocouples (TFTCs) that possess the advantages of small heat capacity and fast response.[56–61] As shown in Figure 1, flexible TFTCs not only inherit the advantages of traditional TFTCs, but also exhibit the characteristics of flexible sensors.[16,22,62] TFTCs are deformable, ultralight, ultrathin, portable, and can be easily attached, making them suitable for application in wearables. Additionally, their fast response times, low interference r in flow filed, *in situ* detection, and passive sensing ability confer them with unique advantages over other temperature sensors.[40,44,48,63–73]

TFTCs are feasible due to the advancements in the flexible substrate technology. Flexible substrates have enabled several applications such as flexible wearable electronics, wearable medical technology, portable personal electronics, and intelligent detection systems. TFTCs deposited on flexible substrates offer strong resistance to bending and durability, which makes them ideal for real-time temperature monitoring of the surfaces of non-planar object, especially surfaces with complex geometries and high curvature. Figure 2 presents a compendium of existing flexible TFTCs in terms of structure, materials, preparation process, performance, optimizations and applications. In this review, the theoretical principles of flexible TFTCs are first outlined in the section principle. In the sections structure and materials and fabrication, the flexible TFTCs are introduced sequentially in terms of structure, material, and preparation process. In the section applications, a summary of the application of TFTCs is presented. Finally, some challenges and future trends are discussed.

PRINCIPLE

The working principle of thermocouples is based on the Seebeck effect, which deals with electromagnetic effects in the circuit of metal conductors when two different metals are connected. A difference in temperature between the two contacts can form a thermal potential in the closed circuit, which can be measured to estimate the temperature. The Seebeck effect describes the process of converting thermal energy to electrical energy in scenarios where contact of different metals produces dissimilar thermoelectric potentials. Generally, a thermocouple is a closed loop formed by two conductors of different materials. When a temperature gradient exists between the hot and cold ends, the charges carried within the conductor or semiconductor move in a directional manner, eventually forming an electric potential at both ends of the thermocouple. Figure 3 shows the working schematic of a flexible TFTC. When using thermocouples to measure temperature, the hot end of the thermocouple is generally connected to the object to be measured, whereas the cold end is disconnected and connected to a multimeter or voltage acquisition system. By measuring the potential difference between the cold ends of the thermocouple poles, the temperature of the hot end is inferred. Then, the temperature of the object in contact with the hot end of the thermocouple is measured.[34,35,37,38,74–78]

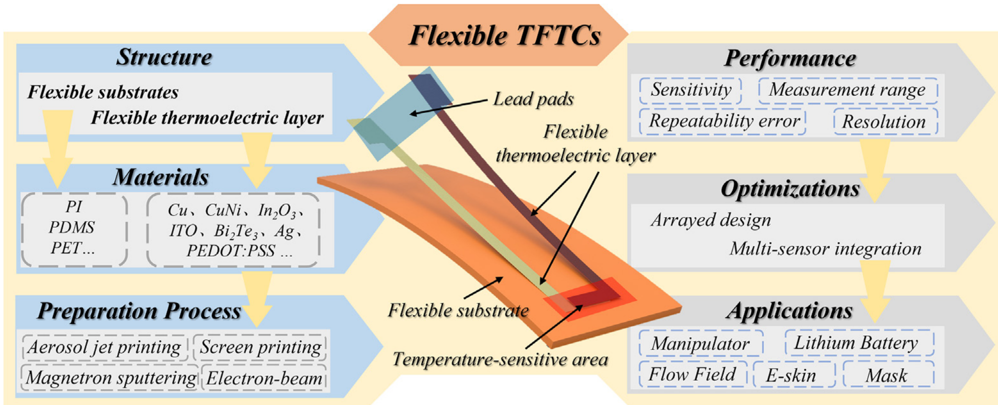

Figure 2. Introduction to the Structure, Materials, Preparation process, Performance, Optimizations and Applications of flexible TFTCs

From the Peltier effect, it is known that different materials possess different electron densities. Hence, at the two ends of the flexible TFTCs, the electrons will move from the higher density end toward the lower density end. The conductor that loses electrons will become positively charged, whereas the conductor that gains electrons will be negatively charged. The potential difference between the two conductors is called the contact electromotive force (EMF). In a thermocouple circuit, if the temperatures of the two conductors A and B at the two contacts are T and T_0, the respective contact EMFs are:

$$E_{AB}(T) = \frac{kT}{e} ln\frac{N_A}{N_B} \quad \text{(Equation 1)}$$

$$E_{AB}(T_0) = \frac{kT_0}{e} ln\frac{N_A}{N_B} \quad \text{(Equation 2)}$$

The total contact EMF in the circuit is:

$$E'_{AB}(T,T_0) = E_{AB}(T) - E_{AB}(T_0) = \frac{k}{e}(T - T_0) ln\frac{N_A}{N_B} \quad \text{(Equation 3)}$$

where k is the Boltzmann constant, e is electron charge, N_A and N_B are free electron densities of conductors A and B, respectively. From Equation 3, it can be deduced that the contact EMF in the thermocouple circuit is related to the nature of the thermoelectric material and the temperature at the contact end. If the two thermoelectrode materials in the thermocouple are identical, then the contact EMF in the loop is zero. When there is a temperature gradient at both ends of the same conductor, free electrons will diffuse from the higher temperature end to the lower temperature end. The high temperature end will lose electrons and become positively charged, while the low temperature end will gain electrons and become negatively charged. As a result of this diffusion, a potential difference is generated between the two ends of the same conductor. This potential difference varies as a function of temperature. If the two ends of the same conductor have different temperatures of T and T_0, then the potential generated at the different temperature ends of conductor A and conductor B is given by:

$$E_A(T,T_0) = \int_{T_0}^{T} \sigma_A dT \quad \text{(Equation 4)}$$

$$E_B(T,T_0) = \int_{T_0}^{T} \sigma_B dT \quad \text{(Equation 5)}$$

From Equations 4 and 5, the total EMF due to the temperature difference in the thermocouple circuit is shown in Equation 6:

$$E''_{AB}(T,T_0) = \int_{T_0}^{T} \sigma_A dT - \int_{T_0}^{T} \sigma_B dT = \int_{T_0}^{T_0} (\sigma_A - \sigma_B) dT \quad \text{(Equation 6)}$$

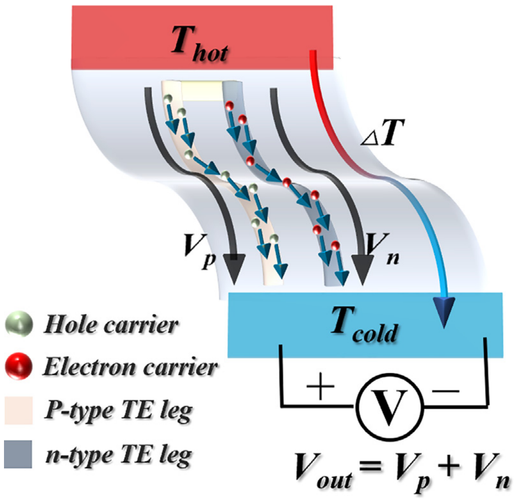

Figure 3. Principle of temperature measurement in flexible TFTCs

where σ_A and σ_B are the Thomson coefficients of conductors A and B, respectively; T is temperature of the hot end of the thermocouple; T_0 is temperature of the cold end of the thermocouple. Equation 6 shows that the EMF caused due to the temperature difference in a thermocouple loop is only related to the material itself and the temperature at both ends. If there is no temperature gradient at the ends of the conductor, the EMF in the thermocouple loop is zero. For a thermocouple consisting of two conductors A and B that are dissimilar, the thermal potential in the entire thermocouple circuit is:

$$E_{AB}(T, T_0) = \frac{k}{e}(T - T_0) ln \frac{N_A}{N_B} + \int_{T_0}^{T} (\sigma_A - \sigma_B) dT \qquad \text{(Equation 7)}$$

It can be inferred from Equation 7 that generated potential is only dependent on the electrode material and the temperature at the contact end. Besides these two, all others factors do not influence the potential. The EMF is zero if the electrodes are identical or if the temperature of the two contacts is equal. The thermal potential in the whole thermocouple circuit is given by:

$$E_{AB}(T, T_0) = \frac{k}{e}(T - T_0) ln \frac{N_A}{N_B} + \int_{T_0}^{T} (\sigma_A - \sigma_B) dT = \int_{T_0}^{T} S_{AB} dT \qquad \text{(Equation 8)}$$

where S_{AB} is the Seebeck coefficient of thermocouple. If the thermocouple electrode material is determined, the thermoelectric voltage of the thermocouple is only related to the temperature difference at the contact end of the thermocouple. The thermal potential in the thermocouple circuit is given by:

$$E_{AB}(T, T_0) = S_{AB}(T - T_0) \qquad \text{(Equation 9)}$$

If the T_0 in Equation 9 is a constant, then the thermocouple thermal potential is a function of the temperature of the hot end. Since the temperature of the hot end of the thermocouple is converted to a voltage, measuring the output voltage of the thermocouple and combining it with the temperature of the cold end yields the temperature of the hot end. If the hot end of the thermocouple and the object to be measured are in close contact, the temperature of the object can be indirectly measured. From this analysis, it can be concluded that the design of the thermocouple requires the composition of the two electrode materials to be different, and the existence of a temperature gradient between the two contact ends.

Homogeneous conductor law

When the two electrodes of a thermocouple are composed of the same homogeneous material, irrespective of the differences in length, cross-sectional shape, and temperature distribution of the conductor, no thermal EMF is generated in the circuit. This is because the free electron density and the Fermi level is the same for identical homogeneous conductors. Hence, the contact EMF of the closed circuit is zero. Since the temperature difference potentials cancel each other, the total EMF in the entire circuit is zero. If the thermocouple is composed of two different homogeneous conductors, the output potential is only related to

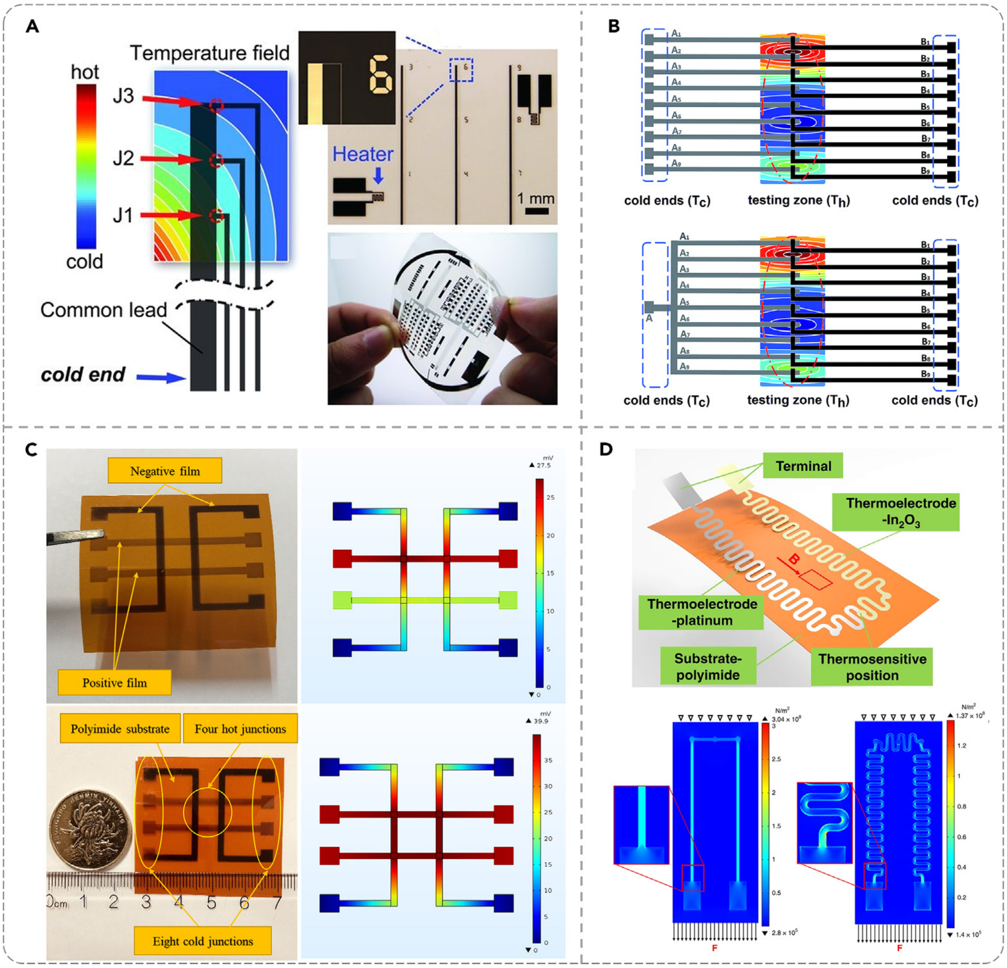

Figure 4. Design of different types of thermoelectric layer structures of flexible TFTCs

(A) Sensor made of single layer metallic film[67] Copyright 2012, John Wiley and Sons, Reproduced with permission).

(B) Common contact pad shared by all leads[90] Copyright 2017, Royal Society of Chemistry, Reproduced with permission).

(C) Four-point conjugate structure[45] Copyright 2020, AIP Publishing, Reproduced with permission).

(D) Micro-three-dimensional flexible TFTCs[65] Copyright 2021, Springer Nature, Reproduced with permission).

the temperature difference between the two contacts. It should be emphasized that the electrode material of the thermocouple must be homogeneous, else the temperature ladder of non-homogeneous materials will generate additional thermal potential, which will lead to the measurement errors.

Intermediate conductor law

If another material conductor is introduced into the thermocouple loop, the introduction of the intermediate conductor does not affect the thermal potential of the entire loop as long as the temperature of the two access ends of the third material conductor is the same. In practice, it is often necessary to introduce another conductor to access the voltage signal.[39–41,43–49,65,73,79–89]

STRUCTURE

Flexible TFTCs usually consist of a thin film substrate with two different thermoelectric materials. A thin film substrate comprises a flexible material, such as polyimide film with polydimethylsiloxane. Its main function is to provide a flexible, thin, and lightweight support structure that allows the thermocouples to adapt to different curved surfaces. Thermoelectric films used in thermocouples are generally composed of two different metals or alloys. These materials are physically or chemically deposited on a thin film substrate. The detection of temperature is achieved according to the thermoelectric effect. In addition, some flexible

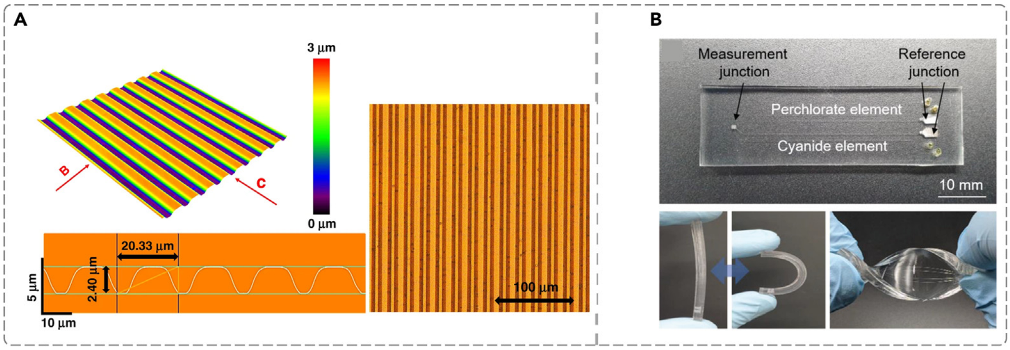

Figure 5. Design of different types of substrates of flexible TFTCs

(A) Wave-shaped polyimide surface[65] Copyright 2021, Springer Nature, Reproduced with permission).

(B) Poured PDMS with channels[91] Copyright 2022, John Wiley and Sons, Reproduced with permission).

TFTCs structures also introduce buffer layers and protection layers to complete their performance. The role of the buffer layer is to reduce the internal stress of the thermoelectric film during operation, whereas the role of the protective layer is to protect the thermoelectric film from damage.

Flexible thermoelectric layer

Liu et al.[67] reported a monolithic TFTC composed of single layer of metal thin film, as shown in Figure 4A. For the most part, the structure of this new TFTC is similar to a conventional thermocouple composed of two metals. However, the two thermoelectrodes of the TFTCs have different widths and consist of the same nickel film (similar in the situation of tungsten, or platinum film). Han et al.[90] used an array structure of tree pairs, as shown in Figure 4B. The proposed structure employed a strategy wherein all the leads of one material in the two thermoelectric electrodes of the TFTCs shared a common contact pad. Using this strategy, an array of N TFTCs used only N+1 contact pads instead of 2N pads that would be required in a conventional configuration, thus simplifying the device design. The two-dimensional (2D) maps of the local temperature distribution obtained by TFTCs in the test were found to be similar to those measured by conventional arrays. Tian et al.[45] designed a four-point conjugate TFTC structure with four test points, where the reliability of the measurement results was improved by simultaneous measurements of the four nodes in a small area, as shown in Figure 4C. Simulation results showed that the thermoelectric potential obtained by the heating of four junctions is greatly improved over the single junction design, which was ascribed to the cross-regulation between the four junctions. The sensitivity of the TFTC using this approach was enhanced from 137.3 $\mu V/^{\circ}C$ to 190.6 $\mu V/^{\circ}C$, thereby exhibiting a significant improvement. In order to enhance stability and decrease the deformation-induced internal stress, Liu et al.[65] designed a curved electrode patterns in the thermoelectric layer of the flexible TFTCs, as shown in Figure 4D. Simulation results showed that adjusting the structure of the thermocouple had no effect on the thermoelectric characteristics of the sensor. However, the resistance of the flexible TFTCs to deformation and damage was dramatically enhanced by employing a curved electrode instead of a straight electrode. Collectively, the structure of flexible TFTCs is rapidly developing toward arraying, flexibility, and simplicity. The shape of the thermoelectric electrode has been improved by reducing the number of leads, which decreases the propensity of the film to be damaged by stress while maintaining the sensing function. In addition, multiple measurement sensors can be arranged in each area to realize the temperature field detection in the part surface.

Flexible substrates

Liu et al.[65] modified the surface of flexible amine substrates, as shown in Figure 5A. Using several MEMS processes such as photolithography, plasma sputtering, and peeling, wave-shaped shapes were prepared on the surface of polyimide substrates, which aimed to increase the deformation and tensile resistance of flexible TFTCs. Jeon et al.[91] utilized cast Polydimethylsiloxane (PDMS) as a flexible substrate for TFTCs, as shown in Figure 5B. The top layer of the PDMS microchannel and the bottom layer of the electrode were precisely aligned under an optical microscopy and assembled by dehydration condensation of the hydroxyl groups formed on the surface of each layer. These TFTCs demonstrated good resistance to deformation, with no significant degradation in temperature measurements despite being subject to external stretching forces.

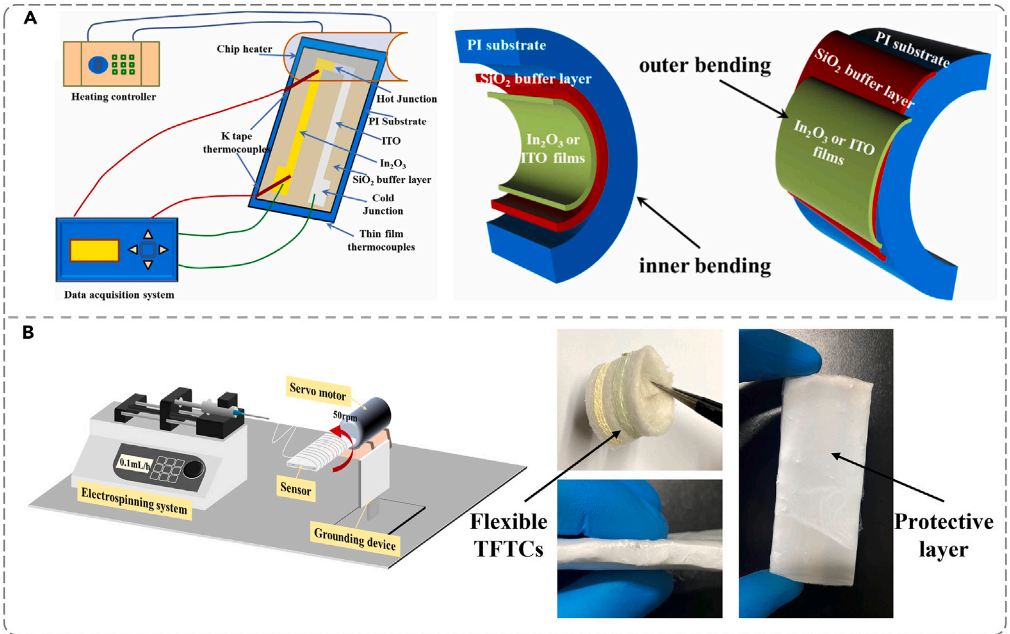

Figure 6. The buffer and protective layer

(A and B) Design of (A) buffer[69] Copyright 2022, Elsevier, Reproduced with permission) and (B) protective[47] Copyright 2023, Elsevier, Reproduced with permission) layer of flexible TFTCs.

Buffer and protective Player

Shi et al.[72] prepared a SiO_2 buffer layer film on a flexible substrate as well as in a thin thermoelectric film layer by magnetron sputtering deposition, as shown in Figure 6A. The SiO_2 layer improved the long-term performance of the thin-film thermocouple during heating and cooling and could decrease the cracks that appeared during the process. Due to the presence of the buffer layer, thermal and mechanical stresses were efficiently suppressed. Besides, in Figure 6A, Liu et al.[65] proposed the concept of adding a protective layer to flexible TFTCs. Since the flexible TFTCs thermoelectric sensitive layer is subject to sensor failure due to volatilization and wear in high temperature environment, they combined with electrostatic spinning process to cover the spun fiber on the surface of flexible TFTCs to form a continuous wrapped protective layer. The well coverage greatly reduces the possibility of damage to the thermoelectric thin film layer.

MATERIALS AND FABRICATION

The sensing material of a flexible thin film thermocouple generally consists of two different metallic or semiconductor materials that can produce the thermoelectric effect. Common materials include copper (Cu), constantan (Cu-Ni), platinum (Pt), palladium (Pd), gallium (Ga), chromium (Cr), silver (Ag), indium oxide (In_2O_3), and indium tin oxide (ITO), among others. In recent years, organic thermoelectric materials such as poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT: PSS), multi-walled carbon nanotubes (MWCNTs), and single-walled carbon nanotubes (SWCNTs) have emerged as sensing materials. Although organic materials offer better flexibility inorganic materials, their sensitivity and operating temperature range are lower than inorganic materials. High thermoelectric coefficient, high electrical conductivity and low thermal conductivity are the characteristics required for a good thermoelectric film. New materials such as nanomaterials, multilayer composites and organic-inorganic hybrid materials are being developed with the aim of improving the sensitivity and stability of flexible TFTCs. From the Table 1, it can be seen that the material systems with Cu/CuNi, In_2O_3/ITO combination are the mainstream direction of the existing flexible TFTCs. Among them, the flexible thermocouple in the Cu/CuNi system benefits from a more consistent preparation, a simple preparation process and a suitable compensation wire. The advantage of the flexible thermocouple in the In_2O_3/ITO system is that it has a large test sensitivity and can capture small temperature changes as well as rapid temperature changes. Polymers are commonly used as flexible substrate materials, including polyimide, PDMS, and PET, among others. However, as the application scenarios change, the demand for alternate materials has driven the development of flexible materials such as aerogel mats, nanofiber mats and mica.[46,47,49] These high

Table 1. Summary of the development of flexible TFTCs in recent years

Substrate	Thermoelectric electrode	Sensitivity (μV/°C)	Temperature range (maximum temperature difference/°C)	Preparation technology	*Reference*
PI	Cu/CuNi	20.6	25∼70	Aerosol jet printing	Sheng et al.[69]
PI	Cu/Ni	8.9	30∼110	Magnetron sputtering technology	Martiny et al.[70]
PI	Cu/CuNi	≈42	27∼32.23	Magnetron sputtering technology	Miao et al.[62]
PI	Cu/CuNi	30.6	Δ120	Aerosol jet printing	Renn et al.[92]
PI	Cu/CuNi	–	45–90	Magnetron sputtering technology & Electron-beam	Tang et al.[93]
PI	Cu/CuNi	–	110∼170	Magnetron sputtering technology	Ali et al.[94]
PI	Cu/CuNi	22.3	20∼80	Evaporation	Konishi and Hirata[95]
PI	Cu/CuNi	40	25∼232	Aerosol jet printing	Rahman et al.[96]
Silicon wafer	Ni/Cr	515	20∼80	Electron-beam	Assumpcao et al.[97]
PI	$Bi_{0.5}Sb_{1.5}Te_3$/PbTe	≈295	23∼323	Flash evaporation method	Kostyuk et al.[71]
PI	BiTe/Sb-Te	192.7	Δ135	Co-evaporation	Lee et al.[98]
PET	Bi_2Te_3/Cu	88	Δ40	Magnetron sputtering technology	Seo et al.[99]
PDMS	Bismuth/Gallium-based mixed alloys	10.54	25∼83	Liquid Metal Injection	Wang et al.[22]
PI	In_2O_3/Pt	204.35	25∼177.8	Magnetron sputtering technology	Liu et al.[44]
PI	In_2O_3/Pt	83	25∼159	Magnetron sputtering technology	Liu et al.[50]
PI	In_2O_3/ITO	171.7	Δ90	Magnetron sputtering technology	Shi et al.[72]
PI	In_2O_3/ITO	25.8	Δ200	Magnetron sputtering technology	Tian et al.[45]
Alumina-silicon oxide	In_2O_3/ITO	226.7	Liquid nitrogen temperature∼1200	Screen printing technology	Liu et al.[46]
PI	In_2O_3/ITO	175.8	25∼223.6	Screen printing process	Liu et al.[73]
Electrostatic spinning-A_2lSiO_5	In_2O_3/ITO	5.625	25∼345	Magnetron sputtering technology Vacuum vapor deposition Ink jet printing technology	Liu et al.[47]
PI	In_2O_3/ITO	35.96	30∼120	Magnetron sputtering technology	Shi et al.[100]
PI	In_2O_3/ITO	50.55	30∼180	Magnetron sputtering technology	Liu et al.[48]
PI	$Ti_3C_2T_x$-Mxene/graphene	53.6	Δ200	Aerosol jet printing	Saeidi-Javash et al.[66]
PET	Different widths Ni thin film	26.2 ± 1.5	Δ40	Magnetron sputtering technology	Liu et al.[67]
PI	Au/Ni	≈36	Δ80	Screen printing	Lee et al.[68]
PDMS	$(K_{3/4}Fe(CN)_6)/(Fe(ClO_4)_{2/3})$,	3430	Δ70	Syringe pump to inject	Jeon et al.[91]

(*Continued on next page*)

Table 1. ***Continued***

Substrate	Thermoelectric electrode	Sensitivity (μV/°C)	Temperature range (maximum temperature difference/°C)	Preparation technology	*Reference*
PI	Alumel (Ni/Al/Si/Mn)/Chromel (Ni/Cr)	41.2	Δ90	Magnetron sputtering technology	Mutyala et al.[101]
Parylene film	Cascaded Ni thin film	55.69 (64-cascaded)	Δ35	Magnetron sputtering technology	Li et al.[79]
Paper	Ga/GaIn$_{21.5}$	0.11	0∼200	Direct printing	Li et al.[102]
PET	Pd/Cr	–	Δ23	Magnetron sputtering technology	Han et al.[90]
PI	V_2O_5/Al	680	23.5–33.8	Resistive thermal evaporation	Bianchi et al.[103]
PET-foil	Carbon black/Silver	5.6	25∼150	Screen printing process	Knoll et al.[64]
PDMS	SWCNTs/AgNW MWCNTs/AgNWs SWCNTs/MWCNTs	37 23 11	Δ90	Ink printing process	Xin et al.[63]
PET	MWCNTs-PEDOT: PSS/Cu	220	0∼100	Solution casting Magnetron sputtering technology	Chen et al.[104]
PET	Al/PEDOT:PSS	32.8 ± 2.7	0∼100	Inkjet-printed	Zhang et al.[105]
PET	MWCNT/Ag	≈6.7	30∼150	Inkjet printing	Jung et al.[106]
PDMS	CNC-PSSH/PEDOT:PSS-WPU	13	Δ35	Drop-casting and drying processes	Xu et al.[107]
Paper	PEDOT:PSS/AgNPs	18.8	Δ125	Inkjet printing technology.	Jung et al.[106]
	PVDF-HFP & NaTFSI & PC (PhNP)	20000	Δ10	Cast	Chi et al.[108]
TEMPO-TOCN	PEDOT:PSS/AgNP	11	25∼150	Inkjet printing technology	Jung et al.[23]
–	MWCNT/Pt	41.0	260K–420K	Electron-beam-induced Pt	Miao et al.[109]
PI	AgNWs/CNT CNTs/PEDOT AgNWs/PEDOT	50.4 ± 5.68 26.9 ± 2.87 14.8 ± 0.83	Δ80	inkjet printing	Albrecht et al.[110]

PVDF-HFP, Polyvinylidene fluoride-hexafluoropropylene; NaTFSI, Sodium bis(trifluoromethylsulfonyl)imide; PC, Propylene carbonate; PET, Polyethylene terephthalate; PI, Polyimide; PDMS, Polydimethylsiloxane; CNCs, Carbon nanocoils; PSSH, PSSHPoly(4-styrenesulfonic acid); WPU, Waterborne polyurethane; AgNW, Ag nanowire.

temperature resistant flexible materials are initially commonly used as insulation materials. However, by combining them with a thin film preparation process, they can become good substrates for high-temperature resistant flexible sensors. The fatal defect of high-temperature carbonization of organic materials is solved by replacing conventional organic materials. Typically, the fabrication of thermoelectric layers has been achieved by additive manufacturing methods. However, the rapid development of MEMS technology has enabled alternate methods for the preparation of thin films. Common manufacturing methods include magnetron sputtering technology, chemical vapor deposition, sol-gel method, screen printing method, and 3d printing method, among others. Depending on the material system, a suitable responsive fabrication process can be adopted to achieve the best service performance of thermoelectric thin films. New manufacturing technologies and processes, such as nano-manufacturing and printing technologies, have shown potential for improving the manufacturing efficiency and quality of flexible TFTCs.

Rahman et al.[96] used rapid aerosol spray printing of nanoparticles combined with low-power laser sintering under inert gas protection to prepare flexible TFTCs of Cu-CuNi on Kapton substrates, as shown in Figure 7A. The laser power was maintained under 400 mW due to the mechanical properties of Kapton. The performance of the prepared TFTCs was repeatedly tested at temperatures up to 140°C. Furthermore, the integrity of the device under bending and twisting for 200 times with different radii was also tested and a stable performance was reported. Xu et al.[107] designed a flexible dual-output fluid sensor using patterned thermoelectric materials, as shown in Figure 7B. Bismuth telluride was deposited on a cut PET substrate using RF magnetron sputtering using a shadow mask method. To realize the array design, copper thermos-electrodes are fabricated under the same conditions using direct current magnetron sputtering technique. Additionally, an expandable array structure consisting of multiple TFTCs was manufactured and applied for measuring fluid temperature over a large area. The arrayed 4x4 (16 pixels) TFTCs were capable of precisely generating different voltage signals and measuring the temperature and dynamics of the working fluid with high accuracy. Jung et al.[23] developed flexible TFTCs using a paper platform with inkjet printing method, as shown in Figure 7C. They used PEDOT: PSS and silver nanoparticles on flexible paper substrates to form the thermoelectric materials of the thermocouples. The TFTCs exhibited a maximum measurement temperature of 150°C. Furthermore, the arrayed TFTCs were integrated with the arrayed pressure sensors to achieve temperature and pressure detection over a specific area, thereby demonstrating great potential for electronic skin applications. Additionally, encapsulating the sensors with PDMS imparted them with excellent bending performance, where the devices were found to maintain their initial properties after up to 1000+ bending cycles.

As shown in Figure 7D, Liu et al.[46] innovatively used an aerogel mat as a flexible substrate. To overcome the roughness of the surface of the flexible substrate, they used an organic polymer as a solvent to mix indium oxide and ITO thermoelectric powders. The two thermoelectric electrodes were separately deposited on the substrate surface using the screen-printing technique. The prepared TFTCs demonstrated excellent temperature measurement capability across a wide temperature range from −190°C to 1200°C. Additionally, the sensor can withstand 100 G of shock acceleration and 2000 Hz high-frequency vibration. These performance indicators validate the application of TFTCs in various extreme environments such as expeditions and evacuations. Liu et al.[44] used magnetron sputtering technique to deposit indium oxide and platinum films onto polyimide films, followed by a stepwise annealing process. The obtained TFTCs exhibited ultrahigh-test sensitivity along with a good performance in terms of repeatability, stability and temperature drift. The sensors were also tested in robotics applications, where accurate measurement of the temperature of the test object under was achieved, thereby demonstrating good prospects in industrial robotics and biomedical fields. Liu et al.[47] used electrospinning technique to prepare high temperature resistant alumina-silicon oxide nanofiber mats. Through parameter optimization, flexible substrates were obtained with excellent mechanical properties and resistance to temperatures up to 970°C. Indium oxide and ITO thermoelectric films were successfully prepared on their surfaces using magnetron sputtering, vacuum vapor deposition and inkjet printing techniques. Collectively, numerous thermoelectric materials have been derived from flexible TFTCs depending on the actual application requirements. In addition to traditional polymer materials, several other flexible substrate materials have also emerged. Along with thermoelectric materials, these substrate materials are compatible with versatile preparation processes, which have the potential to realize flexible TFTCs with higher sensitivity, wider testing range, and superior mechanical properties. Table 1 presents the results of relevant studies on flexible TFTCs over the years. In practice, flexible TFTCs are different from conventional commercial wire thermocouples, because the preparation process of flexible thermo-sensitive layer films is highly influenced by different process parameters and preparation methods. Even thermoelectric thin films prepared in a uniform environment can have different crystal structures depending on the location. Take the Cu/CuNi TFTCs as an

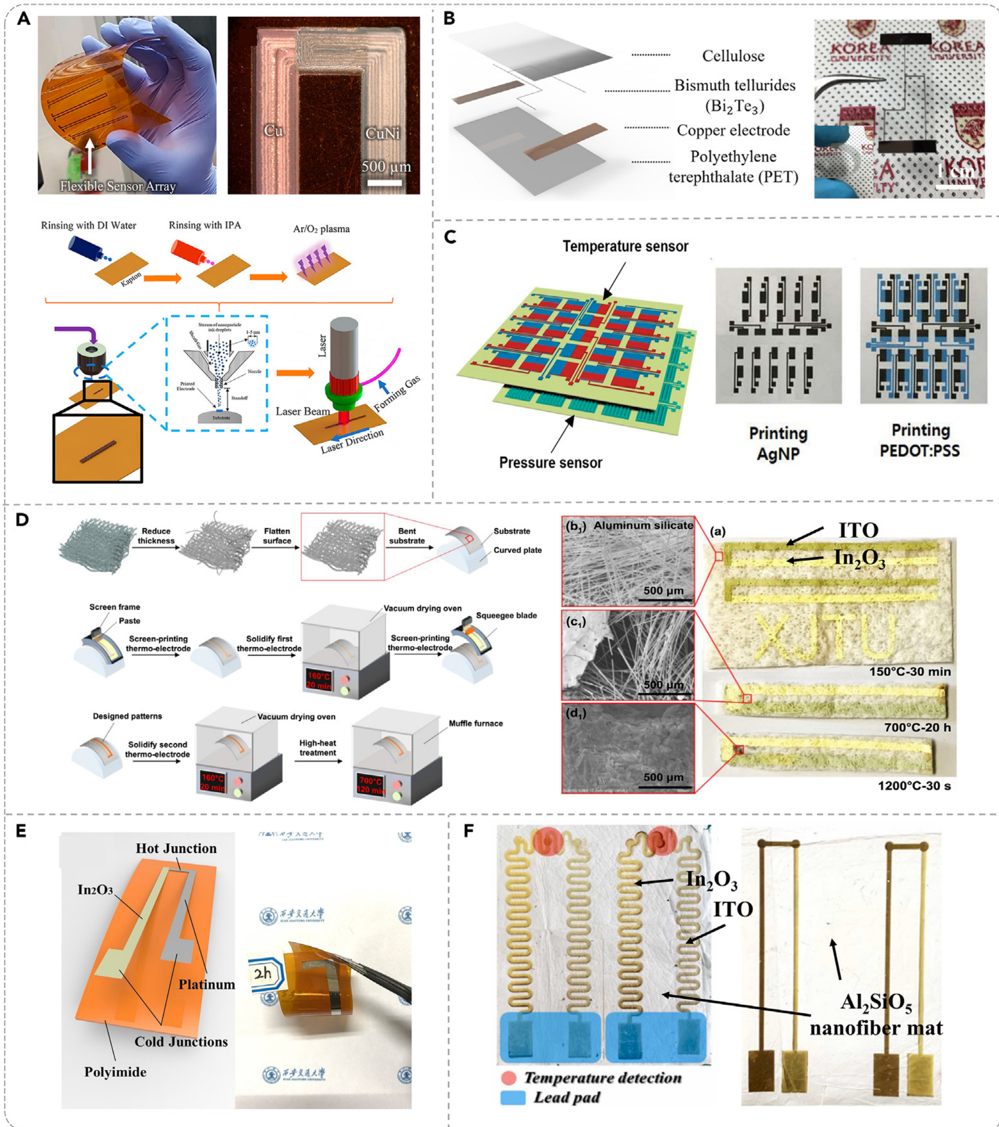

Figure 7. Flexible TFTCs composed of different materials prepared using different processes

(A) Copyright 2019, American Chemical Society, Reproduced with permission.[96]
(B) Copyright 2018, Elsevier, Reproduced with permission.[99]
(C) Copyright 2017, American Chemical Society, Reproduced with permission.[23]
(D) Copyright 2023, IOPscience, Reproduced with permission.[46]
(E) Copyright 2020, Elsevier. Reproduced with permission.[44]
(F) Copyright 2023, Elsevier, Reproduced with permission.[47]

example,[62,69,70,92–95,97] the difference of its Seebeck is that different preparation processes such as aerosol jet printing and magnetron sputtering are used to produce thin films with different microstructures. At the same time, heat treatment in different environments and temperatures can also lead to changes in the internal molecular structure and thermoelectric properties. From the available experience, the preparation of flexible TFTCs by screen-printing technology is of better consistency and has the potential to achieve commercial consistency with wire-type thermocouples.

APPLICATIONS

Before practical application, the prepared flexible TFTCs need to be calibrated and tested as a way to obtain a reference test table for the flexible TFTCs. A small heating table, heating pad, or muffle furnace will usually be used as the heat source to provide the heat load. A standard wire thermocouple is utilized as the standard temperature. The thermoelectric voltage of the flexible TFTCs is collected by a multimeter. The cold end compensation technique is also usually used to keep the cold end of the flexible TFTCs at 0°C using external water circulation or ice water. The calibration system is built to obtain the temperature-output voltage relationship curve of the flexible TFTCs. In addition, the hysteresis error, repeatability error, and temperature drift rate information of the flexible TFTCs will be tested and evaluated in order to better obtain the characteristic information of the flexible TFTCs. In addition to this, laser light is usually used as a thermal load source to irradiate the heat sensitive areas of flexible TFTCs to evaluate their test response speed. Typically, the response speed of TFTCs is in the millimeter range.[34,35,37–40,43–45,48,65,73,111–114]

Flexible TFTCs are widely used in many fields as they can be miniaturized and offer high measurement accuracy. In the biomedical field, flexible TFTCs can be used for the measurement of the temperature of body surface and skin. In addition, they can also be used to measure internal temperatures of organs, such as brain and liver to aid in disease diagnosis. In the field of industrial automation, flexible TFTCs can be used for controlling and monitoring the temperature of various equipment, such as industrial furnaces, boilers, and cooling towers, to improve production efficiency, reduce energy consumption and ensure production quality. In the energy field, flexible TFTCs can be used for solar panel surface temperature measurement to improve the efficiency of solar panels. In the field of thermal power generation, they can be used to measure and control the temperature of boilers to improve efficiency. TFTCs are also routinely used in scenarios where extreme temperatures are encountered, such as in aerospace applications involving the control of rockets and aircraft. Herein, flexible TFTCs can provide accurate temperature measurement and ensure the safety of aircraft. In the field of environmental monitoring, flexible TFTCs can be used for measuring the temperature of soil, seawater, atmosphere, etc. These data can be used for environmental protection, weather prediction, marine investigation, and other fields. In general, the flexible TFTCs have demonstrated efficacy in a wide range of applications, and they are expected to be utilized in more applications in the future.[12,79–89,115–117]

Liu et al.[46] used the properties of flexible sensors to affix flexible TFTCs to the inner wall of respiratory masks, where the respiratory rate and breathing depth were determined from the temperature response, as shown in Figure 8A. Further, the smart mask was able to distinguish different breathing states of a person, such as holding of breath, mouth breathing, nasal breathing, etc. In addition, Miao et al.[62] also applied the flexible TFTCs to nostrils in order to aid in the diagnosis of sleep apnea. The temporal breathing profile for normal breathing generally shows a curve with peaks and troughs, whose absence or irregularities could indicate the occurrence of sleep apnea. Due to TFTCs being ultrathin, ultra-lightweight, and highly sensitive with high response characteristics, they are ideal for detecting human respiratory status in real time and *in situ* for portable healthcare monitoring applications. Liu et al.[46] attached 3 × 3 arrays of flexible TFTCs to the back of a robotic dexterous hand, as shown in Figure 8B. All nine sensors demonstrate a good thermal response when a beaker with hot water was reciprocally placed on the back of the robotic hand. Also, each detection point exhibited independent sensing ability. In addition, an array module consisting of 20 TFTCs was fabricated by.[63] Application of 17 modules to the surface of a robotic hand realized the function of artificial skin that could monitor the surface temperature distribution of the hand. In the experiment, a temperature map was generated when a finger was brought into contact with the array module. The temperature distribution was consistent with the orientation of the finger touching the temperature sensor array. Placing an ice cube on the surface of the module showed a minimum temperature of 1.25°C for the ice, which is consistent with the temperature of an ice-water mixture. These experimental demonstrations validate that the flexible TFTCs can meet the needs of soft robots, robotic hands, and other artificial skin applications. Another potential application of TFTCs is in battery management systems that rely on cell surface temperature monitoring for assessing their aging and safety, as shown in Figure 8C. For instance, Martiny et al.[70] integrated flexible array TFTCs directly into a commercial battery package for *in situ* temperature monitoring of Li-ion pouch batteries. The battery was only able to operate normally for 15 cycles. Although the sensor had no effect on the cell, the adhesion of the sensor overlay weakened after the cycling tests and resulted in electrolyte leakage. However, the tests verified that the flexible TFTCs were capable of carrying out spatially resolved temperature monitoring within the lithium-ion pouch battery.[101] The embedded TFTCs sensors ensure that the entire sensing system is flexible, and damage-proof with good thermal conductivity. The sensors are easily assembled into soft pack lithium batteries, where their temperature can be reliably measured during the charging-discharging cycles. In the aerospace field, Liu et al.[46] placed flexible TFTCs sensors and standard K-type thermocouples at the tail of a model aircraft engine to monitor the tail

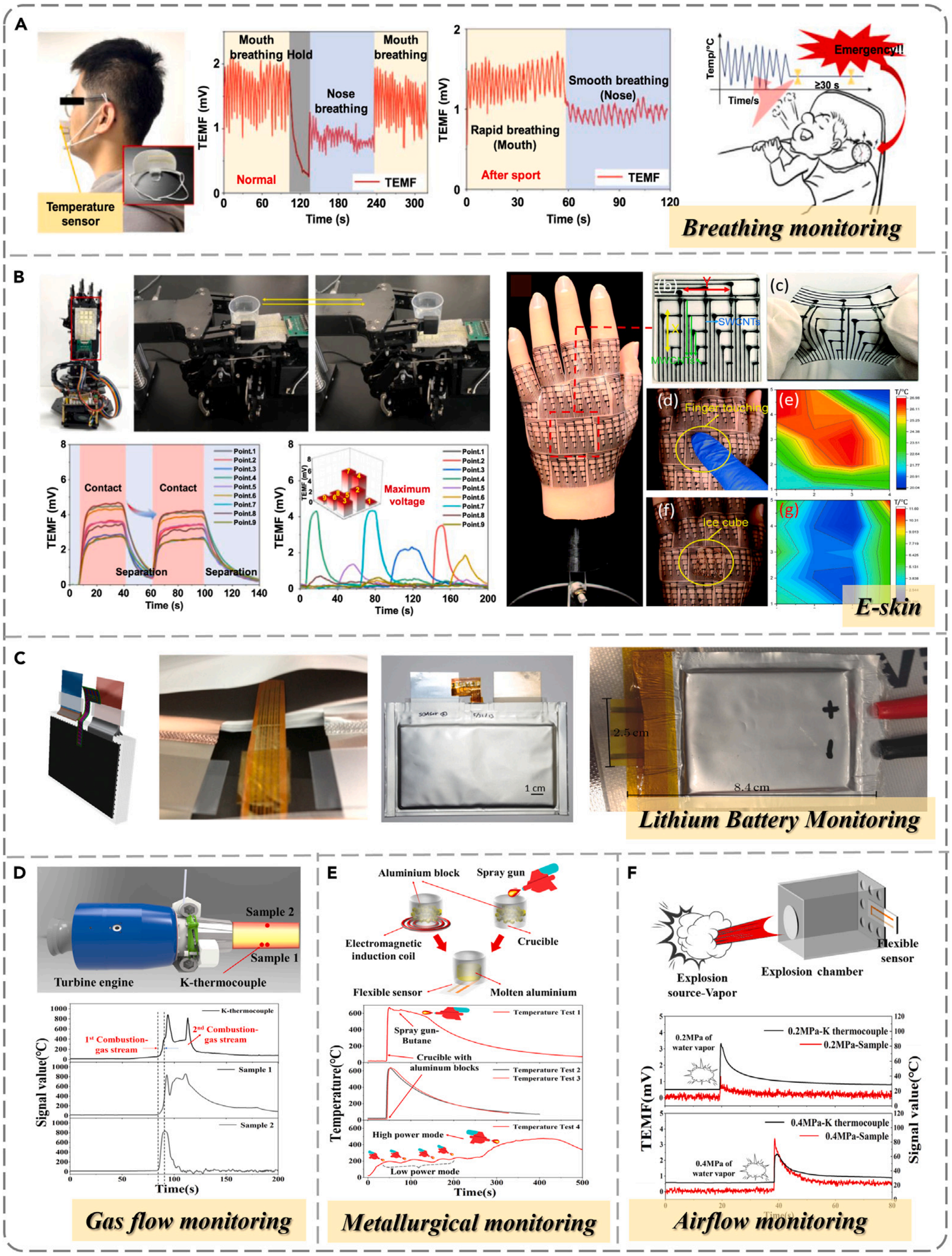

Figure 8. Different application scenarios for flexible TFTCs

(A) Breathing monitoring[46] Copyright 2023, IOPscience, Reproduced with permission).

(B) E-skin[63] Copyright 2019, Royal Society of Chemistry, Reproduced with permission;[46] Copyright 2023, IOPscience, Reproduced with permission).

(C) Lithium battery monitoring[70] Copyright 2022, American Chemical Society, Reproduced with permission;[101] Copyright 2014, Elsevier, Reproduced with permission).

(D) Gas flow monitoring[46] Copyright 2014, Elsevier, Reproduced with permission).

(E) Metallurgical monitoring[46] Copyright 2023, IOPscience, Reproduced with permission).

(F) Airflow monitoring[46] Copyright 2023, IOPscience, Reproduced with permission).

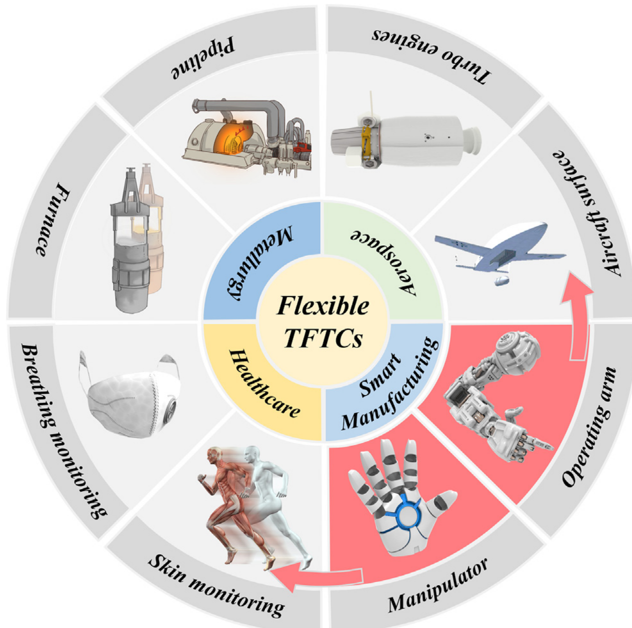

Figure 9. Flexible TFTCs with great application potential in the future

airflow temperature in real time, as shown in Figure 8D. The turbine engine model was tested twice consecutively. The test results showed that the flexible TFTCs responded faster than the standard K-type thermocouple due to the lower thermal conductivity of its flexible substrate and the small heat capacity of the thermally sensitive film layer. In the first test, the flexible TFTCs measured fluid temperatures up to 860.1°C, which was very close to the maximum temperature of 886.1°C measured by standard K-type thermocouples. Liu et al.[46] also applied the flexible TFTCs in controlling metallurgical processes, where temperature measurements play an important role, as shown in Figure 8E. In their experiments, they heated a 1 mm × 1 mm x 1 mm stack of aluminum blocks by electromagnetic induction and oxyacetylene flame. They placed a crucible containing aluminum solution in the temperature sensing part of the flexible TFTCs, which was heated using a lance and then cooled naturally. Real-time temperature measurements of the crucible were recorded by the flexible TFTCs during heating and cooling of aluminum blocks to evaluate the loading of aluminum. In comparison with the traditional thermocouples for point contact temperature detection, the prepared flexible temperature sensor can realize small surface contact at measurement points, which significantly increases the measurement accuracy of crucible wall temperature. Thus, flexible TFTCs show great promise in temperature measurements for metallurgical engineering and metal casting applications. As shown in Figure 8F, Liu et al.[46] applied the flexible TFTCs for measuring the temperature of flow fields by establishing a semi-closed physical blasting system. At the source of the blast, a closed vessel containing water was heated and the pressure inside the chamber was detected with a pressure gauge. The pressure of the water vapor was released by opening the lid of the container. In the chamber, flexible TFTCs were taped to the inner wall to withstand the water vapor impact. As a control, a standard K-type thermocouple was placed next to the flexible TFTCs for comparative testing. Results showed that the flexible TFTCs exhibited a higher response rate to the blast gas than the standard K-Thermocouple. More importantly, the flexible TFTCs could distinguish the difference in temperature between the two blasts at different pressures. The magnitude of the maximum signal detected by the TFTCs increased with the increase of pressure inside the blast chamber. The standard K-type thermocouples exhibited inconsistent signals in two tests, thus validating that the detection accuracy of the prepared flexible TFTCs is due to commercial thermocouples.

Further, Xu et al.[107] developed a sensing system to simultaneously detect fluid temperature and dynamics. Flexible TFTCs were used to generate peak voltages that reflect real-time changes in temperature and fluid dynamics. Additionally, the performance of the sensing system in terms of temperature and kinetics of the working fluid was verified using an arrayed design. The system was found to be suitable for a variety of geometries with sensing resolutions of 0.19K and 0.03 cm/s for fluid temperature and kinetics, respectively. In conclusion, the high response speed, and the small heat capacity of TFTCs enable their application in complex and variable flow fields.

Flexible TFTCs are emerging to be a promising class of sensors with many advantages such as high sensitivity, fast response, low cost, bendability, and wearability. In Figure 9, the following are the future trends of flexible TFTCs.

a) Higher sensitivity and accuracy: Due to advancements in manufacturing technologies and material science, it will be possible to develop flexible TFTCs with even higher sensitivity and accuracy, allowing them to be widely used in more demanding application scenarios.
b) Wider range of applications: Flexible TFTCs are already used in medical, environmental monitoring, energy, and electronic devices, and are envisaged to be used in smart home, automotive, and aerospace applications in the future.
c) Large-scale manufacturing and commercialization: The manufacturing technology of flexible TFTCs continues to advance and will gradually achieve mass production and commercialization. This will reduce costs and promote their popularity in the market.
d) Intelligence and networking: The application potential of flexible TFTCs will be further extended when they are integrated with wireless communication and data processing technology in the future for achieving remote monitoring and control functions.

Currently, flexible TFTCs have initially shown good applications in mechanical dexterous hands, robotic arms, and wearable devices. With the development of new thermoelectric and flexible materials, flexible TFTCs are able to expand to aerospace and metallurgy to achieve temperature field monitoring for more high-temperature components, transient flow fields, and steady-state flow fields. Hence, it can be concluded that flexible TFTCs have a broad development prospect in the future. With the continuous innovation and development of material science, manufacturing technology and application scenarios, flexible TFTCs can be expected to play an increasingly important role in ushering in convenience and innovation to the society at large.

Conclusion

In this review, an exhaustive collection of literature detailing the development of flexible TFTCs is presented. Based on recent research, flexible TFTCs sensors are classified based on temperature response mechanisms, materials, fabrication methods and applications. The selection of materials, the structural design of sensors, and the preparation methods of thermoelectric materials are essential to meet the needs of high-performance temperature sensors. Organic materials have also emerged as excellent thermoelectric materials, in addition to traditional thermoelectric materials. Interestingly, films of many inorganic materials are being designed to be flexible, which can replace polymeric materials due to their superior physical properties. Sensors are being designed to be more flexible with simpler structures and arrayed patterns. In addition to the traditional MEMS processes, printing methods are gaining more popularity, as they are inexpensive, convenient, and a good choice for producing large-area arrays of TFTCs. Currently, the highest sensitivity of flexible TFTCs can reach millivolts per degree Celsius, whereas the highest measured temperature has reached 1200°C. Currently, flexible TFTCs have achieved applications in various fields such as healthcare, smart manufacturing, and electronic skin. However, the existing flexible TFTCs still have certain limitations in terms of temperature sensitivity, test range, stability, and stretchability. Therefore, further research is necessary to develop TFTCs that can operate over wide temperature ranges while ensuring a high degree of flexibility.

ACKNOWLEDGMENTS

This work is supported by The National Key Research and Development Program of China (2020YFB2009100), Fundamental Research Funds for the Central Universities (No. xhj032021016-06). Z.L. received a China Scholarship Council fund (No. 202206280155) for his research stay at National University of Singapore.

AUTHOR CONTRIBUTIONS

Conceptualization: L.Z. and Z.Z.; Investigation: L.J., L.L., C.L., and Z.N.; Funding acquisition: T.B., L.Z., and Z.Z.; Project administration: T.B., Z.L., R.Y., and F.X.; Supervision: T.B., Z.L., R.Y., and F.X.; Writing – original draft: L.Z., C.L., Z.Z., and L.J.; Writing – review and editing: L.Z. and C.L.

DECLARATION OF INTERESTS

The authors declare no competing interests.

REFERENCES

1. Hammock, M.L., Chortos, A., Tee, B.C.K., Tok, J.B.H., and Bao, Z. (2013). 25th anniversary article: the evolution of electronic skin (e-skin): a brief history, design considerations, and recent progress. Adv. Mater 25, 5997–6038.

2. Gao, W., Emaminejad, S., Nyein, H.Y.Y., Challa, S., Chen, K., Peck, A., Fahad, H.M., Ota, H., Shiraki, H., and Kiriya, D. (2016). Fully integrated wearable sensor arrays for multiplexed in situ perspiration analysis. Nature 529, 509–514. https://escholarship.org/uc/item/5jn8d415.

3. Costa, J.C., Spina, F., Lugoda, P., Garcia-Garcia, L., Roggen, D., and Münzenrieder, N. (2019). Flexible sensors—from materials to applications. Technologies 7, 35.

4. Rim, Y.S., Bae, S.H., Chen, H., De Marco, N., and Yang, Y. (2016). Recent progress in materials and devices toward printable and flexible sensors. Adv. Mater. 28, 4415–4440.

5. Gao, W., Ota, H., Kiriya, D., Takei, K., and Javey, A. (2019). Flexible electronics toward wearable sensing. Acc. Chem. Res. 52, 523–533.

6. Sun, Y., and Rogers, J.A. (2007). Inorganic semiconductors for flexible electronics. Adv. Mater. 19, 1897–1916.

7. Wang, P., Hu, M., Wang, H., Chen, Z., Feng, Y., Wang, J., Ling, W., and Huang, Y. (2020). The evolution of flexible electronics: from nature, beyond nature, and to nature. Adv. Sci. 7, 2001116.

8. Yang, Y., and Gao, W. (2019). Wearable and flexible electronics for continuous molecular monitoring. Chem. Soc. Rev. 48, 1465–1491.

9. Lipomi, D.J., Vosgueritchian, M., Tee, B.C.K., Hellstrom, S.L., Lee, J.A., Fox, C.H., and Bao, Z. (2011). Skin-like pressure and strain sensors based on transparent elastic films of carbon nanotubes. Nat. Nanotechnol. 6, 788–792.

10. Yang, J.C., Mun, J., Kwon, S.Y., Park, S., Bao, Z., and Park, S. (2019). Electronic skin: recent progress and future prospects for skin-attachable devices for health monitoring, robotics, and prosthetics. Adv. Mater. 31, 1904765.

11. Wu, H., Huang, Y., Xu, F., Duan, Y., and Yin, Z. (2016). Energy harvesters for wearable and stretchable electronics: from flexibility to stretchability. Adv. Mater. 28, 9881–9919.

12. Wang, X., Liu, Z., and Zhang, T. (2017). Flexible sensing electronics for wearable/attachable health monitoring. Small 13, 1602790.

13. Li, W.D., Ke, K., Jia, J., Pu, J.H., Zhao, X., Bao, R.Y., Liu, Z.Y., Bai, L., Zhang, K., Yang, M.B., and Yang, W. (2022). Recent Advances in Multiresponsive Flexible Sensors towards E-skin: A Delicate Design for Versatile Sensing. Small 18, 2103734.

14. Chowdhury, S.A., Saha, M.C., Patterson, S., Robison, T., and Liu, Y. (2019). Highly conductive polydimethylsiloxane/carbon nanofiber composites for flexible sensor applications. Adv. Mater. Technol. 4, 1800398.

15. Li, J., Bao, R., Tao, J., Peng, Y., and Pan, C. (2018). Recent progress in flexible pressure sensor arrays: From design to applications. J. Mater. Chem. C 6, 11878–11892.

16. Wang, C., Xia, K., Wang, H., Liang, X., Yin, Z., and Zhang, Y. (2019a). Advanced carbon for flexible and wearable electronics. Adv. Mater. 31, 1801072.

17. Yuan, H., Lei, T., Qin, Y., and Yang, R. (2019). Flexible electronic skins based on piezoelectric nanogenerators and piezotronics. Nano Energy 59, 84–90.

18. Niu, Y., Liu, H., He, R., Li, Z., Ren, H., Gao, B., Guo, H., Genin, G.M., and Xu, F. (2020). The new generation of soft and wearable electronics for health monitoring in varying environment: from normal to extreme conditions. Mater. Today 41, 219–242.

19. Zhu, C., Chalmers, E., Chen, L., Wang, Y., Xu, B.B., Li, Y., and Liu, X. (2019). A Nature-Inspired, Flexible Substrate Strategy for Future Wearable Electronics. Small 15, 1902440.

20. Jung, Y.S., Jeong, D.H., Kang, S.B., Kim, F., Jeong, M.H., Lee, K.-S., Son, J.S., Baik, J.M., Kim, J.-S., and Choi, K.J. (2017a). Wearable solar thermoelectric generator driven by unprecedentedly high temperature difference. Nano Energy 40, 663–672.

21. Cao, Z., Koukharenko, E., Tudor, M., Torah, R., and Beeby, S. (2016). Flexible screen printed thermoelectric generator with enhanced processes and materials. Sens. Actuators, A 238, 196–206.

22. Wang, Q., Gao, M., Zhang, L., Deng, Z., and Gui, L. (2019b). A handy flexible micro-thermocouple using low-melting-point metal alloys. Sensors 19, 314.

23. Jung, M., Kim, K., Kim, B., Cheong, H., Shin, K., Kwon, O.-S., Park, J.-J., and Jeon, S. (2017b). Based bimodal sensor for electronic skin applications. ACS Appl. Mater. Interfaces 9, 26974–26982.

24. Jung, M., Kim, K., Kim, B., Lee, K.-J., Kang, J.-W., and Jeon, S. (2017c). Vertically stacked nanocellulose tactile sensor. Nanoscale 9, 17212–17219.

25. Singh, D., Kutbee, A.T., Ghoneim, M.T., Hussain, A.M., and Hussain, M.M. (2018). Strain-Induced Rolled Thin Films for Lightweight Tubular Thermoelectric Generators. Adv. Mater. Technol. 3, 1700192.

26. Guo, Z., Yu, Y., Zhu, W., Zhang, Q., Liu, Y., Zhou, J., Wang, Y., Xing, J., and Deng, Y. (2022). Kirigami-based stretchable, deformable, ultralight thin-film thermoelectric generator for BodyNET application. Adv. Energy Mater. 12, 2102993.

27. Sun, T., Peavey, J.L., Shelby, M.D., Ferguson, S., and O'Connor, B.T. (2015). Heat shrink formation of a corrugated thin film thermoelectric generator. Energy Convers. Manage. 103, 674–680.

28. Zhang, Z., Wang, B., Qiu, J., and Wang, S. (2019). Roll-to-roll printing of spatial wearable thermoelectrics. Manuf. Lett. 21, 28–34.

29. Sahoo, S., Walke, P., Nayak, S.K., Rout, C.S., and Late, D.J. (2021). Recent developments in self-powered smart chemical sensors for wearable electronics. Nano Res. 14, 3669–3689.

30. Liu, H., Zhong, J., Lee, C., Lee, S.-W., and Lin, L. (2018a). A comprehensive review on piezoelectric energy harvesting technology: Materials, mechanisms, and applications. Appl. Phys. Rev. 5, 041306.

31. Soleimani, Z., Zoras, S., Ceranic, B., Cui, Y., and Shahzad, S. (2021). A comprehensive review on the output voltage/power of wearable thermoelectric generators concerning their geometry and thermoelectric materials. Nano Energy 89, 106325.

32. Champier, D. (2017). Thermoelectric generators: A review of applications. Energy Convers. Manage. 140, 167–181.

33. Liu, D., Shi, P., Ren, W., Liu, Y., Liu, M., Zhang, Y., Tian, B., Lin, Q., Jiang, Z., and Ye, Z.-G. (2018b). Enhanced La0. 8Sr0. 2CrO3/Pt thin film thermocouple with Al2O3 coating layer for high temperature sensing. Ceram. Int. 44, S233–S237.

34. Zhang, Z., Liu, J., Cai, R., Liu, Z., Lei, J., Sun, R., Wu, N., Zhao, N., Tian, B., and Zhao, L. (2022a). High-Temperature-Sensing Smart Bolt Based on Indium Tin Oxide/In2O3 Thin-Film Thermocouples with Nickel-Based Single-Crystal Superalloy via Screen Printing. Chemosensors 10, 347.

35. Zhang, Z., Li, S., Tian, B., Liu, Z., Liu, J., Cheng, G., Fan, X., Fang, X., Zhao, N., and Zhao, L. (2022b). Simulation, fabrication, and characteristics of high-temperature, quick-response tungsten–rhenium thin-film thermocouples probe sensor. Meas. Sci. Technol. 33, 105105.

36. Kuzubasoglu, B.A., and Bahadir, S.K. (2020). Flexible temperature sensors: A review. Sens. Actuators, A 315, 112282.

37. Zhang, Z., Tian, B., Cheng, G., Liu, Z., Liu, J., Zhang, B., Lei, J., Zhao, N., Han, F., Fang, X., et al. (2022c). Influences of RF Magnetron Sputtering Power and Gas Flow Rate on a High Conductivity and Low Drift Rate of Tungsten-Rhenium Thin-Film Thermocouples. Nanomaterials *12*, 1120.

38. Zhang, Z., Tian, B., Li, L., Lei, J., Liu, Z., Liu, J., Cheng, G., Zhao, N., Fang, X., and Zhao, L. (2022d). Thermoelectricity and antivibration properties of screen-printed nanodoped In1. 35ZnO2. 11/In2O3 thin-film thermocouples on alumina substrates. Ceram. Int. *48*, 25747–25755.

39. Tian, B., Liu, Y., Zhang, Z., Liu, Z., Zhao, L., Lin, Q., Shi, P., Mao, Q., Lu, D., and Jiang, Z. (2020a). Effect of annealing on the thermoelectricity properties of the WRe26-In2O3 thin film thermocouples. Micromachines *11*, 664.

40. Liu, Z., Tian, B., Liu, J., Zhang, Z., Lin, Q., Mao, Q., Lu, D., and Jiang, Z. (2020a). Influences of annealing temperature on the thermoelectric properties of thin film thermocouples based on a flexible substrate by RF magnetron sputtering. Meas. Sci. Technol. *31*, 095101.

41. Liu, Y., Jiang, H., Zhao, X., Liu, B., Jia, Z., Liang, X., Deng, X., and Zhang, W. (2023a). Effects of nitrogen doping and oxygen vacancies on thermoelectric properties of Pt/N-doped ITO thin film thermocouples: first-principles calculations and experiments. J. Mater. Sci. *58*, 3219–3230.

42. Xie, S., Jiang, H., Zhao, X., and Deng, X. (2023). Fabrication and performances of high-temperature transient response ITO/In2O3 thin-film thermocouples. J. Mater. Sci. Mater. Electron. *34*, 339.

43. Tian, B., Liu, Z., Zhang, Z., Liu, Y., Lin, Q., Peng, S., and Jiang, Z. (2020b). Effect of film deposition rate on the thermoelectric output of tungsten-rhenium thin film thermocouples by DC magnetron sputtering. J. Micromech. Microeng. *30*, 065004.

44. Liu, Z., Tian, B., Fan, X., Liu, J., Zhang, Z., Luo, Y., Zhao, L., Lin, Q., Han, F., and Jiang, Z. (2020b). A temperature sensor based on flexible substrate with ultra-high sensitivity for low temperature measurement. Sens. Actuators, A *315*, 112341.

45. Tian, B., Liu, Z., Wang, C., Liu, Y., Zhang, Z., Lin, Q., and Jiang, Z. (2020c). Flexible four-point conjugate thin film thermocouples with high reliability and sensitivity. Rev. Sci. Instrum. *91*, 045004.

46. Liu, Z., Tian, B., Jiang, Z., Li, S., Lei, J., Zhang, Z., Liu, J., Shi, P., and Lin, Q. (2023b). Flexible temperature sensor with high sensitivity ranging from liquid nitrogen temperature to 1200° C. Int. J. Extreme Manuf. *5*, 015601.

47. Liu, Z., Tian, B., Liu, X., Zhang, X., Li, Y., Zhang, Z., Liu, J., Lin, Q., and Jiang, Z. (2023c). Multifunctional nanofiber mat for high temperature flexible sensors based on electrospinning. J. Alloys Compd. *941*, 168959.

48. Liu, Z., Tian, B., Fan, X., Zhang, Z., Liu, J., Lin, Q., Shi, P., Han, F., Mao, Q., and Jiang, Z. (2020c). Study on the characteristics of thermo-electrodes of various deposition parameters for the flexible temperature sensor. Rev. Sci. Instrum. *91*, 125004.

49. Liu, Z., Tian, B., Li, Y., Lei, J., Zhang, Z., Liu, J., Lin, Q., Lee, C., and Jiang, Z. (2023d). A large-area bionic skin for high-temperature energy harvesting applications. Nano Res. 1–11.

50. Liu, Y., Jiang, H., Zhao, X., Liu, B., Jia, Z., Deng, X., and Zhang, W. (2022). High temperature protection performance of sandwich structure Al2O3/Si3N4/YAlO multilayer films for Pt–Pt10% Rh thin film thermocouples. Ceram. Int. *48*, 33943–33948.

51. Ruan, Y., Xue, M., Teng, J., Wu, Y., and Shi, M. (2022). Horizontal Oxidation Diffusion Behavior of MEMS-Based Tungsten-Rhenium Thin Film Thermocouples. Materials *15*, 5071.

52. Heichal, Y., Chandra, S., and Bordatchev, E. (2005). A fast-response thin film thermocouple to measure rapid surface temperature changes. Exp. Therm. Fluid Sci. *30*, 153–159.

53. Basti, A., Obikawa, T., and Shinozuka, J. (2007). Tools with built-in thin film thermocouple sensors for monitoring cutting temperature. Int. J. Mach. Tools Manuf. *47*, 793–798.

54. Li, T., Shi, T., Tang, Z., Liao, G., Duan, J., Han, J., and He, Z. (2021). Real-time tool wear monitoring using thin-film thermocouple. J. Mater. Process. Technol. *288*, 116901.

55. Liu, H., Sun, W., Chen, Q., and Xu, S. (2011). Thin-film thermocouple array for time-resolved local temperature mapping. IEEE Electron. Device Lett. *32*, 1606–1608.

56. Cruz, S., Azevedo, G., Cano-Raya, C., Manninen, N., and Viana, J.C. (2021). Thermoelectric response of a screen printed silver-nickel thermocouple. Mater. Sci. Eng., B *264*, 114929.

57. Wei, Z., Zhao, J., He, H., Ding, G., Cui, H., and Liu, L. (2021). Future smart battery and management: Advanced sensing from external to embedded multi-dimensional measurement. J. Power Sources *489*, 229462.

58. Su, Y., Ma, C., Chen, J., Wu, H., Luo, W., Peng, Y., Luo, Z., Li, L., Tan, Y., Omisore, O.M., et al. (2020). Printable, highly sensitive flexible temperature sensors for human body temperature monitoring: a review. Nanoscale Res. Lett. *15*, 200.

59. Nag, A., Mukhopadhyay, S.C., and Kosel, J. (2017). Wearable flexible sensors: A review. IEEE Sens. J. *17*, 3949–3960.

60. Fan, Z., Zhang, Y., Pan, L., Ouyang, J., and Zhang, Q. (2021). Recent developments in flexible thermoelectrics: From materials to devices. Renew. Sustain. Energy Rev. *137*, 110448.

61. Versey, N.G., Gore, C.J., Halson, S.L., Plowman, J.S., and Dawson, B.T. (2011). Validity and reliability of temperature measurement by heat flow thermistors, flexible thermocouple probes and thermistors in a stirred water bath. Physiol. Meas. *32*, 1417–1424.

62. Miao, X., Gao, X., Su, K., Li, Y., and Yang, Z. (2022). A Flexible Thermocouple Film Sensor for Respiratory Monitoring. Micromachines *13*, 1873.

63. Xin, Y., Zhou, J., and Lubineau, G. (2019). A highly stretchable strain-insensitive temperature sensor exploits the Seebeck effect in nanoparticle-based printed circuits. J. Mater. Chem. *7*, 24493–24501.

64. Knoll, M., Offenzeller, C., Mayrhofer, B., Jakoby, B., and Hilber, W. (2018). A Screen Printed Thermocouple-Array on a Flexible Substrate for Condition Monitoring (MDPI), p. 803.

65. Liu, Z., Tian, B., Zhang, B., Liu, J., Zhang, Z., Wang, S., Luo, Y., Zhao, L., Shi, P., Lin, Q., and Jiang, Z. (2021a). A thin-film temperature sensor based on a flexible electrode and substrate. Microsyst. Nanoeng *7*, 42.

66. Saeidi-Javash, M., Du, Y., Zeng, M., Wyatt, B.C., Zhang, B., Kempf, N., Anasori, B., and Zhang, Y. (2021). All-printed MXene–graphene nanosheet-based bimodal sensors for simultaneous strain and temperature sensing. ACS Appl. Electron. Mater. *3*, 2341–2348.

67. Liu, H., Sun, W., and Xu, S. (2012). An extremely simple thermocouple made of a single layer of metal. Adv. Mater. *24*, 3275–3279.

68. Lee, C.-Y., Fang, L.-H., Su, A., Jung, G.-B., Huang, Y.T., and Liang, Y.C. (2015). Application of screen printing in flexible miniature thermocouple process development. Int. J. Electrochem. Sci. *10*, 3082–3087.

69. Sheng, A., Khuje, S., Li, Z., Yu, J., and Ren, S. (2022). Conformal Cu–CuNi Thermocouple Using Particle-Free Ink Materials. ACS Appl. Electron. Mater. *4*, 5558–5564.

70. Martiny, N., Rheinfeld, A., Geder, J., Wang, Y., Kraus, W., and Jossen, A. (2014). Development of an all kapton-based thin-film thermocouple matrix for in situ temperature measurement in a lithium ion pouch cell. IEEE Sens. J. *14*, 3377–3384.

71. Kostyuk, O., Dzundza, B., Yavorsky, Y.S., and Dashevsky, Z. (2021). Development of thermal detector based on flexible film thermoelectric module. Phys. Chem. Solid St. *22*, 45–52.

72. Shi, Z., Zhang, J., Wang, W., Zhang, Y., Chen, B., and Shi, P. (2022). Effect of SiO2 buffer layer on thermoelectric response of In2O3/ITO thin film thermocouples. J. Alloys Compd. *902*, 163838.

73. Liu, Z., Tian, B., Zhang, B., Zhang, Z., Liu, J., Zhao, L., Shi, P., Lin, Q., and Jiang, Z. (2021b). High-Performance Temperature

Sensor by Employing Screen Printing Technology. Micromachines *12*, 924.

74. Zaferani, S.H., Ghomashchi, R., and Vashaee, D. (2019). Strategies for engineering phonon transport in Heusler thermoelectric compounds. Renew. Sustain. Energy Rev. *112*, 158–169.

75. Pomeranchuk, I. (1942). On the thermal conductivity of dielectrics at temperatures lower than that of Debye. J. Phys.-USSR *6*, 237.

76. Woolley, J. (1957). Book Review: Introduction to solid state physics. C. KITTEL: Chapman and Hall, 1956. 617 pp., 96 s. J. Mech. Phys. Solid. *6*, 83.

77. Pomeranchuk, I. (1941). On the thermal conductivity of dielectrics. Phys. Rev. *60*, 820.

78. Wen, D.-L., Deng, H.-T., Liu, X., Li, G.-K., Zhang, X.-R., and Zhang, X.-S. (2020). Wearable multi-sensing double-chain thermoelectric generator. Microsyst. Nanoeng. *6*, 68.

79. Li, G., Han, D., Yang, F., Wang, Z., Pi, Y., Wang, W., and Xu, S. (2017a). Linearly enhanced response of thermopower in cascaded array of dual-stripe single-metal thermocouples. Appl. Phys. Lett. *110*, 203505.

80. Chen, Z., Zhao, D., Ma, R., Zhang, X., Rao, J., Yin, Y., Wang, X., and Yi, F. (2021). Flexible temperature sensors based on carbon nanomaterials. J. Mater. Chem. B *9*, 1941–1964.

81. Lichtenwalner, D.J., Hydrick, A.E., and Kingon, A.I. (2007). Flexible thin film temperature and strain sensor array utilizing a novel sensing concept. Sens. Actuators, A *135*, 593–597.

82. Zhao, J., Li, H., Choi, H., Cai, W., Abell, J.A., and Li, X. (2013). Insertable thin film thermocouples for in situ transient temperature monitoring in ultrasonic metal welding of battery tabs. J. Manuf. Process. *15*, 136–140.

83. Zhang, M., Wang, X., Huang, Z., and Rao, W. (2020). Liquid metal based flexible and implantable biosensors. Biosensors *10*, 170.

84. Khan, Y., Ostfeld, A.E., Lochner, C.M., Pierre, A., and Arias, A.C. (2016). Monitoring of vital signs with flexible and wearable medical devices. Adv. Mater. *28*, 4373–4395.

85. Li, Q., Zhang, L.N., Tao, X.M., and Ding, X. (2017b). Review of flexible temperature sensing networks for wearable physiological monitoring. Adv. Healthc. Mater. *6*, 1601371.

86. Nyabadza, A., Vázquez, M., Coyle, S., Fitzpatrick, B., and Brabazon, D. (2021). Review of materials and fabrication methods for flexible nano and micro-scale physical and chemical property sensors. Appl. Sci. *11*, 8563.

87. Cheung, T.W., Liu, T., Yao, M.Y., Tao, Y., Lin, H., and Li, L. (2022). Structural development of a flexible textile-based thermocouple temperature sensor. Text. Res. J. *92*, 1682–1693.

88. Ruoho, M., Juntunen, T., Alasaarela, T., Pudas, M., and Tittonen, I. (2016). Transparent, flexible, and passive thermal touch panel. Adv. Mater. Technol. *1*, 1600204.

89. Zhang, X., and Zhao, L.-D. (2015). Thermoelectric materials: Energy conversion between heat and electricity. J. Materiomics *1*, 92–105.

90. Han, D., Li, G., Zhou, S., Wang, Z., Yang, F., and Xu, S. (2017). To save half contact pads in 2D mapping of local temperatures with a thermocouple array. RSC Adv. *7*, 9100–9105.

91. Jeon, J.G., Kim, H.J., Shin, G., Han, Y., Kim, J.H., Lee, J.H., Lee, J., Lim, H., Ha, S., Bae, M., et al. (2022). High-Precision Ionic Thermocouples Fabricated Using Potassium Ferri/Ferrocyanide and Iron Perchlorate. Adv. Electron. Mater. *8*, 2100693.

92. Renn, M.J., Schrandt, M., Renn, J., and Feng, J.Q. (2017). Localized laser sintering of metal nanoparticle inks printed with Aerosol Jet® technology for flexible electronics. J. Microelectron. Electron. Packag *14*, 132–139.

93. Tang, Y.-Q., Fang, W.-Z., Lin, H., and Tao, W.-Q. (2019). Thin film thermocouple fabrication and its application for real-time temperature measurement inside PEMFC. Int. J. Heat Mass Transfer *141*, 1152–1158.

94. Ali, S.T., Lebæk, J., Nielsen, L.P., Mathiasen, C., Møller, P., and Kær, S.K. (2010). Thin film thermocouples for in situ membrane electrode assembly temperature measurements in a polybenzimidazole-based high temperature proton exchange membrane unit cell. J. Power Sources *195*, 4835–4841.

95. Konishi, S., and Hirata, A. (2019). Flexible temperature sensor integrated with soft pneumatic microactuators for functional microfingers. Sci. Rep. *9*, 15634.

96. Rahman, M.T., Cheng, C.-Y., Karagoz, B., Renn, M., Schrandt, M., Gellman, A., and Panat, R. (2019). High performance flexible temperature sensors via nanoparticle printing. ACS Appl. Nano Mater. *2*, 3280–3291.

97. Assumpcao, D., Kumar, S., Narasimhan, V., Lee, J., and Choo, H. (2018). High-performance flexible metal-on-silicon thermocouple. Sci. Rep. *8*, 13725.

98. Lee, S.H., Shen, H., and Han, S. (2019). Flexible thermoelectric module using Bi-Te and Sb-Te thin films for temperature sensors. J. Electron. Mater. *48*, 5464–5470.

99. Seo, B., Hwang, H., Kang, S., Cha, Y., and Choi, W. (2018). Flexible-detachable dual-output sensors of fluid temperature and dynamics based on structural design of thermoelectric materials. Nano Energy *50*, 733–743.

100. Shi, P., Wang, W., Liu, D., Zhang, J., Ren, W., Tian, B., and Zhang, J. (2019). Structural and electrical properties of flexible ITO/In2O3 thermocouples on PI substrates under tensile stretching. ACS Appl. Electron. Mater. *1*, 1105–1111.

101. Mutyala, M.S.K., Zhao, J., Li, J., Pan, H., Yuan, C., and Li, X. (2014). In-situ temperature measurement in lithium ion battery by transferable flexible thin film thermocouples. J. Power Sources *260*, 43–49.

102. Li, H., Yang, Y., and Liu, J. (2012). Printable tiny thermocouple by liquid metal gallium and its matching metal. Appl. Phys. Lett. *101*, 073511.

103. Bianchi, C., Loureiro, J., Duarte, P., Marques, J., Figueira, J., Ropio, I., and Ferreira, I. (2016). V2O5 thin films for flexible and high sensitivity transparent temperature sensor. Adv. Mater. Technol. *1*, 1600077.

104. Chen, Y., Lei, H., Gao, Z., Liu, J., Zhang, F., Wen, Z., and Sun, X. (2022). Energy autonomous electronic skin with direct temperature-pressure perception. Nano Energy *98*, 107273.

105. Zhang, F., Zang, Y., Huang, D., Di, C.-a., and Zhu, D. (2015). Flexible and self-powered temperature–pressure dual-parameter sensors using microstructure-frame-supported organic thermoelectric materials. Nat. Commun. *6*, 8356.

106. Jung, M., Lee, J., Vishwanath, S.K., Kwon, O.-S., Ahn, C.W., Shin, K., and Jeon, S. (2020). Flexible multimodal sensor inspired by human skin based on hair-type flow, temperature, and pressure. Flexible Printed Electron. *5*, 025003.

107. Xu, S., Fan, Z., Yang, S., Zhao, Y., and Pan, L. (2021). Flexible, self-powered and multi-functional strain sensors comprising a hybrid of carbon nanocoils and conducting polymers. Chem. Eng. J. *404*, 126064.

108. Chi, C., An, M., Qi, X., Li, Y., Zhang, R., Liu, G., Lin, C., Huang, H., Dang, H., Demir, B., et al. (2022). Selectively tuning ionic thermopower in all-solid-state flexible polymer composites for thermal sensing. Nat. Commun. *13*, 221.

109. Miao, T., Shi, S., Yan, S., Ma, W., Zhang, X., Takahashi, K., and Ikuta, T. (2016). Integrative characterization of the thermoelectric performance of an individual multiwalled carbon nanotube. J. Appl. Phys. *120*, 124302.

110. Albrecht, A., Rivadeneyra, A., Bobinger, M., Calia, J.B., Loghin, F.C., Salmeron, J.F., Becherer, M., Lugli, P., and Falco, A. (2018). Scalable deposition of nanomaterial-based temperature sensors for transparent and pervasive electronics. J. Sens. *2018*.

111. Hassan, W.H. (2019). Current research on Internet of Things (IoT) security: A survey. Comput. Netw. *148*, 283–294.

112. Bakker, A., and Huijsing, J.H. (1996). Micropower CMOS temperature sensor with

digital output. IEEE J. Solid-State Circuits *31*, 933–937.

113. Vaz, A., Ubarretxena, A., Zalbide, I., Pardo, D., Solar, H., Garcia-Alonso, A., and Berenguer, R. (2010). Full passive UHF tag with a temperature sensor suitable for human body temperature monitoring. IEEE Trans. Circuits Syst. II Express Briefs *57*, 95–99.

114. Tian, B., Xing, Y., Zhang, X., Liu, Z., Zhang, Z., Liu, J., Zhang, B., Lin, Q., and Jiang, Z. (2022). A High-Precision Three-Dimensional Probe Array Temperature Sensor. Chemosensors *10*, 309.

115. Ye, Z., Ling, Y., Yang, M., Xu, Y., Zhu, L., Yan, Z., and Chen, P.-Y. (2022). A breathable, reusable, and zero-power smart face mask for wireless cough and mask-wearing monitoring. ACS Nano *16*, 5874–5884.

116. Lee, J.H., Heo, J.S., Kim, Y.J., Eom, J., Jung, H.J., Kim, J.W., Kim, I., Park, H.H., Mo, H.S., Kim, Y.H., and Park, S.K. (2020). A behavior-learned cross-reactive sensor matrix for intelligent skin perception. Adv. Mater *32*, 2000969.

117. Liu, Q., Tai, H., Yuan, Z., Zhou, Y., Su, Y., and Jiang, Y. (2019). A high-performances flexible temperature sensor composed of polyethyleneimine/reduced graphene oxide bilayer for real-time monitoring. Adv. Mater. Technol *4*, 1800594.