Review

Flexible ferroelectric wearable devices for medical applications

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SUMMARY

Wearable electronics are becoming increasingly important for medical applications as they have revolutionized the way physiological parameters are monitored. Ferroelectric materials show spontaneous polarization below the Curie temperature, which changes with electric field, temperature, and mechanical deformation. Therefore, they have been widely used in sensor and actuator applications. In addition, these materials can be used for conversion of human-body energy into electricity for powering wearable electronics. In this paper, we review the recent advances in flexible ferroelectric materials for wearable human energy harvesting and sensing. To meet the performance requirements for medical applications, the most suitable materials and manufacturing techniques are reviewed. The approaches used to enhance performance and achieve long-term sustainability and multi-functionality by integrating other active sensing mechanisms (e.g. triboelectric and piezoresistive effects) are discussed. Data processing and transmission as well as the contribution of wearable piezoelectric devices in early disease detection and monitoring vital signs are reviewed.

INTRODUCTION

In the era of the Internet of Things (IoT), our lives have been dramatically improved compared to those of previous generations. This swift progress is attributed to the consistent and rapid development in the collection of information and their analysis, transmission, and storage. This progress has the potential to create significant impact on a range of devices for medical and healthcare monitoring. Currently, intelligent medical devices are able to diagnose, monitor, or treat diseases, and allow people with physical impairments to lead more independent lives. More specifically, flexible multifunctional sensing devices have become one of the emerging technologies in the wearable device sector (Xie et al., 2019). The wearable healthcare device market is rapidly growing and is forecasted to save 1.3 million lives annually, with a market size growth to $72.7 billion by 2021 (Banerjee et al., 2018).

Despite the significant progress in wearable sensing devices, the majority of these systems continue to be powered by batteries. In the healthcare industry, this is a notable drawback as batteries frequently need to be recharged or replaced. One possible solution toward their energy autonomy is to harvest mechanical energy from the human body to generate electricity using a ferroelectric generator, which is able to convert mechanical energy into electrical energy by the *direct piezoelectric effect*. As a result, there is a substantial and growing interest in synthesizing flexible wearable devices using piezoelectric materials as they can both be used for sensing and energy harvesting. The sensing performance of piezoelectric materials is directly related to the piezoelectric strain coefficient d_{ij} (charge per unit force) and g_{ij} (electric field per unit stress) (Duan et al., 2020). In addition, for energy harvesting applications, a low dielectric loss is favorable to maximize the efficiency of conversion from mechanical to electrical energy (Roscow et al., 2015).

As a sub-category of piezoelectrics, *pyroelectric* materials exhibit a spontaneous polarization that changes with temperature. These materials exhibit a change in the level of bound charge when they are heated or cooled. Some pyroelectric materials are *ferroelectric*, in that they exhibit a spontaneous electric polarization that can be switched by an electric field or mechanical stress giving rise to characteristic hysteretic behavior (strain-electric field and polarization-electric field). As a result, all ferroelectric materials are both pyroelectric and piezoelectric, since their polarization changes with temperature and stress, respectively. The inherent multifunctionality of ferroelectric materials to sense and harvest energy from mechanical vibrations (piezoelectric performance) as well as thermal fluctuations (pyroelectric performance) and

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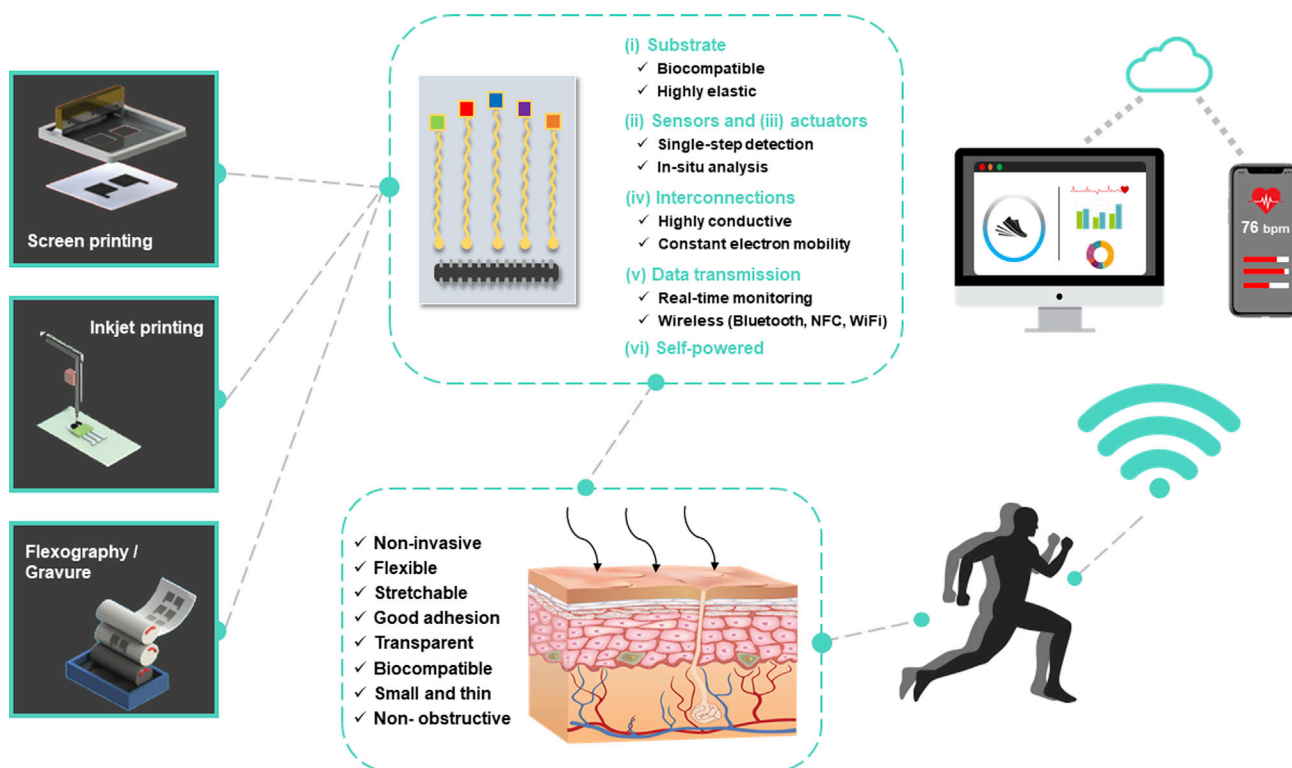
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Figure 1. Wearable medical devices: from the fabrication to the final application
Schematic illustrating a typical self-powered flexible wearable electronic device composed of substrate, sensing, interconnects, wireless transmission, and energy suppling elements from initial fabrication to final application.

their ability to be used in polymer or composite form offer unique advantages to enable mechanical flexibility and functionalization of wearable devices (Ryu et al., 2019; Thakre et al., 2019).

A broad overview, from fabrication to the final application, of a typical self-powered flexible wearable electronic device composition is shown in Figure 1. Typically, wearable electronics are comprised of six main components: (i) substrate, (ii) sensors, (iii) actuators, (iv) interconnections, (v) wireless transmission, and (vi) energy supply (Dharmasena et al., 2019; Gao et al., 2020; Li et al., 2020a). As a result, a stacked layer by layer method is often used to incorporate the different elements together into a final device architecture (Ma et al., 2020). A multicomponent device is necessary to exhibit sufficient mechanical robustness and electronic performance, and each component should be designed to withstand the high strains experienced during operation (Amjadi et al., 2016; Gunawardhana et al., 2020). More specifically, interconnections must be flexible to ensure constant electron mobility, while the piezoelectric active sensing materials must be able to measure stimulus from specific parts of the body and convert them into signals (Wang et al., 2015).

In this paper, we review recent developments in wearable piezoelectric devices and discuss their potential healthcare monitoring applications. We discuss the range of fabrication techniques that can be useful for cost-effective and scalable manufacturing of flexible wearable piezoelectric devices. Finally, we review the most suitable piezoelectric materials with a focus on their capability for detecting multiple physical stimuli and multifunctionality, which can be achieved by combining them with other sensing or harvesting mechanisms, such as *triboelectric* and *piezoresistive* effects.

A better understanding of the required properties and features of wearable flexible devices for medical applications can promote innovation and future developments. The most crucial features for a wearable system are to achieve mechanical flexibility and stretchability (Koo et al., 2018; Ma et al., 2020). Consequently, it can be understood that a device's functional properties are strongly associated with their mechanical properties. For wearable sensing devices to be fully stretchable, they must meet a combination

of different mechanical requirements which include a low Young's modulus, high elastic limit and high ultimate strain (strain to failure). When the Young's modulus is sufficiently small in broad stress range, the desired mechanical and electrical properties of the device are less likely to be affected by any direct contact with the dynamic and complex structure of the human skin (Lipomi, 2016; Tian et al., 2020).

In 21st century clothing, items such as gloves, headsets, wristwatches, armbands, bracelets, and glasses play a prominent role in our lifestyles. These all have the potential to be functionalized if flexible multifunctional devices can be integrated into or onto them. However, the devices must be fully matched to the skin surface. For example, a device mounted on the curved and wavy surface of the skin needs to exhibit small, thin, soft, dry, and non-obstructive form factors. High optical transparency of the materials is also preferred in wearable devices for medical applications, since they are mounted directly on the human skin and need to be aesthetically pleasing and invisible during daily activities (Kim et al., 2018). Good adhesion between the human skin and device surface is also needed, and good adhesion between the different layers of the device ensures appropriate transfer of any applied strains. To achieve an interfacial adhesion between the device and the human body, medical acrylic adhesives have been used previously (Lim et al., 2020). However, this form of adhesion can lead to allergic contact dermatitis and, as a result, recent studies have focused on dry methods, such as van der Walls or electrostatic forces for adequate interfacial adhesion (McNichol et al., 2013).

A wearable flexible device should be designed for long-term wearing and good adhesion with the human skin for the purpose of continuous and highly accurate biometric signal monitoring. In order to prevent skin inflammation, any device must exhibit high permeability to oxygen and water (Lewis, 2006). If the device blocks the sweat glands, the skin is not able to breathe. Furthermore, low toxicity or non-toxic and non-hazardous (e.g. non-flammable) materials are required for wearable flexible device development. A device that is intended for medical applications must also be biocompatible (Wang et al., 2018). Body temperature (BT) can affect negatively both pressure and strain sensors; therefore, a low-temperature coefficient of resistance is desirable so that the effect of temperature on these sensors is minimized (Lim et al., 2020).

Finally, in order to enable transfer of research into an affordable technology, large scalability and cost-effectiveness are the important characteristics that must be considered. Progress in the processing steps to overcome these challenges has been already made (Sreenilayam et al., 2020), and processing strategies have focused on the conversion of the previously used conventional manufacturing processes (microfabrication methods) into large-scale printing techniques. Furthermore, in order to decrease the cost of the final conventional device, solution-synthesized materials are preferred.

MANUFACTURING TECHNIQUES FOR FLEXIBLE WEARABLE DEVICES

For the development of wearable flexible sensing devices, innovative approaches in both materials science and fabrication process are required. Having introduced the main components of a health-care monitoring wearable device, see Figure 1, the fabrication strategies used for the integration of each component into a functional system are now presented. For the development of the individual components, a wide range of processing techniques are necessary. The sensing elements, microprocessor (MPU), and interconnection development usually rely on microfabrication methods, which include chemical vapor deposition, sputtering, reactive ion etching, wet etching, thermal or electron-beam evaporation, and photolithography (duToit and Wardle, 2006; Mannsfeld et al., 2010; Ramadan et al., 2014). However, microfabrication methods usually face challenges when used with flexible substrates. The circuits must be fabricated first on rigid silicon wafers and then transferred subsequently onto flexible/stretchable substrates so that they can conform perfectly with human skin. Either *flip-chip bonding* or a *pick-and-place* process can be used for the transferring of the perfricated circuits (Kim et al., 2015). Deposition and good adhesion between the different surfaces can be achieved using low-temperature solders or adhesive printed electrodes (Khan et al., 2016; Kim et al., 2015). In order to prevent the fracture of devices, the interconnections of the system must be able to accommodate the strains that are transferred through the substrate. This can be achieved by delaminating them from the substrate or by pattering them in kirigami structures, such as a serpentine configuration (Yan et al., 2019). A schematic representation of a serpentine configuration is presented in Figure 2.

Solution-processable materials are the key factor for scalability and cost reduction of sensing element fabrication. Consequently, different approaches are required for the development of flexible sensing devices. Among the available manufacturing techniques, printing techniques are particularly attractive, not
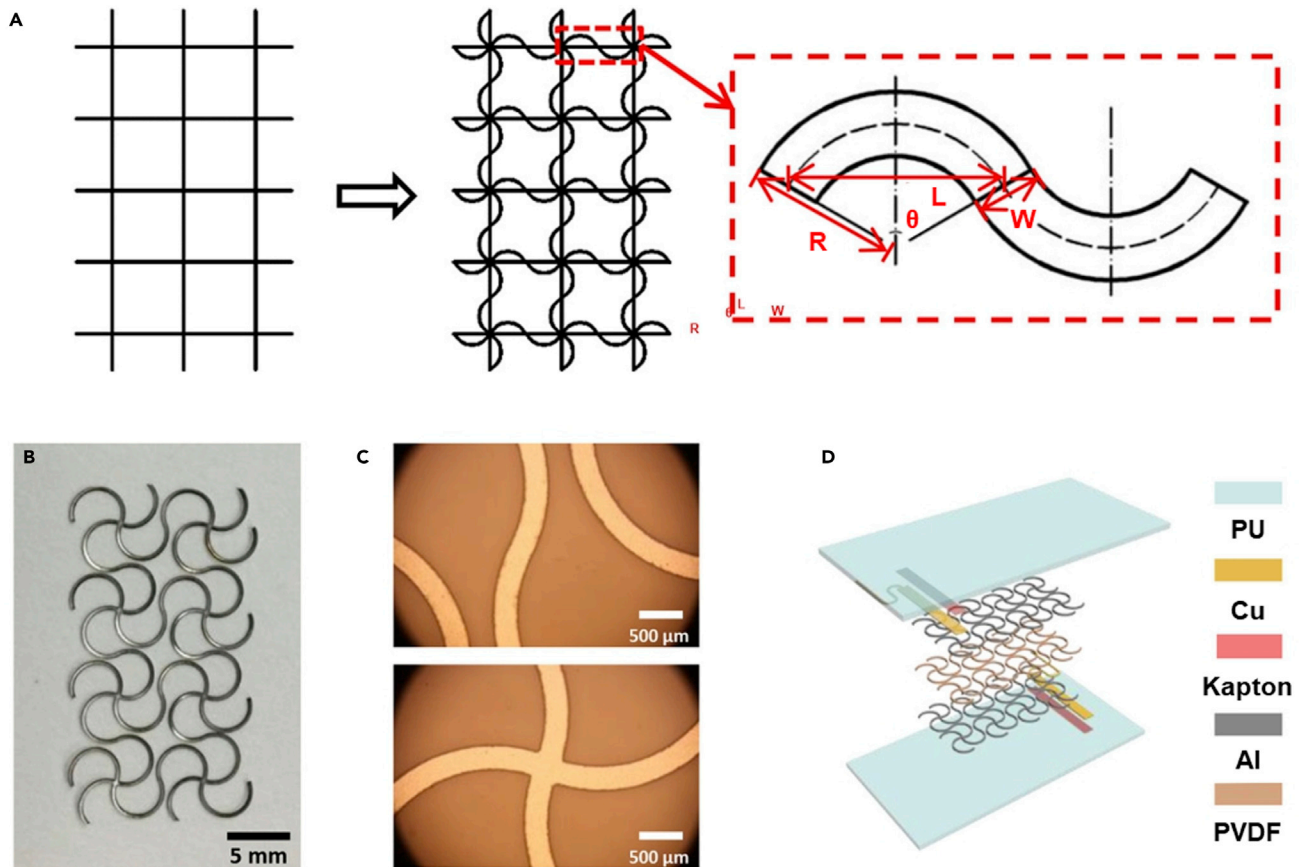
Figure 2. Schematic representation of the serpentine layout

(A) The structure of the presented serpentine configuration is based on a mesh geometry, which consists of serpentine unit cells along with the vertical and horizontal directions. Each serpentine unit cell includes two identical circular arcs with the same arc angle, radius, width, and span.

(B) Optical image of the polyvinylidene difluoride (PVDF) film with a serpentine layout.

(C) Zoomed optical images of the polyvinylidene difluoride (PVDF) film with a serpentine layout indicating that there is not crack.

(D) Exploded view schematic of the micromotion sensor with a serpentine layout

[Copyright 2019, American Chemical Society] (Yan et al., 2019).

only because they reduce the cost of the device in terms of production but are also preferable for large sensor fabrication compared to microfabrication techniques since they are not limited by the wafer throughput. As a result, transfer processes are not required since the sensors can be printed directly onto the flexible plastic or elastomeric substrate. A further advantage of printing techniques is *additivity*, since the desired sensor patterns can be printed onto the substrate in a layer by layer approach. These features also reduce the required steps during manufacturing, leading to a rapid production route (Sreenilayam et al., 2020; Suganuma, 2014a). In the literature, screen and inkjet printing are techniques which are most commonly used for the fabrication of flexible wearable electronic devices (Khan et al., 2016). Processes such as gravure and flexographic printing still require further research in order to be used for commercial manufacturing applications. Low viscosity inks are used to uniformly spin-coat on small areas (Cardoso et al., 2012); however, while spin coating is popular, especially when combined with low viscosity ink processing techniques, the scalability of the approach continues to be challenging. Recent studies (Chen et al., 2017a; Shao et al., 2019) have shown that nanoimprinting lithography, which is a large-area fabrication method with high resolution and efficiency, could be an ideal candidate for future wearable medical devices development.

Furthermore, due to the need for direct integration of flexible sensors with the soft curved surfaces of a human body, particular attention must be given to a material's structural design in order to ensure uniform output generation, robust output stability, and an ability to be fabricated at scale fabrication. Moreover,

as with the abovementioned example of the interconnection with a serpentine configuration, the device geometry can be optimized to enhance the stretchable properties. The printing of multi-pixel arrays in specific patterns is a commonly used geometry structure for printed sensor development since distributed pixels are able to detect the force distribution with higher accuracy compared to a sandwiched planar film configuration (Chen et al., 2020; Ding et al., 2020). A brief discussion on the latest trends for wearable ferroelectric medical device fabrication is given in this section.

Screen printing techniques

Screen printing is a mature technique which has been used to fabricate electronic devices for more than three decades (Krebs, 2009). This technique offers reproducibility, reliability and mass production with low cost (Kim et al., 2017b). A typical screen-printing system consists of a substrate, a screen, a squeegee and a press bed (Emamian et al., 2017). In the screen-printing technique, the squeegee is moved across the screen and forces the ink through the open mesh apertures and onto the substrate. The closed and open pores of the patterned mesh define the desired pattern including the minimum feature size and the deposition thickness. High viscosity inks are preferred (0.5–5 $Pa \times s$), since inks with a low viscosity can easily run through the mesh, reducing the resolution of the desired pattern. If the viscosity of the ink is too high, however, the mesh can become blocked and result in incomplete pattern transfer. Therefore, the properties of the ink and the density of the mesh are important parameters that play a major role on print thickness and resolution. A desired screen printed line should exhibit thickness of 5–250 μm and width of 50–100 μm (Bariya et al., 2018; Krebs, 2009; Tobjörk and Österbacka, 2011). There are two assemblies of screen printing: *flat-bed* and *rotary screen* printing. The process of rotary screen printing allows for a high throughput but it is usually more expensive compared to the manually flat-bed screen printing technique. The main drawback of the screen printing methods is the large volume of non-recoverable ink waste, which leads to further time wastage as the mesh and the stencil must be thoroughly cleaned after each use (Kamarudin et al., 2020).

Screen printed wearable piezoelectric devices have been developed for monitoring physiological signals (Fares et al., 2020; Gonçalves et al., 2019; Laurila et al., 2019; Sato et al., 2019). Sekine et al. (Sekine et al., 2018) created a fully screen-printed ferroelectric poly(vinylidene fluoride-trifluoroethylene) P(VDF-TrFE)-based sensing device for monitoring of human pulse wave/rate on the human skin. Emamian et al. (Emamian et al., 2017) developed a screen printed piezoelectric based touch sensor using ferroelectric PVDF as the active sensing layer. PET (polyethylene terephthalate) and paper were used as flexible substrates. Piezoelectric voltage analysis demonstrated that this printed system could be used as both a touch and force sensor. Fares et al. (Fares et al., 2020) demonstrated a skin attachable piezopolymer screen-printed sensor as shown in Figure 3. A variety of skin patches were designed and developed to provide a sensing capability to a glove and prosthetic hand. The average piezoelectric charge constant for all sensors was measured at incremental preloads (1–3 N) and are presented in a detailed table that can be found in the paper (Fares et al., 2020).

Inkjet printing and 3D printing techniques

In recent years, multi-material *additive manufacturing* (AM) or *3D printing* techniques have led to a revolution in the manufacture of structures with complex electronic components. The majority of 3D printing techniques use a single material for printing complex shaped parts (Bandyopadhyay and Heer, 2018). Fused deposition modeling, stereolithography, 3D material jetting and inkjet printing are the most commonly used 3D printing techniques. Each of these methods rely on localized and controlled dispensing of ink on the receiving substrate in a table-free manner using a nozzle to drive the ink. For an easy and reproducible registration of the ink, these nozzle-based methods require low viscosity inks (10-20 $mPa \times s$) compared to the other printing techniques (Suganuma, 2014b). Within this review, we will refer and analyze inkjet printing since it is the most widely used 3D printing technique for flexible ferroelectric medical devices development.

Inkjet printing is an inexpensive, fully digital, and non-impact method for the development of stretchable and wearable sensing devices. Any inkjet printing process consists of three main components: (i) the inkjet printer, which includes an ink tank and a printer head with micrometer-sized nozzles, (ii) the ink, and (iii) the substrate (Gao et al., 2017). The process provides benefits in terms of a low fabrication temperature, compatibility with substrates (various flexible substrates with planar or non-planar surfaces such as paper, cotton, synthetic textiles, and polymers can be used), waste reduction, controlled deposition, and additive
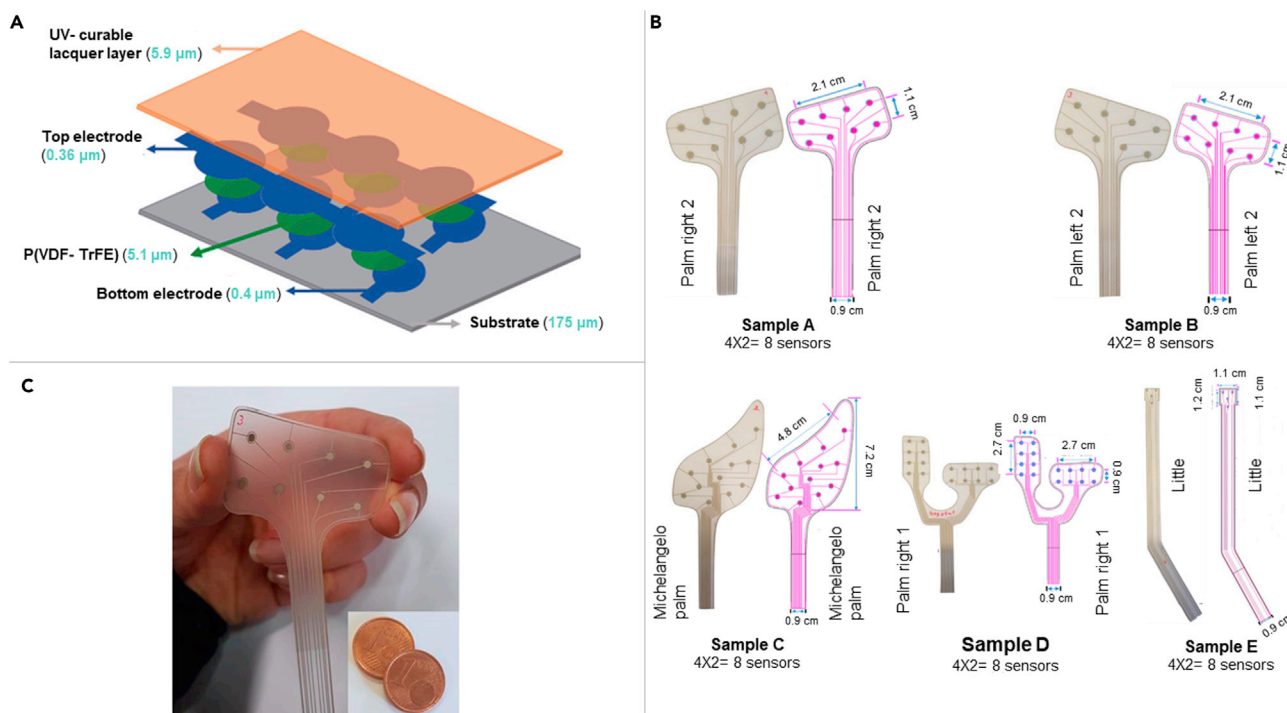
Figure 3. Screen printed PVDF-based wearable sensing device
(A) Sketch of the sensing patch.
(B) Different designs.
(C) Optical image of the final product.
(Fares et al., 2020), [Copyright 2020, MDPI].

patterning (Al-Halhouli et al., 2018; Yin et al., 2010). However, inkjet printing often exhibits limitations regarding the speed of the printing process and potential clogging of the nozzles, even when inks with low viscosity and low evaporation rates are used (Tobjörk and Österbacka, 2011). Another challenge of this technique is the so-called *coffee stain* effect, which is associated with ink droplets evaporating unevenly and results in the creation of a clear ring on their perimeter that affects the homogeneity of the printed pattern (Deegan et al., 1997). There are two modes of inkjet printing, *Drop on Demand* (DoD) and *Continuous Inkjet Printing* (CIJ). The most commonly reported in the literature is the DoD mode that uses a piezoelectric actuator to eject droplets from the nozzle, which has a higher controllability and consistency compared to CIJ printing systems (Al-Halhouli et al., 2018).

By employing an inkjet printer, high-resolution conductive patterns can be printed easily for wearable flexible devices. The performance of an inkjet-printed wearable device depends on the quality of the printed pattern as well as the mechanical flexibility, stretchability, and durability of the final device. In reported work, this form of device has been used to detect temperature changes (Ali et al., 2019; Vuorinen et al., 2016), humidity (Zhang et al., 2018), strain, and pressure on the human body (Karim et al., 2019; Lo et al., 2020). Thuau et al. used an inkjet printer to print fluorinated ferroelectric P(VDF-TrFE) polymer layers and silver electrodes on a polymeric substrate. The inkjet-printed active sensing layer exhibited piezoelectric performance (output voltage of 250 mV) and a dielectric constant ($\varepsilon' = 12$ at 100 Hz) comparable to spin-coated films (Thuau et al., 2017). A remanent polarization of 7.8 μC/cm^2 and d_{31} piezoelectric coefficient of 10.4 pm/V were also obtained. Al-Halhouli et al. used the inkjet printing technique to develop a stretchable and wearable strain gauge sensor (gauge factor >100) which could accurately measure the respiratory volume change in the chest area at five different postures; this included standing, sitting at 90°, Fowler's position at 45°, supine and right lateral recumbent (Al-Halhouli et al., 2020).

Gravure printing techniques

Gravure or *rotogravure* is a contact printing method suitable for high volume and high-quality pattern fabrication on flexible absorbent or non-absorbent substrates (Sreenilayam et al., 2020). A gravure printer

consists of a metallic cylinder in which micron-sized cells are engraved either electromechanically, by chemical etching, or by laser, and an enclosed chamber supplies the cylinder with ink. The metallic cylinders are expensive to produce but they exhibit a long lifetime. In this technique, the ink is transferred by surface tension from the small engraved cavities in the gravure cylinder to the substrate. Both the properties of the inks and the geometric features (i.e. the width and depth) of the microcells play an important role in gravure printing. A satisfactory ink transfer on the substrate can be achieved using low viscosity inks (10-100 m$Pa.s$) and high pressures (1-5 MPa) during the printing process. The gravure printing technique is not suitable for devices where there are not enough printed layers on the substrate as high pressures can easily result in mechanical failure. Another challenge of this technique arises from the roughness of the flexible substrate, which can lead to small unprinted areas (Bariya et al., 2018; Saengchairat et al., 2017; Secor et al., 2014). These limitations can be overcome by optimizing the properties of the ink, the microcells of the cylinder and the substrate materials, as well as by adjusting the pressure and the speed of the printer. Tuukkanen et al. fabricated various solution-processable materials using poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS), as well as composite inks made of carbon nanotubes (CNTs) and carboxymethyl cellulose. The viscosity of the inks used in this study was suitable for gravure and flexographic printed electrodes for ferroelectric thin film sensor development (Tuukkanen et al., 2013).

Flexographic printing technique

Flexographic printing is a high-speed roll-to-roll process that uses a flexible rubber/polymer printing plate cylinder, in which the desired pattern is carved in *bas-relief* to ensure efficient transfer of the ink to the substrate (Deganello et al., 2012). The ink is transferred onto the printing plate cylinder through an anilox roller with small engraved cells embedded on the exterior surface. A fountain roller which is partly immersed in an ink bath continuously supplies the anilox cylinder with ink. A doctor blade is used to remove the excess ink from the non-engraved surface of the anilox roller ensuring the homogeneous distribution of the ink and the good controllability of the wet ink thickness. The thickness is defined by the volume of the small cells in the anilox roller that can carry a specific amount of ink and results in a constant thickness of the transferred ink. A continuous print pattern can be achieved using inks with low viscosity and surface tension and by controlling the printing pressure. The viscosity of the ink for flexographic printing is usually <500 m$Pa.s$. Flexographic and gravure printing techniques share many similarities (i.e. the ink viscosity requirement, usage of cylinders and plates, high volume printing, etc.) although the resolution in flexographic printing is usually lower as the patterns (30-80 μm) on the flexible printing plate can be distorted during printing. A further common challenge in flexographic printing is the formation of halo-like shapes (coffee-strain effect) around the edges of a pattern, which are formed when high pressures are applied to the substrate. A schematic of flexographic and gravure printing methods is shown in Figure 4. In the case of the flexographic method, Figure 4 (a), the ink is firstly transferred to the printing plate of the printing cylinder before filling the cells of the anilox roller and finally being transferred to the substrate. In the gravure printing method, Figure 4 (b), the ink fills the cells of the gravure cylinder and is then transferred directly onto the substrate.

Flexography is a popular method since it can be used for printing on a variety of substrates such as thin, flexible, and solid materials, thick cardboards, papers, fabrics, and packaging materials with rough surfaces (Izdebska, 2016). Flexographic printing has been also used for the development of piezoelectric energy harvesting devices. For example, Ali et al. (Ali et al., 2014) developed a fully printed piezoelectric ZnO/polymer hybrid diode-based rectifier on a PET substrate. A printed piezoelectric device exhibited output voltage of 0.425 V. The output voltage increased linearly with the number of the printed devices connected in series.

In Table 1, the abovementioned fabrication techniques are summarized and compared with respect to the required ink viscosity, the achievable printing resolution, the printed film thickness and the printing speed for each method. Furthermore, other parameters of interest include the required processing steps, the material wastage and details of whether the technique can be used for printing on flexible substrates. The ideal process for the commercialization of flexible medical devices should consist of a minimum number of processing steps and the different layers of the device should be roll-to-roll (R2R) printed. The properties of the ink, such as surface tension, evaporation rate and viscosity, and its surface characteristics such as surface energy, roughness, and porosity, determine the processing speed and the achievable thickness of the layers (Al-Halhouli et al., 2020; Bariya et al., 2018; Kamarudin et al., 2020; Saengchairat et al., 2017; Sreenilayam et al., 2020; Tobjörk and Österbacka, 2011).
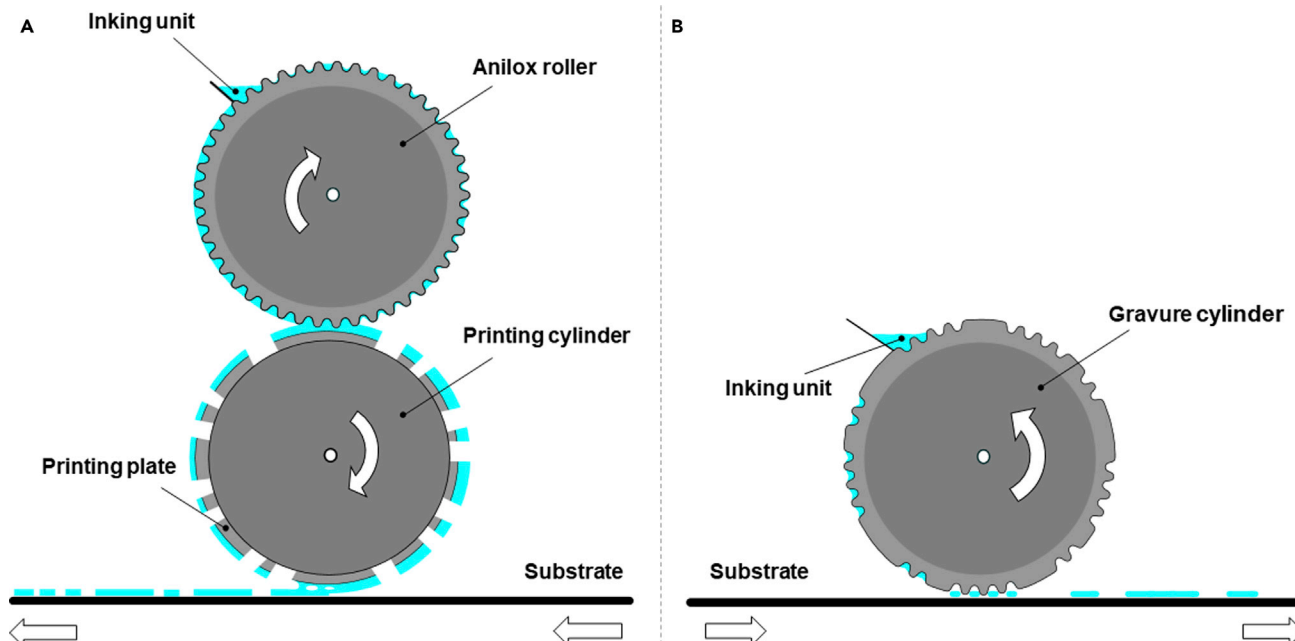
Figure 4. Schematics
(A) Flexographic printing technique.
(B) Gravure printing technique.

MATERIALS

Materials selection plays a decisive factor in the design of wearable piezoelectric devices for health-care applications. A seamless, non-invasive, durable interface must be ensured before the device can be placed on human skin. The main components of multifunctional, flexible, and stretchable sensing devices have already been presented in the introduction. In this section, materials for flexible piezoelectric devices will be overviewed, including flexible substrates, the use of piezoelectric materials as an active sensing material, and conductive flexible electrodes. In addition, the strategies for battery-free, human motion energy harvesting wearable devices will be discussed. Therefore, particular attention will be given to the interaction between each different component and how these affect the performance of the final device needs particular attention.

Substrates

In the last three decades, in electronic device manufacturing many rigid materials such as silicon, silica (SiO_2) and glass have been used as substrates (Lou et al., 2020). In the case of medical applications, the

Table 1. Comparison of printing fabrication techniques for medical applications

Fabrication technique	Screen printing	Inkjet printing	Gravure printing	Flexography printing
Form	Stencil/R2R	Digital (DoD/CIJ)	R2R	R2R
Ink viscosity (mPa·s)	500–5000	10–20	10–100	<500
Printing resolution (μm)	30–100	15–100	50–200	30–80
Line width (μm)	50–100	10–50	10–100	10–100
Line thickness (μm)	5–250	1–10	<1	<1
Printing speed (m/min)	70	1	1000	1000
Process mode (step)	Multiple	Single	Multiple	Multiple
Flexible substrate	Yes	Yes	Yes	Yes
Material wastage	Yes	No	Yes	Yes

selection of the most suitable substrate plays a key role toward to achieving flexibility and stretchability. More specifically, the selected substrate must meet all the physical, thermal, mechanical, and optical criteria of the device. Skin-like conformability, stretchability, ease of manufacturing, dimensional stability, heat resistance, and gas-impermeability are considered as the most important requirements for a flexible substrate. Therefore, in order to achieve such a goal, polymers, soft silicone elastomers and more ecological (e.g. recyclable) materials such as cellulose paper and natural textiles have been proposed (Kamarudin et al., 2020; Liu et al., 2018). Thermoplastic polymers such as PET (Emamian et al., 2017), polyurethane (PU) (Nayak et al., 2013) and polycarbonate (PC) (Babu and de With, 2014) are popular substrates due to their optical transparency and good deformability. Another polymer that is widely used is polydimethyloxane (PDMS); this is a silicone-based non-toxic, non-flammable, biocompatible organic material with excellent elasticity (up to 1000% elongation) (Sappati and Bhadra, 2018, 2020). These characteristics make it a strong candidate for wearable medical devices development. Furthermore, polyethylene naphthalate (PEN) (Khan et al., 2015), polyimide (PI) (Yoon et al., 2016), polyvinyl alcohol (PVA) (Park et al., 2016) and some trademarked elastomeric materials such as ExoFlex, Silbione and DragonSkin have been reported in the literature as potential flexible substrates (Khan et al., 2015; Yu et al., 2020, p. 3). The silicone-based elastomeric materials are able to conform on different surfaces with varied geometries and textures. In addition, they are generally biocompatible, chemically inert, and present a high degree of deformability.

In addition to synthetic substrates, natural materials are also of interest for wearable, flexible systems. This category of materials includes fibers and textiles derived from silk or cotton. These environmentally friendly substrates are promising since they are inexpensive, exhibit desirable characteristics such as biodegradability, good biocompatibility, sustainability and versatility, and satisfy most of the mechanical requirements of irregular deformation (Kamarudin et al., 2020; Wang et al., 2017b). Physiological monitoring devices using paper substrates have also been developed (Chen et al., 2018; Yao et al., 2017), which are of significant interest due to their recyclability, abundance (derived from renewable resources) and mechanical flexibility (Kamarudin et al., 2020).

Sensing materials

The most important component of a functional flexible wearable system is its active functional (sensing, actuating and harvesting) element. Physical data detection depends on wearable sensors force-triggered changes in the electrical parameters such as piezoelectricity (voltage or strain induced through applied mechanical or electrical stress), piezoresistivity (change in resistance with mechanical stress) (Lee et al., 2015), piezocapacitance (change in capacitance with mechanical stress) (Nur et al., 2018) and triboelectricity (contact electrification) (Meng et al., 2019). Mechanical deformation such as stretching, bending, pressing and twisting lead to changes in the electrical parameters of device sensing components. The cross-sectional area of the device is changed by such mechanical deformation, altering the distance between the conductive electrodes and the active sensing components of the device. This enables external monitoring of changes in resistivity, capacitance or generated voltage, for example, and creates a mechanically sensitive material. Structural design to further tailor the performance is also important. In this review, we have focused on ferroelectric-based wearable medical devices since these sensing materials are able to detect multiple stimuli, such mechanical stresses and pressures due to their piezoelectric properties, as well as temperature fluctuations due to pyroelectric effects.

Piezoelectric materials

Piezoelectricity was first discovered by the Curie brothers in 1880 and describes the change in the electric polarization of a material in response to an applied mechanical stress or strain (Elahi et al., 2018). The generated voltage can be used to drive a current in an external circuit. Therefore, piezoelectric materials can be both used for sensing and energy harvesting applications (Roscow et al., 2015; Xie et al., 2019). This is known as the direct piezoelectric effect and is described by the following equation:

$$D_i = d_{ij}\sigma_j + \varepsilon_{ik}E_k \tag{Equation 1}$$

where D_i is the dielectric displacement in response to the applied stress, σ_j, under short circuit conditions, d_{ij} is the piezoelectric strain coefficient, ε_{ik} is the dielectric permittivity, and E_k is the electric field. The subscripts i, j, and k refer to the different directions within the piezoelectric materials using matrix notation. The piezoelectric strain coefficient is defined by two subscripts. The first subscript denotes the direction of polarization while the second subscript refers to the applied or induced mechanical strain (Soin et al., 2016). A piezoelectric sensor converts mechanical loads into electrical charge and its performance is directly related

to the piezoelectric charge coefficient, d_{ij}, and piezoelectric voltage coefficient, g_{ij}. The crystallographic orientation of a piezoelectric crystal, or the polarization direction in the case of a ferroelectric ceramic, defines one of its three main operational modes for external stimuli detection: longitudinal (d_{33}), transverse (d_{31}), and shear (d_{15}). For instance, the electric charge which is produced by an applied strain on the 3-direction is given by the Equation (2):

$$Q_3 = d_{33}F_3 \qquad \text{(Equation 2)}$$

where the d_{33} is the piezoelectric charge coefficient and F_3 is the applied force in the same direction. In sensing applications, the open-circuit voltage coefficient, g_{ij}, is of importance too, which determines the voltage sensitivity of a material in response to an applied stress. The following Equation (3) expresses the relationship between the piezoelectric voltage coefficient and the piezoelectric strain coefficient, where $\varepsilon_{33}^{\sigma}$ is the dielectric permittivity at constant stress:

$$g_{ij} = \frac{d_{ij}}{\varepsilon_{33}^{\sigma}} \qquad \text{(Equation 3)}$$

Piezoelectricity is the main principle of the piezoelectric type pressure/strain sensing materials. The piezoelectric electromechanical interaction can occur in a specific class of material. This kind of material can be divided in non-centrosymmetric crystal structure inorganic materials and in semi-crystalline polymeric materials. The former class includes ferroelectric materials such as lead titanate ($PbTiO_3$), lead zirconate titanate ($PbZrTiO_3$) (Dagdeviren et al, 2014, 2015) and modified lead zirconate titanate ($PbZr_xTi_{1-x}O_3$) (Winestook et al., 2019), barium titanate ($BaTiO_3$) (Guan et al., 2020), strontium bismuth titanate ($SrBi_4Ti_4O_{15}$), potassium sodium niobate (Bairagi and Ali, 2020), and potassium niobate ($KNbO_3$) (Yang et al., 2012). On the other hand, piezoelectric polymers for pressure/strain sensing consist of polypropylene ferroelectret (PPFE) (Wu et al., 2015), where dipoles originate from electrically charged pores, and ferroelectric polymers such as polyvinylidene fluoride PVDF (Chiu et al., 2013) and P(VDF-TrFE) (Chen et al, 2015). However, not all of the above materials can be used with the same ease of manufacturing. Inorganic materials are usually brittle, less flexible, require high processing temperatures, and are more expensive than piezoelectric polymers, while lead zirconate titanate (PZT) and its associated materials, even if they represent excellent piezoelectric performance, are toxic and harmful for their surrounding environment (Qian et al., 2020). However, while ferroelectric polymers are inherently flexible, their low d_{ij} piezoelectric coefficients can limit performance, although the low permittivity of polymer can lead to high g_{ij} coefficients, see Equation 3. Researchers have attempted to improve biocompatibility of inorganic materials (Kim et al., 2017a) and piezoelectric performance of organic materials (Yuan et al., 2020) by integrating different materials in a system and purposing new structures.

A game-changer in the fabrication of sensing elements based on piezo-composite materials was solution-processable composites. They have recently gained considerable interest from scientists, engineers, and medical practitioners since composite materials can overcome the barriers of both organic and inorganic materials as they combine their different advantages providing enhanced actuation and sensing performance. The flexibility and the stretchability as well as the piezoelectric performance of the polymer composites can be improved by choosing the most suitable polymer matrix and the piezoelectric reinforcing phase, respectively (Mahanty et al., 2020; Qian et al., 2020; Wang et al., 2017a; Yang et al., 2020b). The working principle of a wearable piezoelectric device is presented in Figure 5. Under normal conditions, the piezoelectric-based device is electrically neutral and consequently, there is no electrical flow in the circuit. When an external force is applied in one direction, the piezoelectric layer undergoes a change in electrical polarization and positive and negative charges accumulate on surfaces to balance this change in polarization. When the external force is released, an opposite current is generated and the active sensing layer returns to its original state. This reciprocating motion alternately changes the charge distribution and force electrons to flow back and forth, forming a continuous output AC signal (Qian et al., 2020).

Flexible electrodes

Another key component of a dynamic, large area, deformable, and wearable sensing device is the creation of flexible thin film electrodes. An ideal flexible conducting material should exhibit high electrical conductivity and robustness even under extreme and complex mechanical deformation conditions. Furthermore, mechanical flexibility and stability are prioritized characteristics for research and development. To achieve such a flexible electrode material with high stretchability, low processing temperature and low resistance, a variety of conductive materials and processing technologies have been studied. Different conventional
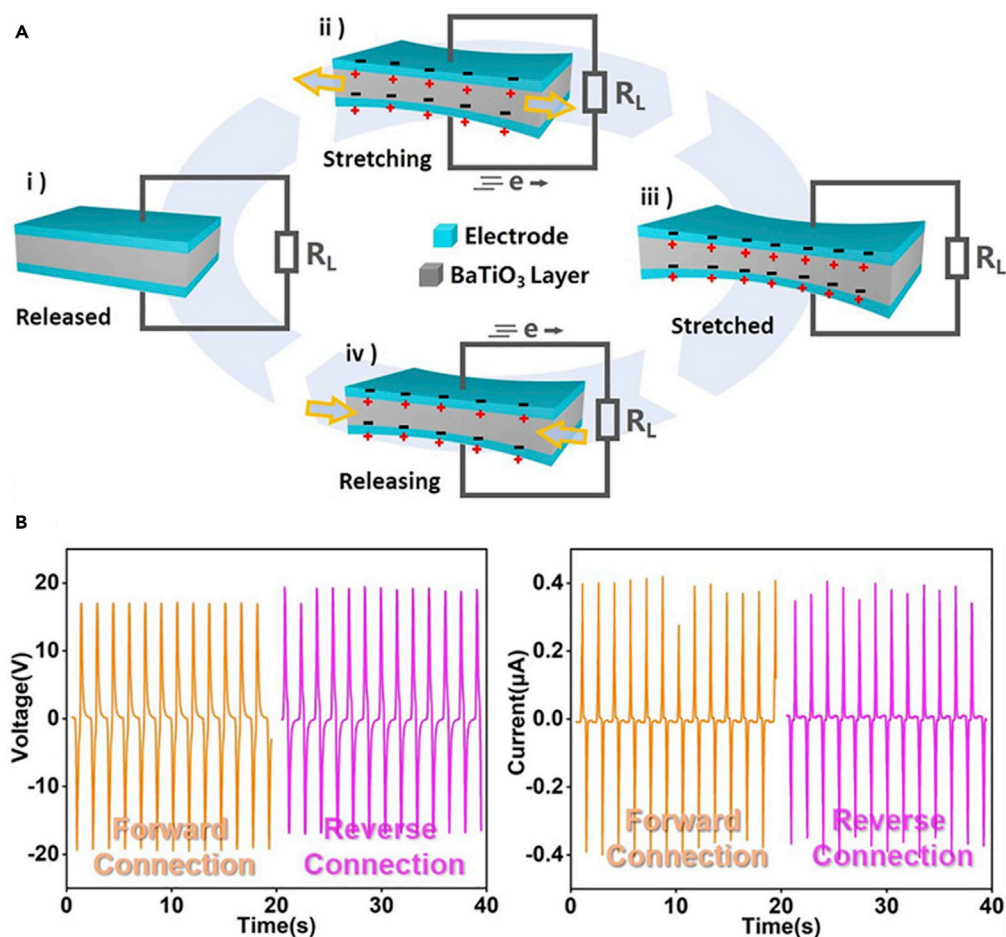
Figure 5. The working principle of a wearable piezoelectric device
(A) Charge distribution of ferroelectric barium titanate based composite in four different working situations: (i) released, (ii) stretching, (iii) stretched and (iv) releasing.
(B) The max value of V_{oc} and I_{sc} during working conditions.
[Copyright 2020, Elsevier] (Qian et al., 2020).

inorganic materials such as platinum (Pt) (Dagdeviren et al., 2014), gold (Au) (Gonçalves et al., 2019), aluminum (Al) (Zhao et al., 2020), indium tin oxide (Luo et al., 2017), silver (Ag) (Yoon et al., 2020) and copper (Cu) (Rajala et al., 2017) in a variety of forms such as films, nanoparticles, nanowires or flakes, as well as some organic materials, such as CNTs and graphene, are generally considered as the most attractive options for developing conductive, flexible electrodes and interconnection elements. Inorganic materials are usually rigid which limits their use in wearable flexible systems. Their performance can be optimized when they are in nanotube, nanowire and nanoparticle form as they are more transparent, electrically stable, durable, and flexible. For instance, Choi et al. developed a highly conductive, biocompatible, and flexible nanocomposite using integrated poly(styrene-butadiene-styrene) elastomer with long Ag-Au nanowires (Choi et al., 2018). Carbon-based materials, such as CNTs and graphene, are a common choice for use as electrodes as they exhibit excellent sensitivity, mechanical, and electrical properties (Lou et al., 2020).

Power harvesting and storage

Self-powered piezoelectric flexible devices do not require an external source of power to operate their sensing activity or process and transmit the produced data, eliminating the need for battery usage, which is currently one of the key limiting factors in the wearables industry. Devices such as these can act as both a sensor and a power generator. Mechanical energy from physical motion can be converted into electrical energy by soft, flexible piezoelectric harvesters. The harvested power from these devices can be stored

in a capacitor or a flexible battery and further used for device operation. The advantage of not having to use a battery is the miniaturization of device, and not needing to recharge or replace the battery, thereby providing autonomous operation (Jin et al., 2017). A variety of figures of merit (FOMs) have been generated to evaluate the energy conversion efficiency of a piezoelectric. A dimensionless FOM has been proposed for piezoelectric materials in energy harvesting devices at on-resonance and off-resonance cases.

$$FOM_{on-resonance} = \left(\frac{k_{31} \; Q_m}{S_{11}^E} \right) \qquad \text{(Equation 4)}$$

$$FOM_{off-resonance} = \left(\frac{d_{31} \; g_{31}}{\tan \delta} \right) \qquad \text{(Equation 5)}$$

Where k_{31} is the transversal electromechanical coupling factor, Q_m is the mechanical quality factor, S_{11}^E is the elastic compliance at the constant electric field, d_{31} is the transversal piezoelectric strain constant, g_{31} is the transversal piezoelectric voltage constant, and $\tan \delta$ is the loss factor (Xu, 2012). Both sensing and harvesting FOMs can be improved through forming composite materials. The material property requirements for sensing and harvesting are similar, i.e. we typically want a high piezoelectric charge coefficient, d_{ij}, and low permittivity, ε_{33}^σ, to maximize g_{ij} and $d_{ij}g_{ij}$, which is an advantage for multi-functional applications.

Recent research has shown promising results by embedding piezoelectric materials into flexible wearable devices. While ferroelectric and piezoelectric materials such as PZT (Park et al., 2014; Qi et al., 2011), BaTiO$_3$ (Lin et al., 2012), and lead magnesium niobate-lead titanate (PMN-PT) (Xu et al., 2013) are good candidates for energy harvesting, piezoelectrets are also of interest, which derive their piezoelectric properties for charged pores. For example, Wu et al., demonstrated self-powered and wearable cellular polypropylene piezoelectret-based system. Cellular polypropylene piezoelectret, first discovered in 1990 by Kari Kirjavainen et al. (Saengchairat et al., 2017), is flexible, lightweight, inexpensive, and has similar properties to traditional piezoelectric materials. This system can be used simultaneously for biological signal detection (such as coughing and arterial pulse) and energy harvesting from body movements. The flexible generator was able to reach a maximum peak power density of ~52.8 mW/m^2 (Wu et al., 2015). Recently, Rovisco et al. fabricated a self-sustainable multifunctional microstructure composite system made of nanowires, synthesized by a seed-layer free hydrothermal route mixed with PDMS (ZnSnO$_3$@PDMS), which exhibited an output voltage, current, and instantaneous power density of 120 V, 13 µA, and 230 µW, respectively (Rovisco et al., 2020). Yuan et al. tried to improve the piezoelectric coefficient of ferroelectric P(VDF-TrFE) for enhanced mechanical energy harvesting by proposing a copolymer processing method and a flextensional mechanism in a rugby ball configuration. This system exhibited an output voltage of 88.62 V and a current of 353 mA, which were ten times larger than for a single layer P(VDF-TrFE) harvester under the same pressure (Yuan et al., 2020).

MULTIFUNCTIONAL SYSTEMS

Bifunctional systems

This literature review focuses on wearable flexible energy conversion devices that are able to convert mechanical energy into electrical energy. Piezoelectric sensors can detect mechanical signals with varying intensity from different body parts; however, all these signals are either pressure or strain, which limits the functionally of the device. Therefore, in recent years, attempts have been undertaken to improve the performance output of the device. This can be easily achieved by integrating other energy converting and sensing systems into the device. Living in an era where technology is evolving rapidly, it is easily understandable that comfortability and flexibility are no longer sufficient for piezoelectric medical devices. In order to remain attractive and competitive, multifunctionality is a potential solution and will now be described.

Piezoelectric and pyroelectric systems

Temperature is one of the most important factors to monitor in the human body. Through temperature monitoring, different human activities can be detected, and the health condition of a person can be determined. The most common materials for temperature sensing are pyroelectric materials (often based on ferroelectric compositions) and temperature resistive (Trung and Lee, 2016). Temperature-sensing devices can provide valuable experimental data in many areas of medicine, such as pneumological and cardiovascular disease monitoring. As previously mentioned, piezoelectricity and pyroelectricity are both present in

poled ferroelectric materials. Therefore, this category of materials is an ideal choice for simultaneous thermal and mechanical sensing. In a pyroelectric sensing element, variations in temperature change the spontaneous polarization of the pyroelectric material, which results in a short-circuit flow of current; this is analogous to piezoelectric materials whose polarization changes with stress. The polarization level in the pyroelectric material decreases with increasing temperature and vice versa. The thermal fluctuations lead to a change in dipole orientation, which further leads in the change in the level of bound charges on the material surface. The Curie temperature is the upper limit of a ferroelectric-based pyroelectric material since above this temperature the spontaneous polarization is reduced to zero. However, this is often not an issue for a wearable medical device as their operating temperature is in the vicinity of the human BT ($\sim$37°C); nevertheless, there can be potential issues with regard to washing or sterilization at more elevated temperatures.

The most widely used pyroelectric materials can be categorized into three types: (i) monocrystalline materials which include deuterium triglyceride sulfate, triglyceride sulfateand potassium tantalum niobate, (ii) inorganic ferroelectric and piezoelectric ceramic materials such as PZT, PMN, barium strontium titanate, ZnO, and $BaTiO_3$, and lastly (iii) polymeric (ferroelectric PVDF and its copolymers are mostly used) and polymer-based composite materials (PVDF/ZnO, PVDF/PT, PVDF/PZT, PMN/PT, etc.) (Li et al., 2020c). Viola et al. developed an ultrathin (1.2 μm) comfortable and flexible multimodal temperature and force sensor by coupling charge-modulated organic field-effect transistor with a pyro-/piezo-electric PVDF sensing element. The sensing area was separated by introducing a floating gate organic thin film transistor in a bottom-gate/bottom-contact configuration. Moreover, it was possible to discriminate between simultaneously applied temperature and pressure stimuli by coupling sensing surfaces made of poled and unpoled spin-coated PVDF-TrFE), see Figure 6 (a) (Viola et al., 2018). Chen et al. proposed the development of the first skin-attachable inorganic materials materials-based flexible hybrid generator using micropatterned ferroelectric single-crystal $(1-x)Pb(Mg,Nb)O_3$-$xPbTiO_3$ (PMN-PT) ribbons. Single-crystal PMN-PT/SU8 (photoresist polymer) was able to scavenge mechanical and thermal energy from human related multimodal stimuli (pressure, strain, temperature, sound, light, and illumination) (Chen et al., 2017b). Recently, Li et al. demonstrated a 3D multilayer sandwich assembly of flexible organic polypyrrole (PPy) electrodes, electrospun PVDF nano-fibers and electrosprayed $CsPbBr_3$/PVDF beads with good output performance and high sensitivity. Piezoelectric characterization demonstrated the multilayer assembly presented an open-circuit voltage of 10.3 V and a short-circuit current density of 1.29 μA/cm^2 (Li et al., 2020b). Furthermore, a pyroelectric current (100 nA/cm) and output voltage (0.5 V) was obtained upon a temperature rise from 25°C to 35°C.

Piezoelectric and triboelectric systems

Due to their relatively high output and the simplicity of the fabrication processes involved, triboelectric materials have gained much attention in recent years for sensing and energy scavenging (Dharmasena et al., 2018; Dharmasena and Silva, 2019; Patnam et al., 2020). Their performance is based on the triboelectric effect which is caused by the contact electrification when two different materials contact each other and then separate. A variety of inorganic materials such as MoS_2, $MoSe_2$, TiO_2, Si, WS_2, and WSe_2 and organic materials such as silk, cotton, nylon, Kapton, perfluoroalkoxy (PFA), polytetrafluorethylene (PTFE), ethylene-vinyl acetate copolymer, PET, PDMS, and PVDF have been deployed as dissimilar surfaces for the preparation of sensing and energy-generating triboelectric materials (Huang et al., 2020a). The larger electronegative affinity difference between the two different surfaces contributes to higher output performance. Among these, organic materials are usually preferred for the development of wearable piezo/triboelectric generators and are placed as a friction layer on the human skin (Guo et al., 2018; Yu et al., 2019). Human skin can also act as a negative triboelectric layer (Cao et al., 2018), offering the potential to fully integrate devices onto the human body.

The main obstacle of triboelectric materials is their low output current. However, by hybridizing triboelectric with piezoelectric materials, their output performance can be enhanced. For instance, a hybrid device combining these two mechanisms can generate electrical current from mechanical energy either by individual piezoelectric and triboelectric effects or from both at the same time as synergetic effect. Patnam et al. recently developed a hybrid nanogenerator composite film based on the synergetic piezoelectric/triboelectric effect by incorporating calcium-doped barium zirconate titanate (Patnam et al., 2020) (Ca-BZT) into PDMS aiming to harvest energy from mechanical motion. This device exhibited a maximum electrical output performance of 550 V, short-circuit current of 34 μA and power density of 23.6 W/m^2. More
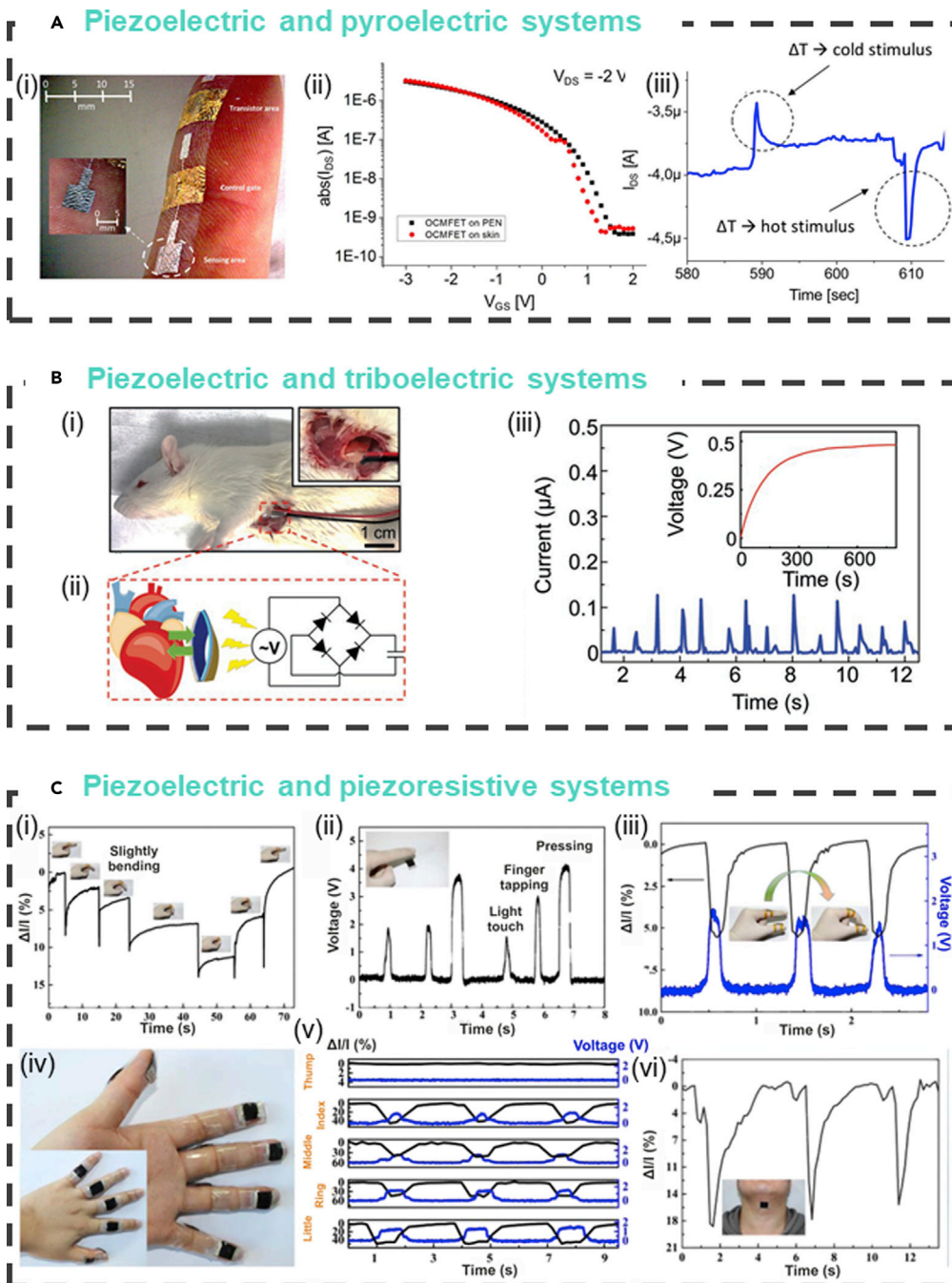
Figure 6. Multifunctional wearable flexible devices for healthcare medical application

(A) (i) The developed charge-modulated organic field-effect transistor placed on human skin; (ii) Its electrical characterization before being peeled-off from the PEN carrier and after its placement onto the skin; and (iii) the temperature sensing preliminary results. [Copyright 2018, Nature Research] (Viola et al., 2018).

(B) (i) Images of a tribo/piezo sensor located in the subdermal chest region of a rat, as well as (ii) the schematic of the energy harvesting process and the circuit, and (iii) the generated electrical current signal obtained from the beating heart. [Copyright 2020, John Wiley and Sons] (Huang et al., 2020b).

(C) Detection of different human motions and flexural measurement using the flexible multifunctional PAN-C/BTO-based sensor in different configurations. (i) A sensor attached to the knuckle where it can detect different bending degrees of the finger. (ii) A self-powered tactile sensor which can detect different contact forces. (iii) Through the combination of two working modes, a sensor on the knuckle (flex sensing mode) and on the fingertip (self-powered pressure sensing mode)

Figure 6. Continued
express a pinching motion. (iv) A smart sensing system that was proposed by integrating two sensors on each finger where the signal could be used to express different hand gesture as shown on image (v). (vi) A sensor designed to detect a swallowing action.
[Copyright 2018, American Chemical Society] (Zhao et al., 2018)

recently, Huang et al. presented a flexible tribo-/piezo-electric nanogenerator made of genetically engineered recombinant triboelectric spider silk protein and a ferroelectric PVDF layer. Between these two different materials, a PET layer was introduced which acted as the electrification layer. The device exhibited outstanding output performance and energy transformation efficiency (open circuit voltage $V_{OC} \approx 300\ V$, short circuit current $I_{SC} \approx 72\ \mu A$, energy conversion efficiency = 50.9%), see Figure 6 (b) (Huang et al., 2020b).

Piezoelectric and piezoresistive systems

In addition to piezoelectricity, other mechanisms such piezoresistivity and piezocapacitance can be used for pressure and strain sensing. When a pressure is applied to a piezoresistive sensing material, its resistance changes in response to that stimuli. This category of active sensing materials is also promising for health monitoring and attached on skin applications due to their ease of manufacturing, low energy consumption (Sosa et al., 2015) (typically current consumption between 1 mA and 50 mA) and their broad range of pressure detection (Chen et al., 2019) (e.g. 259.32 1/kPa in the range of 0 - 2.5 kPa). Piezoelectric and piezoresistive materials can be combined into a wearable system, creating a dual-function sensing device with improved performance. The piezoresistive materials that have mainly been used in the literature are elastomeric-based (such as PU and PDMS) composite materials with conductive fillers (such as CNTs and reduced graphene oxide, R-GO) as a reinforcing phase. However, compared to piezoelectrics, these materials are not able to easily detect low-pressure stimulus in medical electronics applications (Pramanik and Saha, 2006). In order to overcome these limitations and increase their sensing performance, microstructured and porous-structured piezoresistive materials have been proposed (Jung et al., 2014; Li et al., 2019). Zhao et al. developed a multifunctional sensor made of carbonized electrospun polyacrylonitrile/barium titanate (PAN-C/BTO) nanofiber film. In this work, piezoelectric, piezoresistive, and triboelectric effects were combined in a device introducing a dual-function mechanical sensor for swallowing, finger flexure, finger tapping, and walking gait detection. The fabricated sensor could independently and simultaneously detect pressure and curvature via the piezoelectric $BaTiO_3$ nanoparticles and the impedance change of the conductive nanofibers, see Figure 6 (c) (Zhao et al., 2018).

Trifunctional systems

All the abovementioned mechanisms can be combined, either all together or integrated with other functionalities, such as magnetic, electrostatic and photoelectric mechanisms, to create a multifunctional energy generating devices. For example, Yoon et al. demonstrated a highly flexible and comfortable stress-monitoring multilayer patch that integrated three different sensors, which included skin temperature, skin conductance, and pulse rate (HR). The pulse wave sensor was fabricated using a flexible piezoelectric membrane supported by a perforated polyamide membrane. This skin-attachable sensing patch was claimed to be highly responsive and capable of detecting multimodal physiological and emotional signals (Yoon et al., 2016). Lee et al. developed a tri-functional device based on all powder-processing methods by using ZnS powder as a phosphor layer and a PZT as a dielectric layer (Lee et al., 2018a). This device was capable of simultaneously generating light based on the electroluminescent (EL) and sound based on the piezoelectric effect. Moreover, the device was also able to harvest the reverse-piezoelectric energy and generate a piezoelectric-driven EL light when pressure was applied onto its surface. The best recorded luminous efficiency (1.3 lm/W) was achieved at 40 V and 1000 Hz.

Data processing and transmission

A wearable medical device can use a variety of sensors to obtain complex information in the process of physical interaction with the environment. The embedded sensors are typically transducers that generate signals through an energy conversion process. Before exporting the data, the generated signals should be pre-processed by appropriate data processing methods in order to be interpreted as meaningful information for medical monitoring or diagnosis (Servati et al., 2017). During the design of a wearable sensing device, the electric signal type, measurement range and frequency must be considered. The measurement range determines the required resolution of the signal for analysis. In addition, non-invasively obtained signals are often low in intensity and noisy. In order to avoid this problem, an analog front-end controller
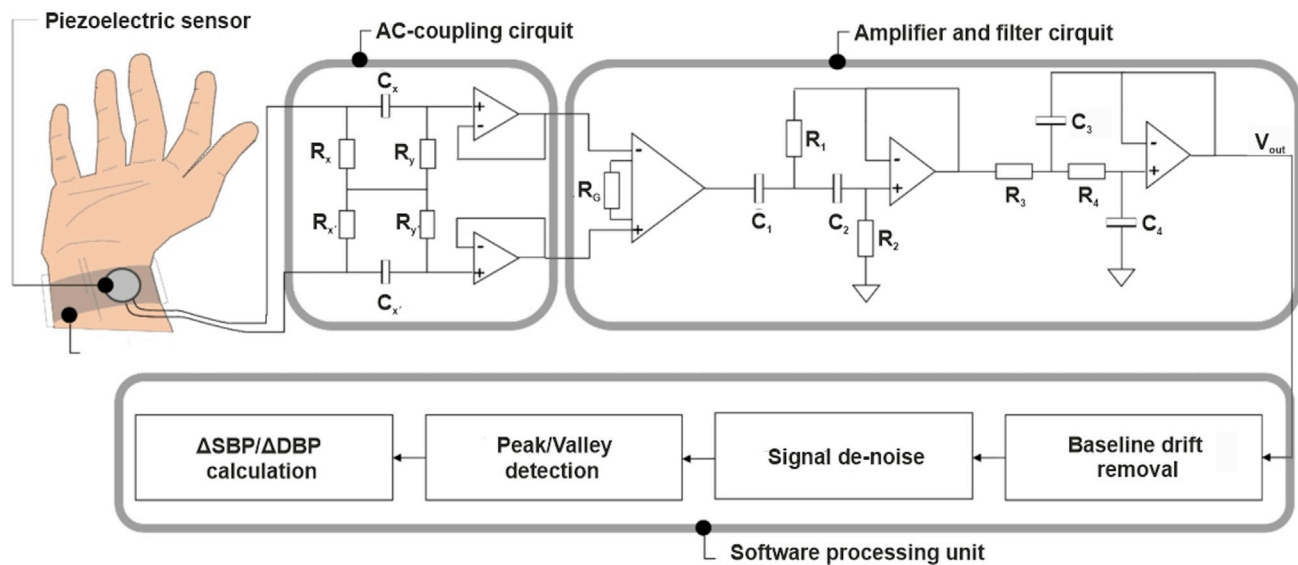
Figure 7. Schematic of the process of data-relay from a wearable piezoelectric device
[Copyright 2020, MDPI] (Wang and Lin, 2020)

(AFEC) is used to reduce the noise and to increase the power of the signal at levels that can be further processed. The active sensing materials inside the device are usually connected to an AFEC in which the electric signals are filtered and amplified. Afterward, an analog-to-digital converter (ADC) is used to convert the signal. When the signal is finally converted to digital, it is read and processed by a microcontroller or a MPU (Khan et al., 2016). To demonstrate clearly how a wireless wearable piezoelectric device works, a schematic representation of a system for continuous beat-to-beat systolic blood pressure (SBP) and diastolic blood pressure (DBP) signals processing is shown in Figure 7. This system consists of three main parts: (i) a piezo-electric sensor, (ii) a front-end analog circuit, and (iii) the software processing unit. The piezoelectric sensor converts pressure from pulsation variations of the radial artery into electrical signals. The generated signals amplified and filtered by the front-end circuit and finally, the post-processing unit is responsible for detecting SBP and DBP feature points and calculating pressure changes between the adjacent waves (Wang and Lin, 2020).

In recent years, new wireless technologies including Bluetooth (power consumption = 1 - 100 mW, range <50 m), NFC (Near Field Communication) (very low power consumption, range <20 cm), Zigbee (very low power consumption, range = 10-75 m) and WiFi (power consumption = 60 - 70 mW, range <100 m) have been developed for data transmission (Liang and Yuan, 2016; Song et al., 2019; Tian et al., 2019a, 2019b). After the processing of the detected signals, the data are transferred from the embedded sensor node to the monitoring unit and the communication network. The selection of the wireless communication network depends on many factors such as power consumption, the cost of set-up, and the range of the transmitted signal, for example. Bluetooth seems to be the most reasonable choice as it is low cost, does not need complex hardware, and is widely compatible with other systems. At the final stage, the data are transmitted wirelessly from the wearable sensing device to smartphones, laptops, or any other consumer electronic device for data interpretation and visualization. Mokhtari and Ahmad demonstrated a Bluetooth-based portable piezoelectric sensor for continuous monitoring of cardiac activity. The low energy Bluetooth sensor was able to transmit the collected data in real-time and send it to a laptop (Mokhtari and Al Ahmad, 2019).

BIOMEDICAL APPLICATIONS AND HEALTH-CARE MONITORING

Pressure and strain sensors for vital signs monitoring

Monitoring the vital signs from a human body is the underlying motivation of wearable electronics for health-care applications. BT, HR, respiration rate (RR), and blood pressure (BP) are routinely monitored by health professionals and are characterized as the four primary vital signs. These measurements can be
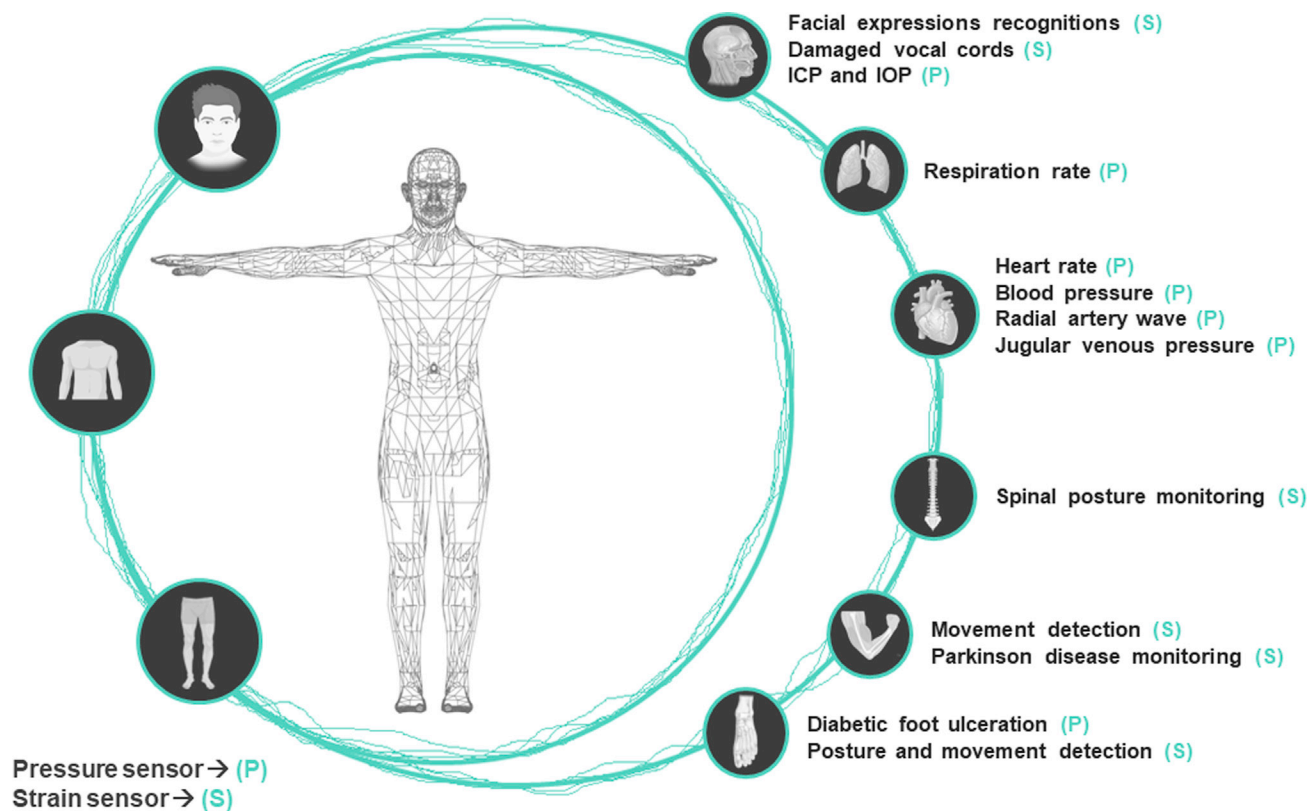
Figure 8. Wearable flexible piezoelectric devices for health monitoring applications

useful as they show a first image of a patient's general health and give clues about possible diseases. Physical sensing platforms are used to measure a range of physical responses such as tensile strain, pressure, temperature, and humidity at the surface of the body (Rosato et al., 2004). A human body can produce pressures ranging from less than 10 kPa to more than 100 kPa (Mannsfeld et al., 2010). Physical activity in human body parts produces different amounts of pressure with highest pressures located in the feet, due to body weight and movement. BP, RR, heart rate, radial artery wave, jugular venous, and vocal cords usually produce medium-range pressures. Intrabody pressures like intracranial pressure and intraocular pressures exhibit values lower than 10 kPa. The monitoring and detection of all above mentioned human body produced pressures are important in the diagnosis and prevention of serious medical diseases such as heart failure, respiratory disorders, cardiovascular disease, damaged vocal cords, sports-related injuries, sleep apnea-hypopnea syndrome, and diabetic foot ulceration (Trung and Lee, 2016). All the mentioned pressure signals are grouped by sensing location and are presented on Figure 8.

Wearable strain sensors can be attached directly on human skin or in clothing for human motion monitoring in medicine. The strain level of skin at various positions on the body relates to different physiological signs. Detectable human motions can be categorized in large scale and small-scale motions. The former consists of leg, spine, arm and hand bending movements while the latter include the subtle movements of the chest, face, and neck during swallowing, breathing and speaking. This category of wearable sensing devices are of interest as they can be used to diagnose and monitor respiratory disorders, damaged vocal cords, Parkinson's disease, posture and movement, facial expression, the degree of change of spinal posture and skin sclerosis (Trung and Lee, 2016). For further clarification, Figure 8 is a schematic representation of body parts where strain sensors can be attached. Such flexible devices that can be worn on the skin and detect pressure and strain changes have gained significant attention in the recent years and are of importance in the healthcare industry. In this section, the latest research into wearable piezoelectric health-care systems that have been used for real-time diagnosis and treatment of patients are introduced.

Heart rate

In a living human, the heart pumps oxygenated blood and other nutrients to the body through arteries and removes carbon dioxide through the lungs in a repetitive circular process known as the cardiac cycle. The heart rate, or pulse, is the frequency of the cardiac cycle measured by the number of contractions (beats) of the heart per minute (bpm). The heart rate varies according to any change in a person's physical or mental state (Achten and Jeukendrup, 2003). Heart rate analysis is a valuable non-invasive method for detecting early evidence of cardiovascular events such as bradycardia (rapid decrease in heart rate) and tachycardia (rapid increase in heart rate) (Golzar et al., 2017). Due to the fact that these events occur suddenly, pressure and strain sensing wearable devices can be used for a long period of time in order to measure the heart rate by detecting the systolic peaks from the radial artery at the wrist or from the carotid artery at the neck (Maity et al., 2020; Park et al., 2017; Wang et al., 2017a; Yi et al., 2020). For example, Chun et al. fabricated a self-powered mechanoreceptor incorporating an artificial ion-channel system and piezoelectric films in the same device which was able to identify the characteristics of radial artery pressure waveform; see Figure 9 (a) (Chun et al., 2018). Yang et al. recently proposed a three-dimensional hierarchically interlocked piezo-electric PVDF/ZnO nanofiber-based sensor through epitaxial growth of piezoelectric ZnO nanorods on the surface of electrospun PVDF nanofibers, ensuring good flexibility and high gas permeability. This sensor exhibited high sensing performance in both bending and pressing mode and could be used for the detection of the complex subtle physiological signals of wrist pulse, respiration and muscle behavior (Yang et al., 2020a).

Respiration rate

The respiration cycle is a critical physiological task in humans because without sufficient oxygen inhalation into the lungs and carbon dioxide removal through nose or mouth, patients are at risk of permanent injury or even death (AL-Khalidi et al., 2011). A healthy adult at rest usually breathes with a rate of 12-20 breaths per minute (Güder et al., 2016). Through the monitoring of the alternations in RR, serious clinical events such as cardiac arrests can be predicted and prevented (Cretikos et al., 2007). More specifically, the abnormal RR is usually an important marker of serious lung diseases such as asthma, apnea, dyspnea, tachypnea, hyperpnea, hypopnea, orthopnea, bradypnea, platypnea, chronic obstructive pulmonary disease, Cheyne–Stokes respiration, Kussmaul breathing, and Biot's respiration, among others (Atalay et al., 2015). Wearable respiration sensing devices are usually mounted on the chest and abdomen region to respond to the expansion and contraction of the diaphragm (Abu-Khalaf et al., 2018; Chiu et al., 2013; Mahbub et al., 2017; Xin et al., 2014). Lei et al. proposed a small size, light weight, easy to use, low cost, and portable PVDF-based sensor patch for respiration detections in both static (sitting) and dynamic (walking) condition. The sensor patch produced electrical signals from the periodical deformations for the human chest during the respiratory movements, see Figure 9 (b) (Lei et al., 2015). Pressure sensors can be also applied near to the mouth or nose area to detect the flow of breath with high accuracy (Yaghouby et al., 2016). Liu et al. developed a wearable self-powered PVDF sensor for respiration and health-care monitoring by electro-spinning the piezoelectric active material on a silicon substrate. Due to the periodic inhalation and exhalation, the piezoelectric nanogenerator generated an output open-circuit voltage (up to 1.5 V) and a short-circuit current (up to 400 nA). Apart from the RR, this sensor was also able to detect human gestures and vocal cord vibrations (Liu et al., 2017). However, respiratory sensors placed on the chest region are more comfortable and, as such, for normal activity tracking, where the patient does not suffer from a serious pulmonary disease, they are usually preferred.

Blood pressure

BP is another key parameter in cardiovascular disease monitoring. The cuff sphygmomanometer is one of the oldest and most established diagnostic tools for BP measurement. However, it is not well suited for continuous monitoring because it can only detect two basic pressure values, the (greatest heart contraction) SBP and the (greatest heart dilation) DPB (Dagdeviren et al., 2014). Another technique for BP monitoring is the arterial cannula method. This method uses a catheter which is inserted into the blood vessel and measures the pressure with high accuracy. Nevertheless, such an invasive technique induces potential risk and is uncomfortable for the patient (Lakhal and Robert-Edan, 2017). As a consequence, continuous non-invasive BP monitoring remains an important demand. Dagdeviren et al. developed a BP sensing device by integrating ferroelectric PZT in an array of capacitors on an ultra-conformal elastomeric substrate (Dagdeviren et al., 2014). Pulse pressure measurements in different locations of the body were possible due to the high sensitivity of this device. Recently, Wang and Lin proposed a low-cost wearable
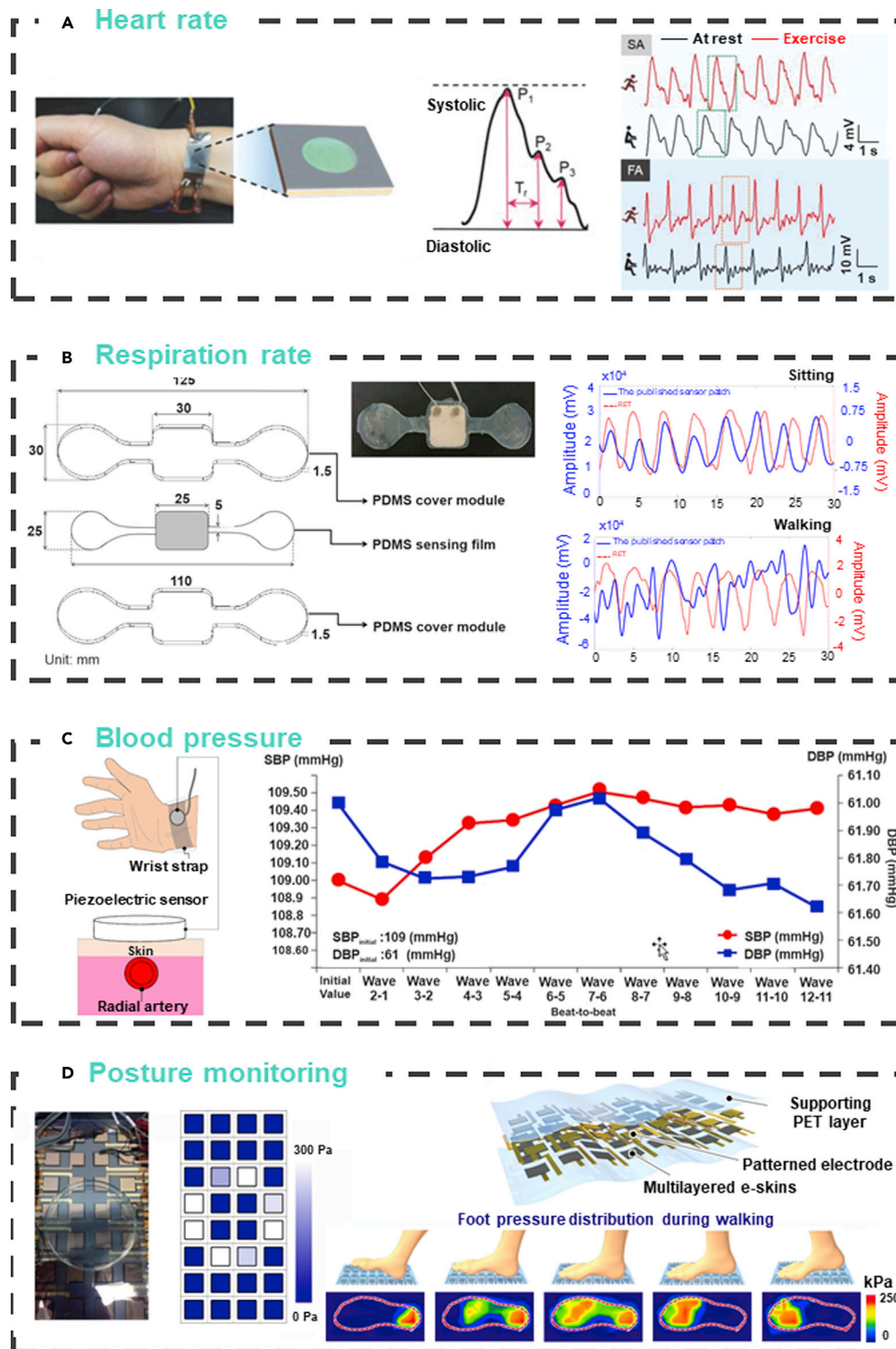
A Heart rate

B Respiration rate

C Blood pressure

D Posture monitoring

Figure 9. Fabricated wearable flexible devices used for medical applications

(A) Photograph of the attachable patch sensor on human wrist to measure the radial artery. Three typical types of pulse waves (P1, P2, and P3) in radial artery measurements and the round-trip time of a reflected wave from the hand periphery are shown, as are characteristic signals before and after exercise. [Copyright 2018, John Wiley and Sons] (Chun et al., 2018).

Figure 9. *Continued*

(B) Schematics and photograph of the sensor patch as well as the comparison of the electrical signals acquired from the proposed sensor patch and a commercial respiratory effort transducer (RET) under static (sitting), and dynamic (walking) conditions. [Copyright 2015, MDPI] (Lei et al., 2015).

(C) Schematic of the sensor measurement location and the beat-to beat blood pressure measurements for 12 beats. [Copyright 2020, MDPI] (Wang and Lin, 2020).

(D) Schematic and photograph of the smart insole consisting of 4 × 8-pixel arrays of multilayer e-skins, patterned electrodes, and supporting PET layers. Schematics of five walking motions on the smart insole to monitor the foot pressure distribution and their corresponding pressure maps. [Copyright 2018, American Chemical Society] (Lee et al., 2018b).

piezoelectric system for continuous beat-to-beat SBP and DBP pressure measurement (Wang and Lin, 2020). This system used a piezoelectric pressure sensor to directly convert the voltage difference between the consecutive systolic and diastolic feature point of pressure pulse wave into pressure difference, see Figure 9 (c).

Posture monitoring

The interaction of the surface of the feet with the environment during locomotion leads to the generation of foot plantar pressure. Problems in the foot area are highly important and its diagnosis at an early stage can prevent serious lower limb problems, such as foot ulceration in diabetic patients (Abdul Razak et al., 2012). In 2017, Cha et al. demonstrated a position monitoring system mounted on the inner side of a patient cloth using unobtrusive piezoelectric sensors (Cha et al., 2017). Flexible piezoelectric sensors were inserted close to the knee and hip region of the patient cloth and produced electrical signals every time that patient's knee and hip was bent. The output data were transferred to a computer via Bluetooth. In the same year, Rajala et al. proposed an in-sole single-axis piezoelectric sensor made of PVDF coated with copper electrodes on both sides for plantar pressure measurement (Rajala et al., 2017). The sensor was capable of detecting pressure up to 486 kPa and was proposed as a method to prevent pressure ulcers. Finally, Lee et al. developed a low consumption flexible ferroelectric sensor with ultrahigh-pressure sensitivity (47.7 1/kPa, 1.3 Pa minimum detection) and linear response over an exceptionally broad pressure range (0.0013–353 kPa) for weak gas flow, acoustic sound, wrist pulse pressure, respiration, and foot pressure detection, see Figure 9 (d) (Lee et al., 2018b).

Other medical applications

Dong et al. reported the development of epitaxially grown single-crystalline ferroelectric barium titanate membranes. Barium titanate is brittle and prone to fracture upon bending. However, these $BaTiO_3$ membranes were able to undergo 180° folding during *in situ* bending tests, thus demonstrating superelastic and ultra-flexible performance. These epitaxial ferroelectric membranes are excellent candidates for use in electronic skin medical applications (Dong et al., 2019). Sun et al. designed, developed, and tested a thin and flexible piezoelectric device which consisted of an aluminum nitride piezoelectric thin film on a compliant PDMS substrate for decoding facial strains and predicting facial kinematics. This device was tested on both patients with amyotrophic lateral sclerosis and healthy individuals. When coupled with appropriate algorithms, the system was able to decode different facial movements which makes this integrated system an ideal candidate for use in both clinical settings as nonverbal communication technology and real-time monitoring of neuromuscular conditions (Sun et al., 2020). Kenet et al. presented a low-cost endoscopy visualization device that aimed to decrease a patient's pain and time lost due to complications experienced during a colonoscopy procedure. The device consisted of a piezoelectric PVDF sensing cable which was able to detect the applied forces during endoscopic looping (looping of the colonoscopy shaft within a patient that leads to stretching of the intestine), bending or compression. The piezoelectric cable was inserted within the working channel of the colonoscope before the colonoscopy procedure. During the procedure, the piezoelectric sensor could detect extreme forces and bending, and thereby send a notification to the connected monitor. When the colonoscope reaches the desired location, the piezoelectric cable can be removed allowing the working channel open for use by other tools (Kenet et al., 2020).

CONCLUSIONS AND FUTURE RESEARCH

The rapid development of material science and engineering is stimulating progress in the development of wearable sensing devices. These devices are expected to have a major impact on the health sector and improving the quality of lives by providing real-time monitoring of serious medical deceases. To meet

the market demands, researchers have placed great effort in researching, developing and designing highly active sensing elements with novel properties. Ferroelectric materials have a bright future in wearable electronics industry due to their inherent multifunctionality (e.g. their excellent piezo-, pyro and dielectric properties) that are used in the many sensing and harvesting applications proposed to date.

This review has covered the recent progress of the state-of-the-art flexible piezoelectric wearable devices for medical applications from a materials, processing, and device development approach. Furthermore, the range of possible wearable and health-care-related applications for piezoelectric energy harvesters were identified and presented. Scalable AM processes, such as printing methods, have shown auspicious results toward low cost and high-performance of wearable piezoelectric devices fabrication. In addition, recent developments in techniques such as 3D printing techniques and nanoimprinting technology could provide additional potential for high resolution, low cost and larger-area fabricating ability and are worthy for study for future wearable medical devices. Moreover, hybrid material combinations are necessary for the acceleration of wearable sensing platforms, such as exploiting the potential pyroelectric properties of ferroelectric materials or combining with other sensing mechanisms.

Despite the promising future of these devices due to the encouraging results have been achieved, further technological advances in power output efficiency and cost-effective manufacturing are necessary for the commercialization of future wearable systems. For example, problems like long-term stability, sensitivity, and biocompatibility must be further improved to meet the standard requirements from diagnostic devices, as defined by regulatory agencies. Lastly, there is also a need to effectively reduce the power consumption of the devices, including data acquisition and transmission, which is critical for long term operations. In recent years, researchers have focused on the development of self-powered sensing devices that can operate continuously without external power supply since they represent a promising candidate to achieve long-term sustainability in wearable sensors. New research directions on the field may focus on the integration of multiple ultrathin components or the integration of components with multifunctional capabilities into a flexible piezoelectric sensing system. Therefore, advanced and sophisticated manufacturing processes as well as the rational selection and the design of sensing materials are of utmost necessity.

AUTHOR CONTRIBUTIONS
Z.M.T. conducted the literature review and wrote the manuscript. H.K., J.I.R, and C.R.B. conceived the idea and discussed and revised the manuscript.

DECLARATION OF INTEREST
The authors declare no competing interests.

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