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Piezoelectric nanogenerators for personalized healthcare

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The development of flexible piezoelectric nanogenerators has experienced rapid progress in the past decade and is serving as the technological foundation of future state-of-the-art personalized healthcare. Due to their highly efficient mechanical-to-electrical energy conversion, easy implementation, and self-powering nature, these devices permit a plethora of innovative healthcare applications in the space of active sensing, electrical stimulation therapy, as well as passive human biomechanical energy harvesting to third party power on-body devices. This article gives a comprehensive review of the piezoelectric nanogenerators for personalized healthcare. After a brief introduction to the fundamental physical science of the piezoelectric effect, material engineering strategies, device structural designs, and human-body centered energy harvesting, sensing, and therapeutics applications are also systematically discussed. In addition, the challenges and opportunities of utilizing piezoelectric nanogenerators for self-powered bioelectronics and personalized healthcare are outlined in detail.

1. Introduction

With the significant progress of sensing and information network technology, the Internet is no longer limited to enabling interconnectability and interoperability on traditional smart devices such as computers and mobile phones. Novel technological platforms can nowadays also extend these interaction capabilities to everyday objects and individuals, and gradually form an Internet of Things (IoT) paradigm covering an ever-growing number of different aspects of human life.[1,2] Alongside the above, the development of emerging nanotechnology and materials science have themselves also promoted the development of soft bioelectronics.[3–16] Combined with the IoT platforms, intelligent wearable devices have been widely employed in health monitoring,[17–21] human-machine fusion,[22–25] artificial intelligence,[26–28] and many other applications.[29–33] This field has emerged as favored by both the scientific and industrial

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communities. More specifically, with the rapid development of 5G technology, wearable bioelectronics can be connected to each other to form sensor networks, thereby providing big data insights for personalized healthcare with profound socioeconomic impact.[34–36] In recent years, with the reform and development of medical care, theragnostic models have gradually shifted from hospital-centric to patient-centric, greatly encouraging the development of wearable bioelectronics for personalized healthcare.[37–44] With the help of popularized smart terminals, customized flexible wearable sensors can be used to monitor physical activities,[45–49] vital signs,[50–57] and even infectious diseases like coronavirus,[58] providing a non-invasive and attractive dynamic health assessment and personalized medical treatment.

Wearable bioelectronics enable the change of the current reactive and disease-centric healthcare system to a personalized model with a focus on disease prevention and health promotion. Among them, wearable sensors, which can capture valuable information from the body and permit human health monitoring, are one of the core components of a fully-fledged Internet of Medical Things (IoMT) system. However, continuous and stable power supply for these wearable devices remains

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a major challenge. Traditionally, the standard powering method relies on batteries, with electronic devices equipped with independent power sources to achieve independent operation.[59–61] One way to effectively address such a challenge is to develop self-powered sensors.[62–66] Since the human body yields enormous quantities of passive biomechanical energy in the forms of physical activity, cardiac/lung motions, blood circulation, and so on,[67–71] converting these energies into electricity could expectantly decouple wearable devices from traditional batteries' limited lifetime and frequent replacements, eventually allowing for self-powered operations. Especially for implanted devices, self-powered devices can effectively prevent secondary surgeries needed to replace power sources, providing tremendous advantages and important application prospects.[72–75] Several technologies can convert these biomechanical energies into electrical energy, including but not limited to the piezoelectric effect,[76–78] triboelectric effect,[79–87] magnetoelastic effect,[88–91] and electromagnetic effect.[92–94] Among them, piezoelectric nanogenerators (PENGs) based on the piezoelectric effect have attracted significant attention since their invention in 2006.[95] Due to the inherent characteristics of piezoelectric materials, PENGs can permit reversible conversion of mechanical energy and electrical energy, allowing high-efficiency electromechanical coupling, a fast response profile and simple engineered structures. PENGs can not only act as energy harvesters,[96–102] but also provide active sensing capabilities including in the form of tactile sensing,[103–113] stress or strain sensing,[114–120] acoustic sensing,[121–126] and so on. When attached to the human body, PENGs can allow the monitoring of weak physiological signals of human activities[127–136] including pulse detection,[137–145] respiratory monitoring,[146–151] tissue elastic modulus calculation,[152,153] blood flow monitoring,[154,155] *etc.* Besides these applications, the generated electricity is also widely used for *in situ* electrical stimulation,[156–160] cell activity modulation,[161–173] tissue regeneration,[174–183] and drug delivery.[184–187] In recent years, with the emergence of new materials and the development of new processing technologies, PENG technology has developed rapidly and made significant progress in the field of energy, sensing, and therapy.

This review intends to highlight and cover recent PENG progress in the aforementioned three fields, with a focus on their biomedical applications within personalized healthcare (Fig. 1).[188–196] To better promote the understanding of PENGs and their applications in healthcare, we begin the review with a brief introduction of the fundamental physical science behind their working modes. Subsequently, a number of typical piezoelectric materials are described, including inorganic materials, organic materials, piezoelectric composites, and natural piezoelectric materials. Research progress of various piezoelectric material-based bioelectronics is further covered along with real-life biomedical applications that include on-body energy harvesting, sensing, and electrical stimulation therapy. In the last section, the challenges and opportunities of utilizing PENGs for personalized healthcare are outlined in detail.

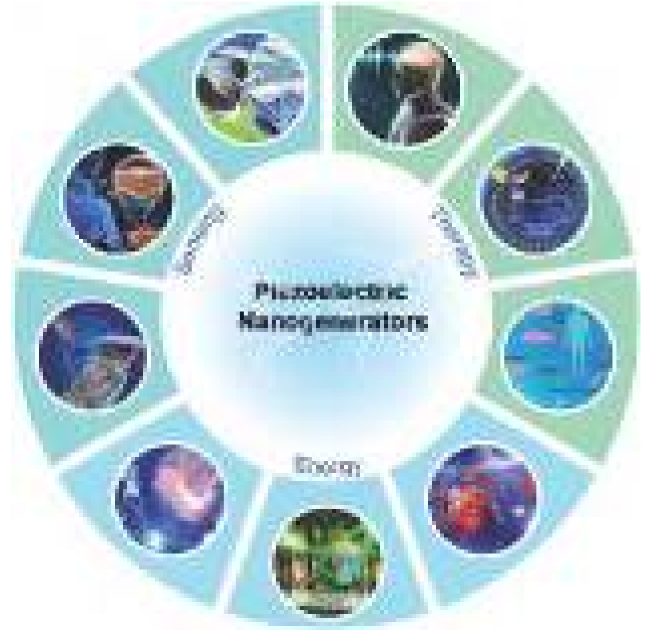

Fig. 1 PENGs for personalized healthcare, including human body associated energy harvesting, self-powered sensing, and electrical stimulation therapy. Reproduced with permission from ref. 188 Copyright 2015, Royal Society of Chemistry. Reproduced with permission from ref. 189 Copyright 2020, Wiley. Reproduced with permission from ref. 190 Copyright 2015, Elsevier. Reproduced with permission from ref. 191 Copyright 2019, Wiley. Reproduced with permission from ref. 192 Copyright 2020, Royal Society of Chemistry. Reproduced with permission from ref. 193 Copyright 2013, Royal Society of Chemistry. Reproduced with permission from ref. 194 Copyright 2019, Elsevier. Reproduced with permission from ref. 195 Copyright 2018, American Chemical Society. Reproduced with permission from ref. 196 Copyright 2018, Royal Society of Chemistry.

2. Fundamental of piezoelectricity

2.1 Piezoelectric effect

The fundamental working principle of PENGs rests on the piezoelectric effect of piezoelectric materials, first discovered by the Curie brothers in 1880.[197] The piezoelectric effect is characterized by a linear relationship between mechanical strain/stress and electrical variables. When some dielectrics are deformed under an external force in a certain direction, the equivalent positive and negative charges appear on two opposing surfaces due to the occurrence of internal polarization. Once the external force is removed, an uncharged state is recovered. This phenomenon is defined as the direct piezoelectric effect. Conversely, when an electric field is applied to the dielectric in the polarization direction, these dielectrics also deform. After the electric field is removed, the deformation of the dielectric disappears. This phenomenon is called the inverse piezoelectric effect. Those materials exhibiting the piezoelectric effect are called piezoelectric materials. Fig. 2 depicts the mechanisms of piezoelectricity (the process to induce polarization) and three typical piezoelectric materials. When the polarization direction of the material (original shape) is along the 3-direction (as shown in Fig. 2a), a direct piezoelectric effect is produced (illustrated in Fig. 2b and c), with

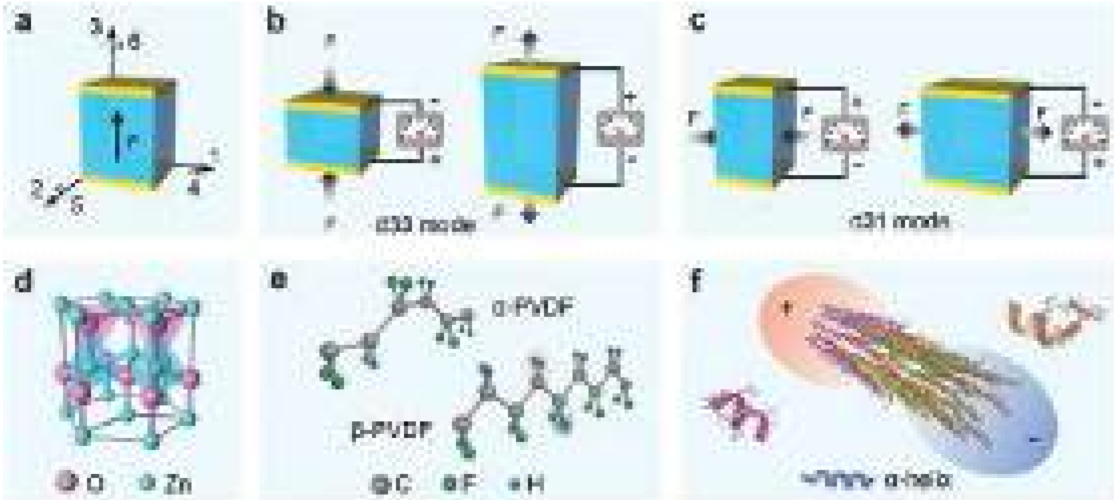

Fig. 2 Piezoelectric materials and piezoelectricity. (a) Direction of forces affecting a piezoelectric element. Illustration of the d_{33} (b) and d_{31} (c) working modes. (d) Wurtzite-structure model of inorganic piezoelectric material ZnO. (e) The α- and β-phase structure of organic piezoelectric polymer PVDF. (f) Natural piezoelectric material of M13 bacteriophage.

current flowing in an external circuit after a force is applied to the piezoelectric material sandwiched between two electrodes (typical structure of PENG). The direction of the applied stress can be either along the polar axis (3-direction) or perpendicular to it (1-direction), resulting in two common PENG configurations, longitudinal mode (d_{33}) and transverse mode (d_{31}). When stress/strain is applied to the piezoelectric material in a parallel manner along the 3-axis, and if the potential generated occurs along the same axis, the design produced is known as d_{33}-mode (Fig. 2b). If the potential generated occurs perpendicular to the direction of applied stress/strain (1-axis), it is called the d_{31}-mode as illustrated in Fig. 2c. In addition, when a piezoelectric material is subjected to a shear force, there is another shear mode (d_{15}) modality known,[198] which can also permit conversion between mechanical deformation and electricity. Relying on the piezoelectric effect, PENGs can effectively convert external strain or stress into electricity, thereby achieving energy harvesting, vital signs sensing, electrical stimulation therapy, and so forth.

2.2 Piezoelectric materials

Piezoelectric materials are materials that display the piezoelectric effect, *i.e.*, the ability to generate electrical charges in response to applied mechanical stress, and *vice versa*. The key feature of piezoelectric materials is the presence of non-coincident positive and negative charge centers (*i.e.*, non-centrosymmetric materials) within the material.[199–201] Depending on the nature of the materials, piezoelectric materials can be roughly divided into four categories: inorganic, organic, composite, and natural piezoelectric materials. Various piezoelectric materials have been developed for PENGs over the past decades, such as zinc oxide (ZnO),[202–209] aluminum nitride (AlN),[210–212] barium titanate ($BaTiO_3$),[213–215] lead zirconate titanate (PZT),[216–218] $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT),[219–221] molybdenum sulfide (MoS_2),[222–228] poly(vinylidene fluoride) (PVDF),[229–233] poly(L-lactic acid) (PLLA),[234–237] bacterial viruses,[238–241] engineered eggshell,[242,243] engineered fish scale[244] and their composites.[245–248] Taking the typical inorganic piezoelectric material ZnO as an example, its wurtzite structure can be described as multiple alternating planes of tetrahedral coordinates of Zn and O stacked along the c-axis, as shown in Fig. 2d.[205] In the absence of external force, the positive charge center is localized on Zn and the negative charge center is localized on O, with both charges balancing each other. When stretched or compressed by external forces, the positive and negative charge centers separate to form a dipole that provides a piezoelectric potential. Indeed, piezoelectricity in organic materials is generated by the process of reorienting molecular dipoles in bulk polymers. PVDF and its derivatives are the most studied organic piezoelectric materials, with these being semi-crystalline polymers synthesized by the polymerization of $H_2C{=}CF_2$ monomers.[249,250] As depicted in Fig. 2e, α and β phases are the most common conformations. The β-phase, with a fully *trans*-planar zig-zag conformation, is responsible for most of the obtained piezoelectric response, due to its polar structure with oriented hydrogen and fluoride unit cells along with the carbon backbone.[251] For natural piezoelectric materials, piezoelectricity mainly originates from the ordered nature of nanocrystalline or liquid-crystalline in biomaterials, due to the internal rotation of atomic groups interrelated with the asymmetric carbon atoms.[252] For example, the M13 phage has a rod-shaped structure composed of α-helical structure, with its dipole moment directed from the amino-terminal to the carboxyl-terminal, as illustrated in Fig. 2f. Because of the lack of reverse symmetry, the film arranged by the M13 phage also shows inherent piezoelectric properties.[253] In addition to the monolithic materials, the piezoelectric composite containing inorganic materials and polymers represents another piezoelectric material of relevance. The composites combine the outstanding mechanical flexibility of organic polymers as well as the high piezoelectric performance of inorganic ceramics, demonstrating an enhanced overall performance. In recent years, significant progress has been made in the preparation of the aforementioned piezoelectric materials as well as their applications in self-powered wearable and implantable biomedical devices.

3. Material engineering and structural design

With the rapid development of wearable technology, wearable bioelectronics face mounting pressure to deliver consistently improving mechanical and electrical performance.[4–7] In order to conformally fit and deform synchronously with the target tissue (*in vitro* or *in vivo*), the engineered devices need optimized flexibility and stretchability, while ensuring good electrical output performance. To achieve these goals, there are two general strategies: material engineering and structural design, as described in Fig. 3.[253–266] This section analyzes the preparation of different piezoelectric materials and their structural designs which are engineered to meet the performance requirements mandated by wearable electronics.

3.1 Inorganic piezoelectric materials

Inorganic piezoelectric materials have attracted great attention in PENGs due to their excellent electrical output profiles. Many inorganic piezoelectric materials have been developed for PENG, such as the aforementioned ZnO,[202–209] AlN,[210–212] $BaTiO_3$,[213–215] PZT,[216–218] PMN-PT,[219–221] MoS_2,[222–226] WSe,[267] SnS,[268] *etc.* However, most of the traditional inorganic piezoelectric materials are bulky and inherently brittle, which restricts their application as wearable bioelectronics. To this end, a great deal of research has been carried out around optimizing the bending and stretching capabilities of PENGs to achieve a high degree of flexibility while maintaining sufficient electromechanical coupling efficiency. The most common strategy is to incorporate the thinned piezoelectric films onto flexible substrates, thereby providing the piezoelectric devices with flexibility. One recent example involves ultra-thin PZT films (500 nm thickness) created on an oxidized silicon wafer through the chemical lift-off method, and subsequently

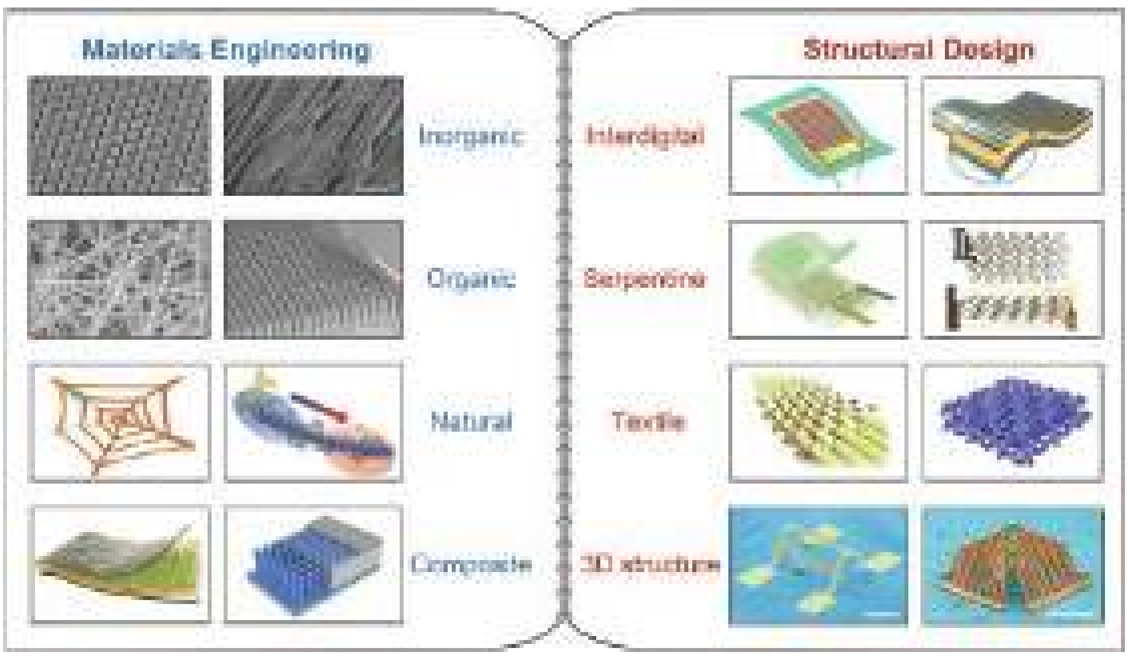

Fig. 3 Materials engineering and structural design for PENGs. Inorganic: SEM image of patterned ZnO nanorods (left). Scale bar, 2 μm. Reproduced with permission from ref. 254 Copyright 2014, Hindawi. SEM image of the assembled Se nanowires (right). Scale bar, 10 μm. Reproduced with permission from ref. 255 Copyright 2019, Elsevier. Organic: SEM image of PVDF nanofibers (left). Scale bar, 5 μm. Reproduced with permission from ref. 256 Copyright 2019, Elsevier. SEM images of the imprinted P(VDF-TrFE) micropillars (right). Scale bar, 30 μm. Reproduced with permission from ref. 257 Copyright 2018, IOP Publishing. Natural: schematic of spider silk structure (left). Reproduced with permission from ref. 258 Copyright 2018, Elsevier. Schematic of a single coat protein (right). Reproduced with permission from ref. 253 Copyright 2012, Nature Publishing Group. Composite: schematic illustration presenting the structure of the PVDF-niobate-based PENG (left). Reproduced with permission from ref. 259 Copyright 2018, American Chemical Society. Schematic diagram represents the structure of P(VDF-TrFE)/BNNTs nanocomposite micropillar-based PENG (right). Reproduced with permission from ref. 260 Copyright 2019, Elsevier. Interdigital: schematic illustration of interdigital PENG (left). Reproduced with permission from ref. 261 Copyright 2016, Wiley. Schematic diagram of PENG with a cross electrode (right). Reproduced with permission from ref. 262 Copyright 2015, Wiley. Serpentine: schematic (left) and photographs (right) of the stretchable PVDF e-tattoo. Reproduced with permission from ref. 263 Copyright 2019, Wiley. Textile: schematic of the PVDF-based textile (left). Reproduced with permission from ref. 264 Copyright 2015, IOP publishing. Schematic of the fabric PENG (right). Reproduced with permission from ref. 265 Copyright 2017, Springer Nature. 3D structure: SEM images of ultralow-stiffness PVDF mesostructure (left) and 3D PVDF mesostructure with electrodes (right). Scale bars, 500 μm. Reproduced with permission from ref. 266 Copyright 2019, Springer Nature.

transferred onto thin flexible polyimide substrates together with the serpentine configurations of metal traces. Such a device was able to yield a low modulus and stretchable mechanics.[152] Furthermore, forming the material into a wavy/bent geometry and combining it with an elastomeric substrate such as polydimethylsiloxane (PDMS) provided a structure that could be stretched to a large degree of strain (∼8%) without fracture.[269,270] In addition to the chemical lift-off method, laser lift-off (LLO) is another effective method to obtain piezoelectric films.[216,271,272] LLO is a dry-type transfer technique that utilizes an excimer laser to vaporize the interface between the piezoelectric thin film and the mother substrate, allowing for the entire piezoelectric thin film to be detached and transferred. Hwang *et al.* fabricated aerosol-deposited PZT film through the LLO method combined interdigital electrode design, the device exhibited good electromechanical performance.[261] The LLO method provides a simple, stable, and large-area transfer of high-quality piezoelectric ceramic films annealed at high temperature, which has the advantages of high efficiency and large-scale production compared to wet-etching processes. Unlike thinning the piezoelectric film, discretizing the piezoelectric film is another effective route to improve the flexibility of the device. This strategy can be used to synthesize films (in the form of nanowires or nanorods) directly onto the target substrate at low temperatures through solution-phase synthesis methods.[273–278] Due to the discontinuity of the nanowire effectively preventing stress concentration during the deformation process, the device displays an overall better flexibility.[279,280] In addition, the nanostructure formed further enhances the effective piezoelectric constants compared to their bulk counterparts due to size effects,[281,282] favoring the application of wearable PENGs. Interestingly, Shan *et al.* recently reported a gravity-driven sintering method to directly fabricate curved and compact piezoceramics by exploiting gravitational force and the high-temperature viscous behavior of the sintered ceramic specimens, which was inspiring in engineering piezoelectric ceramics for wearable applications.[283]

3.2 Organic piezoelectric materials

Among the various materials exhibiting piezoelectricity, polymers have attracted much attention because of their outstanding flexibility, versatile designability, low processing temperature, and ease of processing over inorganic piezoelectric materials, all of which are critical for wearable PENG. There are many piezoelectric polymers used for PENGs, including PVDF,[229–233] PLLA,[234–237] poly(vinylidene fluoride trifluoroethylene) (P(VDF-TrFE)),[284–289] poly(vinylidene fluoride hexafluoropropylene) (PVDF-HFP),[290,291] poly(vinylidene fluoride chlorotrifluoroethylene) (P(VDF-CTFE)),[292] gelatin,[293] polyamides (PA),[294] polyacrylonitrile (PAN),[295–297] polyimides (PI),[298] polyurethanes (PU),[299,300] *etc.* However, piezopolymers have weak electromechanical coupling and low piezoelectric coefficients, which result in low energy conversion efficiency and sensitivity. Improving the electrical properties of piezopolymer materials has now become a top priority in the community. Emerging nanofabrication technologies that could improve the piezoelectric response and expand the range of device structures have been developed. For instance, PVDF is a commonly-used organic piezoelectric material, the β phase, which exhibits surface charge separation caused by the parallel arrangement of dipoles, is the main contributor to the piezoelectric performance. Thus, various strategies have been employed to enhance the electrical performance brought by the presence of a higher β phase ratio in the material,[301] such as mechanical stretching,[302] melting under high pressure,[303] thermal annealing,[304] external electrical poling,[291] and ultrafast cooling.[305] Among them, electrospinning based on external electrical poling can produce piezopolymers in the form of nanofibers and nanowires with enhanced piezoelectric properties, which has been widely investigated. By rationally designing collectors and parameters, fibers can also be arranged in various forms, such as random mats,[306,307] aligned strands,[308,309] highly-aligned arrays,[310] nanoribbons,[311] or woven/knitted into textiles.[312–318] These strategies are helpful for the functional design and performance improvement of piezoelectric devices. Additionally, the polar electroactive phase can also be promoted in PVDF by employing an external filler in the absence of electrical poling.[319–321] In terms of structural design, nanotemplate confinement is an effective strategy, as immersing PVDF in an anodized aluminum template (AAO) permitted the formation of a random array of PVDF nanowire arrays to significantly increase the β-phase content of the polymer.[289,322,323] This approach is brought about as the stress induced by the nanotemplate promotes the transition from the α to the β-phase. In addition to this approach, various kinds of micro/nanostructures, such as nanowires,[324,325] micropillars,[285,326] pyramid-shaped films,[284] serpentine structures,[263,327] curved structures,[328] porous structures,[329,330] twisted yarns[331,332] have also shown enhanced outputs. Furthermore, introducing pores into polymers to form piezoelectrets through polarization is another effective method to obtain organic piezoelectric materials with high performance.[333–337] Interestingly, Yuan *et al.* recently reported a 3D-printed multilayer β-phase P(VDF-TrFE) copolymer which does not require high-temperature annealing or complicated transfer processes to yield high performance and exhibits a much higher effective piezoelectric coefficient output ($d_{33} \sim 130$ pC N^{-1} for six 10 μm layers).[192] This approach provides yet another convenient method for the preparation of high-performance wearable devices. Furthermore, transforming these planar structures into complex three-dimensional (3D) frameworks could provide novel operation modes,[266] demonstrating the untapped potential of PENG-related applications and attracting widespread attention. Indeed, Yang *et al.* recently demonstrated that the copolymerization of pyromellitic dianhydride with *p*-phenylenediamine and a specially designed NH_2-ended imidazole ionic liquid could yield a device with a piezoelectric coefficient of d_{33} as high as 420 pC N^{-1}, indicating great potential for wearable applications.[338]

3.3 Piezoelectric composites

Piezoelectric composites comprising mainly inorganic and organic materials are also of interest. Since desired properties can be achieved by adjusting the ratio of the component materials, extensive research has been conducted in recent years in this space. In general, piezoelectric composites combine the good mechanical flexibility of piezoelectric polymers and the outstanding electrical properties of inorganic piezoelectric materials, offering materials with a decent comprehensive performance that can be used in an even wider range of applications.[339,340] Various alternative concepts for piezoelectric composite have been developed, including those incorporating fillers of different morphologies into the polymer matrix, to construct 0-3 (fillers in form of nanoparticles),[341–352] 1-3 (fillers in form of nanowires, nanotubes, or fibers),[353–358] or 2-2 (fillers in form of nanosheets or laminates) geometries.[221,359] Both the filler and the polymer matrix can be piezoelectric materials or non-piezoelectric materials, in the form of three combinations of piezoelectric composites: piezoelectric fillers and non-piezoelectric polymers; non-piezoelectric fillers and piezoelectric polymers; and piezoelectric fillers and piezoelectric polymers. Piezoelectric fillers and non-piezoelectric polymers, mainly include mixed piezoelectric nanomaterials such as $BaTiO_3$, PZT or potassium sodium niobate (KNN) with non-piezoelectric polymers such as PDMS, rubber or epoxy resin to reduce the brittleness of ceramic materials.[360–364] In contrast, non-piezoelectric fillers and piezoelectric polymers mainly contain conductive materials (rGO, Ag NPs, carbon nanotubes, graphene),[365–372] metal oxides (MgO, TiO_2, Fe_3O_4, $CoFe_2O_4$)[373–378] or other materials (MXene,[379] sugar,[380] $MAPbI_3$,[349] polyaniline[381]) within piezoelectric polymers like PVDF or P(VDF-TrFE), enhancing the dipole polarization of the polymer to improve the electrical output performance of the composites. When both the fillers and the polymers are piezoelectric, the resulting composites not only improve mechanical flexibility but also enhance electrical output performance *via* the piezoelectric synergy.[103,246,382,383] In addition, much like piezoelectric polymers, many piezoelectric composites are also suitable for massive production using solvent-assisted extrusion technology, *i.e.*, electrospinning and 3D printing,[192,342,384–387] greatly facilitating the design and large-scale fabrication of wearable piezoelectric devices.

3.4 Natural piezoelectric materials

Although the previously discussed inorganic piezoelectric materials, polymers, and composites have been well developed, their fabrication processes often involve the use of some environmentally unsafe components (such as heavy metal ions, organic solvents), which are largely limited for on-body applications, especially implantable devices. More interestingly, piezoelectricity has been found in many natural materials, such as wood,[388] bone,[389,390] tendons,[391] viruses,[238–241] eggshells,[242,243] spider silk,[258] fish scales,[244] porcine skin,[392] chitin,[393,394] amino acids,[395] peptides,[396] deoxyribonucleic acid (DNA) and proteins.[397–399] Such nature-driven nontoxic, biodegradable piezoelectric materials have emerged as a new paradigm toward the development of next-generation wearable and smart electronic systems. However, natural piezoelectric materials usually exhibit weak piezoelectric properties with inherently uncontrollable shapes. In this situation, synthetic natural piezoelectric materials with designable structures and better piezoelectric properties have attracted an ever-growing amount of research efforts. Among many natural materials, the M13 bacteriophage has been identified as the most attractive candidate for the development of wearable PENGs due to its unique structure (Fig. 2f). To improve its electrical performance, one effective strategy is to modulate its film morphology through a self-templating assembly process of two- and three-dimensional hierarchical nanostructures.[239] Another method is to change the alignment direction of the phage. *Via* these modifications, the electrical output performance of M13 bacteriophage-based PENG was increased 2.6-fold when the phage was vertically aligned to construct nanopillars using enforced infiltration.[240] Some hybrid methods have also been explored to improve the electrical performance of M13 phage-based PENG. For instance, by combining the self-assembly of phages in a microfluidic channel, and the surface modification of phages through genetic engineering, the electrical output could be significantly enhanced to 2.8 V, about 100 times higher than that of in-plane aligned phages.[238] Recently, Guerin *et al.* obtained high shear piezoelectricity (178 pm V^{-1}) in the amino acid crystal β-glycine through efficient packing of the molecules along certain crystallographic planes and directions, which has further advanced the utility of natural piezoelectric materials.[400] In addition, the biocompatibility and biodegradability of these natural piezoelectric materials are highly beneficial for implantable applications, suggesting that PENGs using natural piezoelectric materials show great potential for healthcare applications.

3.5 Outlook on piezoelectric materials

As elaborated above, great progress has been made in both material design and structural design for piezoelectric materials. Recently, the piezoelectric properties of many new materials have also been discovered, including for materials: MXene,[401] ZnPc,[402] black phosphorus,[403] selenium nanowires,[255] tellurium nanowires,[404] monolayer hexagonal boron nitride,[405] single-component organic ferroelectric[406] *etc.* On this basis, the electrical and mechanical performance of the PENG has been further improved and expanded to fulfill more unconventional requirements, showing a promising application prospect in wearable bioelectronics, though several challenges that limit their further development must still be addressed.

Materials. In recent years, with the in-depth study of the morphotropic phase boundaries (MPBs) theory, both in ceramics and polymers, many piezoelectric materials with ultrahigh electrical properties have been obtained.[407–410] A recent report demonstrated that rhombohedral PMN-PT crystals could simultaneously achieve an ultrahigh piezoelectric coefficient of d_{33} (>2100 pC N^{-1}) and perfect transparency *via* domain engineering, which are highly desirable for wearable applications.[411] Meanwhile, with the rapid development of molecular piezoelectric materials, a series of novel metal-free organic perovskite piezoelectric materials exhibiting excellent piezoelectricity of 1500 pC N^{-1} (stronger than PZT) without high-temperature sintering, have been developed, making them an attractive choice for a variety of applications in flexible and wearable devices.[412–415] However, the relevant applications of such high-performance piezoelectric materials in PENGs are barely reported, further development and exploration are highly recommended. In particular, the degradable properties of piezoelectric materials are extremely advantageous for wearable devices, especially for implantable devices, and should be further investigated.[416,417] Indeed, most of the current biodegradable piezoelectric materials are natural piezoelectric materials with weak electrical performance, thus further research on improving their electrical performance is of paramount importance.

Preparation. At present, the most widely used high-performance inorganic piezoelectric materials are lead-based materials (mainly PZT). Despite its excellent piezoelectric properties, PZT contains heavy metals (Pb), which seriously limits its applications in wearable and implantable electronics. In this regard, Pb-free piezoelectric materials, such as $BaTiO_3$,[213–215] KNN,[418,419] $(Na_{0.5}Bi_{0.5})TiO_3$-$BaTiO_3$,[420] *etc.*, have been developed and exhibit electrical properties comparable to those of PZT. Nevertheless, most inorganic piezoelectric materials require high-temperature sintering during manufacturing, such as high-performance $BaTiO_3$-based ceramics that can only be prepared at a temperature of *ca.* 1450 °C.[421] This high-temperature requirement is a major obstacle for wearable applications. In addition, although some materials are naturally piezoelectric, most piezoelectric materials need to be repolarized with high-voltage electric fields to yield better electrical properties, whether inorganic, organic, natural piezoelectric materials or piezoelectric composites. Solutions on how to lower the high-temperature and high-voltage electric fields required in the preparation process of piezoelectric materials remain an urgent challenge.

Working mode. It is well known that the piezoelectric coefficients of most piezoelectric materials have a magnitude relationship in the order of $d_{15} > d_{33} > d_{31}$.[422] For example, the piezoelectric coefficient d_{15} of bulk PZT was reported to be 494 pm V^{-1}, larger than the other piezoelectric coefficients d_{33} (223 pm V^{-1}) and d_{31} (−93.5 pm V^{-1}).[423] However, most devices are currently operating in either the d_{33} or the d_{31} mode, whereas fewer tests using the d_{15} shear mode have been performed. If the piezoelectric response to the d_{15} mode were to be explored, a better piezoelectric output could perhaps be obtained, which in turn could improve the device's performance and applicability. Hence, establishing novel ways to develop piezoelectric devices in various modes through structural or material design, is also worth exploring.

Mechanism. Since the discovery of piezoelectric materials, plenty of research has been conducted to understand the underlying working mechanism.[225,403,424–426] For linear elastic regular crystals, there exists a continuous piezoelectric equation that can quantitatively describe the relationship between force and electricity coupling.[427] However, to date, no definite theoretical model can quantitatively describe the electromechanical coupling relationship of doped piezoelectric materials or piezoelectric composites. Although many reports attribute the performance improvement to the piezoelectric synergy,[103,246,382,383] the underlying mechanism is indeed unclear. The contribution of different material components to the electromechanical coupling performance also needs to be decoupled to enable better material and structural design. Moreover, a major issue for semiconductor-based piezoelectric materials is the presence of the screening effect[279,428–433] due to the presence of free carriers within the semiconductor, which will screen piezoelectric charges, subsequently reducing the electrical output. Therefore, establishing the mechanism of screening effect and providing a more fundamental solution for suppressing the screening effect are answers urgently required to optimize the performance of PENGs.

4. PENGs for energy harvesting

Due to their outstanding electromechanical coupling performance, piezoelectric materials can effectively convert mechanical energy into electrical energy and have thus been primarily developed for energy harvesting. The abundant biomechanical activities of the human body including walking, running, limb movement (*e.g.*, finger bending), and even the heart beating, can provide rich passive energy sources for PENGs. Medical devices currently implanted in the human body, such as cardiac pacemakers, defibrillators, brain stimulators, and cochlear implants, all rely on conventional batteries that are limited in use because of inherent factors linked to capacity, size, and lifespan, rendering their use problematic.[188,434,435] The emergence of PENGs provides a good solution to these shortcomings. In the past few years, researchers have conducted numerous studies to convert passive biomechanical energy into electrical energy through PENGs to provide a power supply for low-power electronic devices. In this section, the most recent research on the harvesting of passive biomechanical energy from human activities by PENGs using different piezoelectric materials is discussed.

4.1 Inorganic materials-based PENGs

PENGs based on inorganic piezoelectric materials have been extensively studied given their excellent electrical output performance. However, most traditional inorganic piezoelectric materials are bulky, inherently stiff, and brittle, unable to withstand sizable deformations. A common strategy used to address such limitations is to integrate the bulky piezoelectric material into locations that are subjected to relatively controlled deformations, such as footwear, and permit the harnessing of energy from human walking or running.[436–441] The focus of these studies revolved mainly around structural design modifications. For example, Qian *et al.* embedded piezoelectric stacks composed of PZT to amplify the vertical heel-strike force for higher energy conversion efficiency of the PENG harvester.[442] However, these devices can only respond to relatively large external forces and endure only relatively small deformations. To further improve the flexibility of the device, Yang *et al.* proposed a PENG based on a single ZnO nanowire, completely different from the traditional bulk piezoelectric

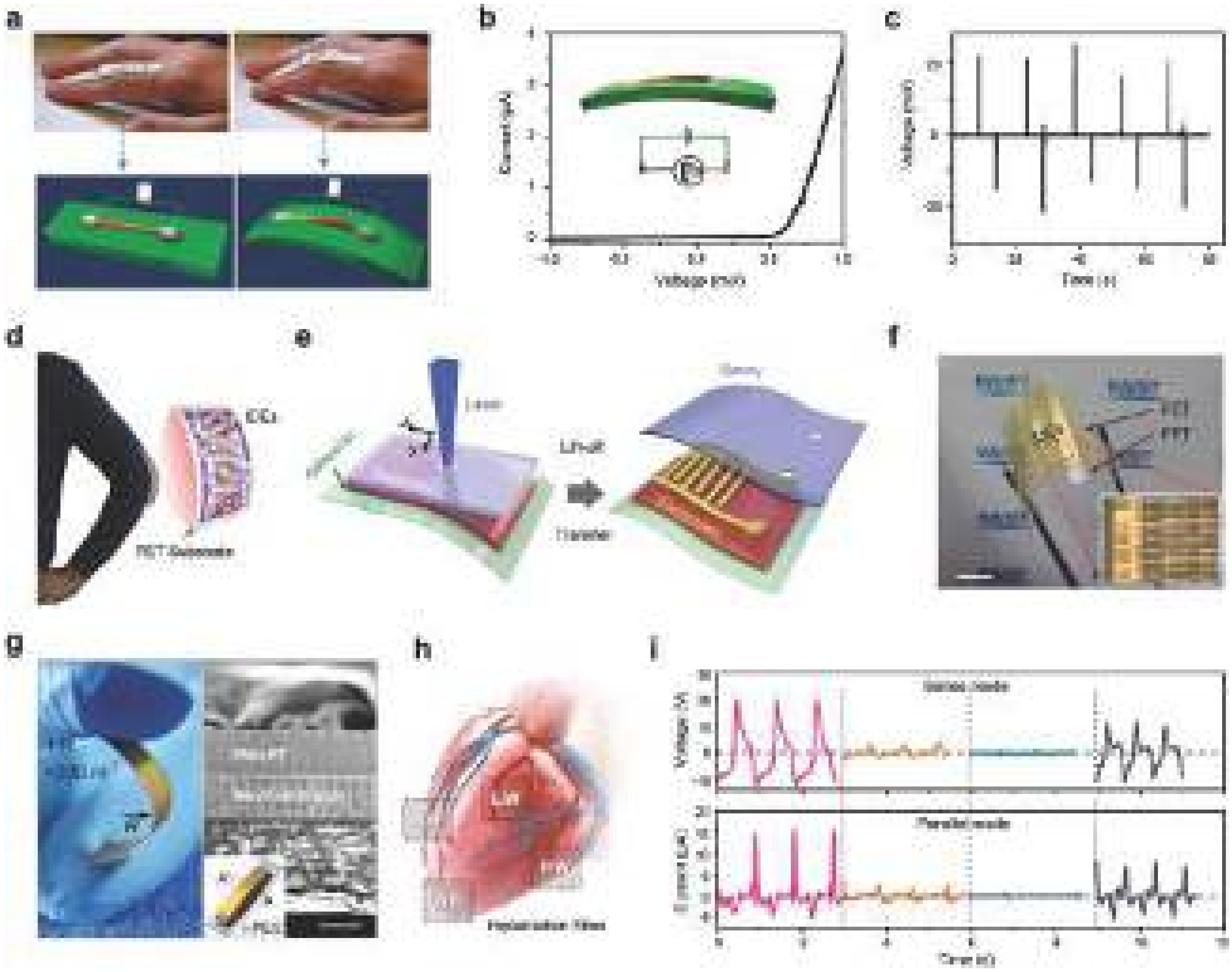

Fig. 4 Inorganic piezoelectric materials based energy harvesting. (a) Schematic diagram of ZnO nanowire-based PENG attached to a human index finger and bent with the finger. Reproduced with permission from ref. 443 Copyright 2009, American Chemical Society. (b) *I–V* characteristics of the PENG. The inset illustrates the schematic of the PENG and its connection configuration about the measurement system. Reproduced with permission from ref. 444 Copyright 2010, Wiley. (c) Open-circuit voltage generated by PENG when the finger was periodically bent back and forth. Reproduced with permission from ref. 443 Copyright 2009, American Chemical Society. (d) Schematic illustration of wearable applications using a flexible PZT energy harvester. Reproduced with permission from ref. 458 Copyright 2019, Elsevier. (e) Schematic diagram of the fabrication process for a flexible PZT thin film-based PENG using the LLO method. Reproduced with permission from ref. 216 Copyright 2014, Wiley. (f) Photograph of a flexible PZT energy harvester bent by tweezer. Scale bar, 1 cm. Reproduced with permission from ref. 458 Copyright 2019, Elsevier. (g) Photograph of a piezoelectric composite bent by human fingers and cross-sectional SEM image of the piezoelectric composite on the elastic skeleton. Scale bar, 50 μm. (h) Schematic view illustrating different implantation sites. (i) The output performance of implanted PENGs in series (upper) and parallel (lower) mode from different implantation sites. Reproduced with permission from ref. 472 Copyright 2019, American Chemical Society.

material.[443] In this approach, the two ends of the single ZnO wire (diameter was 100–800 nm with lengths of ∼100–500 μm) were placed in contact with metal electrodes, laterally bonded, and packaged on a flexible substrate, as shown in the lower part of Fig. 4a. The deformation of the ZnO wire generated piezoelectric potential within the wire *via* piezoelectricity, much like the ZnO which wire acted as a charge pump to drive electron flow back and forth in the external circuit. The as-fabricated ZnO wire-based PENG exhibited Schottky behavior as shown in the *I–V* characteristic curve (Fig. 4b), with piezoelectric potential effectively preserved.[444,445] When the ZnO wire was attached to the joint of the index finger, irregular finger motion could produce a voltage up to 25 mV, as shown in Fig. 4c. Since then, more research efforts have led to the development of nanowire-based PENGs.[446] For instance, Zhang *et al.* reported a ZnO-nanorods patterned textile used to harvest biomechanical energy *via* means of palm clapping or finger bending.[447] Similarly, a PENG built with a single 0.65PMN-0.35PT nanowire could generate a current of 1.5 nA *via* bending motions.[448] Jin *et al.* recently reported a PENG developed with vertically aligned PZT nanorod arrays, which could generate a voltage up to 8 V when tapped with a human finger.[449] Indeed, a large number of studies have adopted a similar method of discretizing traditional bulk piezoelectric materials into nanowires,[273,450] nanorods,[447,451] nanotubes[452,453] or nanosheets[454] to realize flexible energy harvesting.

To further improve the mechanical flexibility of PENGs, incorporating thin piezoelectric films onto flexible substrates is another effective strategy.[455–457] For instance, Park *et al.* developed a flexible energy harvester based on an eco-friendly $BaTiO_3$ thin film which was built through radiofrequency magnetron sputtering and soft lithographic transfer.[214] Although this device exhibited a certain degree of flexibility, the output performance was low (voltage of ~1.0 V and current of ~26 nA), limited by the narrow horizontal length of the piezoelectric active regions. To overcome this limitation, a large-area PZT thin film was developed on a flexible substrate through a laser lift-off process, a widely commercialized technique that can be used to detach functional film from sapphire substrates (Fig. 4e).[216] With the flexibility of such a substrate, PZT thin-film PENGs can readily collect energy from small-scale human motions, such as from elbow movements (conceptualized in Fig. 4d).[458] When a 2 μm PZT film was combined with lateral interdigitated electrodes, the resulting device exhibited high flexibility and durability upon bending (Fig. 4f), facilitating eventual attachment to clothing or human skin. When the interdigitated electrodes were designed with an electrode width of 100 μm and inter-electrode gap of 100 μm, an open-circuit voltage of 165 V and a short-circuit current of 1.5 μA could be obtained *via* continuous cyclical bending/relaxing motion (Inset of Fig. 4f).

In addition to converting biomechanical energy from motions of joints and limbs, including the finger,[110,459,460] jaw,[461] elbow,[332,462] ankle,[463] knee[464–467] or shoulder,[287,468] PENGs can also be used to harvest energy within the body, by coupling to motions generated by the heartbeat,[101,191,469] respiration,[102] internal muscle contraction,[443,444,470] even blood circulation.[155,471] Typically, Dagdeviren *et al.* proposed a PENG based on multilayered PZT ribbon, which could yield high-efficiency mechanical-to-electrical energy conversion from the natural contraction and relaxation of the heart, lungs, and diaphragm.[102] This as-fabricated PENG generated ~4.32 V output when exposed to a heart rate of 120 beats per minute. To further enhance the electrical output, Hwang *et al.* constructed a flexible PENG based on single-crystalline 0.72PMN-0.28PT thin film *via* a mechanical Ni exfoliation process.[219] The resulting device converted minimal biomechanical motion and mechanical deformation into electric energy, reaching 0.223 mA in current and 8.2 V output voltage peaks. Recently, Li *et al.* worked a double-layered implantable PENG (Fig. 4g), in which the same single-crystalline 0.72PMN-0.28PT with a thickness of 50 μm served as the piezoelectric layer.[472] This piezoelectric layer was bonded on the elastic skeleton of PET to give it flexibility, and no visible resulting damage was detected despite the piezoelectric layer being bent to curvature of ~200 m^{-1}. When the device was implanted at the apex (AP) of the heart (Fig. 4h), the maximal output voltage of ~20 V was yielded in the series mode, and the maximal output current of ~15 μA was yielded in the parallel mode (Fig. 4i). Using this approach, the researchers were able to power a commercial cardiac pacemaker utilizing said implantable PENG, an impressive step toward fabricating a self-powered cardiac pacemaker.

4.2 Organic materials-based PENGs

Compared with inorganic piezoelectric materials, piezoelectric polymers are very popular in the field of wearable energy harvesting because of their excellent flexibility. Numerous works have been reported on piezoelectric polymer-based energy harvesters that can scavenge biomechanical energy from human activities, especially those based on PVDF-related polymers.[473–478] Owing to their structural flexibility and processing simplicity, the polymer can be easily fabricated into thin, light, and flexible film systems. For instance, Zhao *et al.* developed a shoe-embedded PVDF energy harvester by mounting an 8-layer PVDF film on the inner sole responsible for gathering energy under the heel of the foot.[479] With a brisk walk at 1 Hz, a peak power of 4 mW could be obtained by this prototype. However, such films are relatively thick and less flexible. Pi *et al.* adopted the spin coating method to prepare thinner P(VDF-TrFE) film as the functional layer of a PENG.[286] The as-prepared nanogenerator exhibited an open-circuit voltage of up to 7 V and a short-circuit current of 58 nA. Compared with the aforementioned thick film, the spin-coated film showed better flexibility, although its flexibility was still undermined by the substrate. To further improve the performance, polymer films with well-controlled nanostructures can be designed. Mao *et al.* presented a large-area sponge-like PENG based on the mesoporous PVDF thin-film, made through a simple casting-etching process in the wafer scale, as illustrated in Fig. 5a.[480] This device could be directly attached to a body surface, such as a finger (Fig. 5b), and can effectively convert the mechanical energy of surface oscillations into electrical energy, enabling energy harvesting.[481] Alongside this example, a surface morphology engineering technology alternative was proposed to enhance the output performance of a PENG, based on P(VDF-TrFE) film, using a simple solvent annealing method at room temperature.[288] This surface morphology engineered PENG presented 8 times enhanced output voltage and current due to its well-aligned electrical dipoles. When the device was attached to a wrist, the movement of the wrist could generate a peak-to-peak voltage of up to 3 V. To further improve the electrical output performance, large strain stretching and high electric field polarization have also been proved to be effective approaches. Yu *et al.* demonstrated that electrospun PVDF nanofibers could effectively harvest the energy generated during walking or running when fabricated inside an insole.[229] The large squeezing force and the high electric field of electrospinning simultaneously provided *in situ* mechanical stretching and electrical polarization for the α-to-β-phase transition of PVDF.[69] Furthermore, Zhang *et al.* designed 3D interdigital electrodes to optimize the performance of flexible PENGs based on aligned P(VDF-TrFE) nanofibers.[482] As illustrated in Fig. 5c, the aligned P(VDF-TrFE) nanofibers were directly electrospun onto the 3D interdigital electrodes and encapsulated in PDMS. From the cross-section of the grooves in PDMS (Fig. 5d), it was obvious

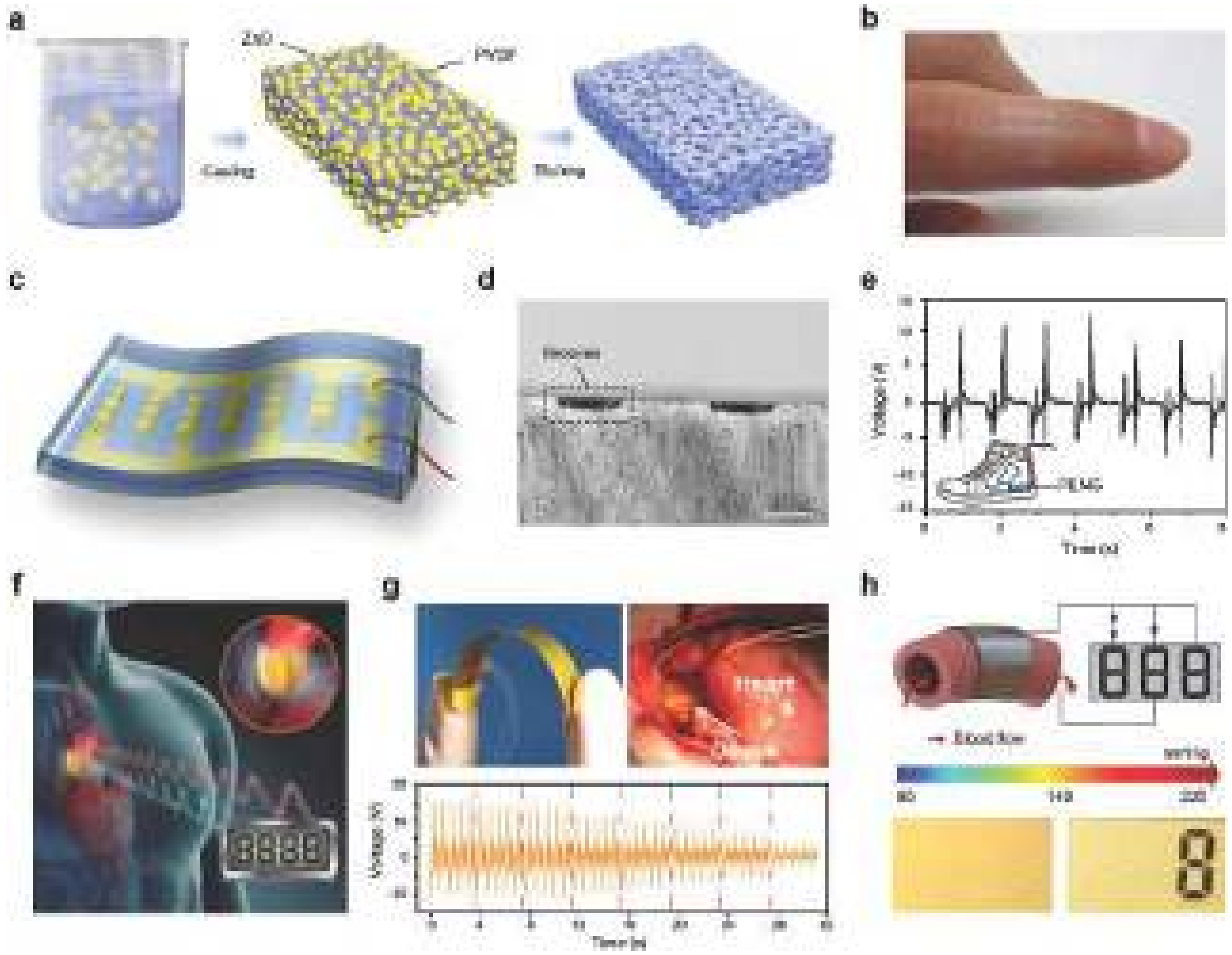

Fig. 5 Organic piezoelectric materials based energy harvesting. (a) Schematic procedure for fabricating mesoporous piezoelectric PVDF thin films. (b) Photograph of the mesoporous PVDF thin film adhered to the finger surface. Reproduced with permission from ref. 480 Copyright 2014, Wiley. (c) Schematic of a PENG based on aligned nanofibers and 3D interdigital electrodes. (d) Photomicrograph of a cross-section of the grooves in PDMS. Scale bar, 500 μm. (e) The output voltages generated by a person walking slowly. Reproduced with permission from ref. 482 Copyright 2019, Elsevier. (f) Schematic diagram of PENG used *in vivo* when wrapped on the aorta. (g) Photograph of the device showing its flexibility and wrapping on the ascending aorta of a pig (upper part). Output voltages of the device varying with different pressure applications (lower part). (h) System diagram and photo of LCD lit by a PENG when the systolic blood pressure was higher than 140 mmHg. Reproduced with permission from ref. 155 Copyright 2016, Elsevier.

that the structure of the groove was consistent with an inverted micropatterned quadrangular frustum pyramid. Both the gaps and sidewalls of micropatterns were all filled with aligned P(VDF-TrFE) nanofibers, an addition that further increased contact area between P(VDF-TrFE) nanofibers and 3D interdigital electrodes, resulting in improved electrical output performance. When wearing the fabricated PENG in a shoe, slow walking could yield a peak-to-peak voltage of up to 15 V, as shown in Fig. 5e.

In addition to the 3D electrode, Christine *et al.* demonstrated that using a melt-spinning process to weave 100% PVDF fibers into two-dimensional (2D) or three-dimensional (3D) braided fabrics, can also optimize the electrical output of the device.[483] More interestingly, Han *et al.* recently reported a sophisticated 3D piezoelectric microsystem based on PVDF that can work in complex motion patterns, thereby creating more opportunities for energy harvesting, especially when applied to small-scale systems.[266] Due to the nonlinear deformations from the 3D structure, the device can harvest energy from broadband motions. When the device was attached to the dorsal surface of the hand, a peak voltage >0.5 mV could be produced *via* an alternating closed fist/open hand motion. Indeed, after implantation of this device by the hind leg region of a mouse, a peak amplitude around 1 mV could be induced using trotting or climbing motions, with the 3D structure architecture providing more deformation capabilities than its 2D counterpart (in specific cases). In addition to this 3D structured PENG, many other polymer-based PENGs have been reported to collect a larger amount of kinetic energy *in vivo* that is not normally utilized. For example, Cheng *et al.* evaluated the feasibility and efficacy of a PVDF film-based implantable PENG to collect the energy produced by blood circulation (schematically shown in

Fig. 5f).[155] The device was wrapped onto an ascending porcine aorta, as shown in the upper part of Fig. 5g. The corresponding output voltage produced by the device showed similar peak flow pressure variation tendencies in the *in vitro* experiment, as shown in the lower part of Fig. 5g. The device was wrapped onto the aorta and a battery-free liquid crystal display (LCD) was fixed onto the porcine thoracic wall for *in vivo* demonstration. When the blood pressure exceeded 140 mmHg, the LCD could be turned on to display the systolic blood pressure of the animal, validating how such a set-up could be used to predict the risk of hypertension (Fig. 5h).

In addition to the above materials and engineering solutions, polymers filled with oval-shaped air voids also exhibit strong piezoelectricity after charging. These materials, collectively known as piezoelectret, have aroused much attention.[141,335,484] With outstanding mechanical property, Li *et al.* presented a PENG based on a cellular polypropylene piezoelectret,[485] which could be folded, allowing a doubling of the piezoelectric coefficient d_{33}, compared to its unfolded state. When a PENG composed of 7 piezoelectric layers was pressed by hand, the output voltage and current could reach higher than $\sim$50 V and $\sim$5 μA, respectively. As a result, the device was able to simultaneously power 20 LEDs upon pressure-driven activation. On this basis, Luo *et al.* fabricated a flexible insole with 80 layers of cellular PP, which was able to power a commercial ZigBee wireless transmitter *via* electricity generation from every 3 to 4 footsteps.[486] Apart from the polymers discussed above, other polymers such as PAN, PI, and PU also display piezoelectric properties and show potential in wearable energy harvesting.[295–300]

4.3 Piezoelectric composites-based PENGs

In order to obtain better overall performance, the preparation of piezoelectric composites is a compelling strategy, which combines the advantages of inorganic materials (*e.g.*, strong polarity, fast response) and organic polymers (*e.g.*, good flexibility, easy processability). This strategy has been extensively adopted to fabricate flexible PENGs, especially for wearable energy harvesting.[487–496] As discussed in Section 3.3, there are three different combinations of piezoelectric composites depending on the piezoelectric properties of the filler or matrix. In the case of piezoelectric fillers and non-piezoelectric polymers, some flexibility can be obtained at the expense of the piezoelectric properties of inorganic piezoelectric fillers. Li *et al.* blended the $Bi_4Ti_3O_{12}$ nanoplates and PDMS to form sandwiched multilayers to serve as piezoelectric functional layers, which could bend with arm.[497] In another work, Zhang *et al.* reported a high-performance flexible PENG consisting of 3-D cellular-structured PZT ceramic foam filled with PDMS, as shown in Fig. 6a.[218] Through a versatile preparation process, this 3D composite film could be conveniently fabricated over large areas and easily stretched/bent by fingers without damage (Fig. 6b), yielding output voltages ranging from $\sim$22 to $\sim$65 V as the tensile strain increased from 5% to 15% (Fig. 6c). Almusallam *et al.* demonstrated that the composites of PZT nanoparticle and polymeric binder were printable and could harvest energy *via* forming an active film on textile surfaces.[498] When they are non-piezoelectric fillers and piezoelectric polymers, the materials are inherently flexible and the polarization orientation of the piezoelectric polymers is enhanced by the fillers to obtain higher piezoelectric properties. For instance, Dutta *et al.* developed a NiO@SiO_2/PVDF nanocomposite with the enhanced piezoelectric output due to the electrostatic interaction between the negatively charged surface of NiO@SiO_2 nanoparticles and the positive $-CH_2$ group of PVDF, as schematically shown in Fig. 6d.[499] In addition, the piezoelectric properties of the composite could be further enhanced when both the filler and the polymer are piezoelectric, due to the presence of piezoelectric synergy. For instance, both the piezoelectric $BaTiO_3$ and PVDF were adopted to electrospun core–shell structured piezoelectric nanofibers, as shown in Fig. 6e.[500] The introduction of inorganic nanoparticles into the core and shell of the composite nanofibers (Fig. 6f) was capable of effectively enhancing the piezoelectric response and polarization effect. Similarly, Zhang *et al.* developed a fully lead-free PVDF-niobate-based PENG with enhanced performance through the formation of a polymorphic phase boundary.[259] Although such devices offer certain flexibility, they cannot be stretched. In this regard, Siddiqui *et al.* fabricated a stretchable PENG by combining the piezoelectric nanofibers and graphite electrode on a PDMS stretchable substrate.[501] Due to the stacked structure of the multiple free-standing nanofiber layers and the omnidirectionally stretched substrate, the designed PENG could deliver the highest peak power density of 1.76 μW cm^{-2} when subjected to a 40% stretching strain. Jeong *et al.* developed a hyper-stretchable energy harvesting device based on an elastic composite consisting of PMN-PT particles, multi-walled carbon nanotubes and silicone rubber.[346] This highly elastic PENG could generate an electrical output under various mechanical deformations (*e.g.*, twisting, folding, and crumpling) with a stretchability that reached up to 300%. Interestingly, based on a template-assisted sol–gel method, a three-order hierarchical piezoceramic-polymer textile was constructed by intertwining multiple ceramic fibers at the submillimeter scale (Fig. 6g).[502] As a result, the introduction of the hierarchical porous structure led to larger mechanical deformation, greatly improving the electrical output. Recently, Gu *et al.* proposed a three-dimensional intercalated electrode design to further improve the output performance of the PENG,[503] as shown in Fig. 6h. Here the piezoelectric composite was sandwiched between multiple pairs of electrodes, a radically different approach from the traditional 2D interdigital electrode. Under an external force, the small current generated in each unit could be combined to form a large output current *via* the designed 3D intercalation electrodes. When this composite was spin-coated into a thin film, the device could be placed directly under the insole cushion of a shoe to act as a human-motion-driven power source, showing great potential in scavenging biomechanical energy from low-frequency human motions such as walking or elbow joint movement, as schematically illustrated in Fig. 6i. Generally, compared with inorganic piezoelectric ceramics and organic

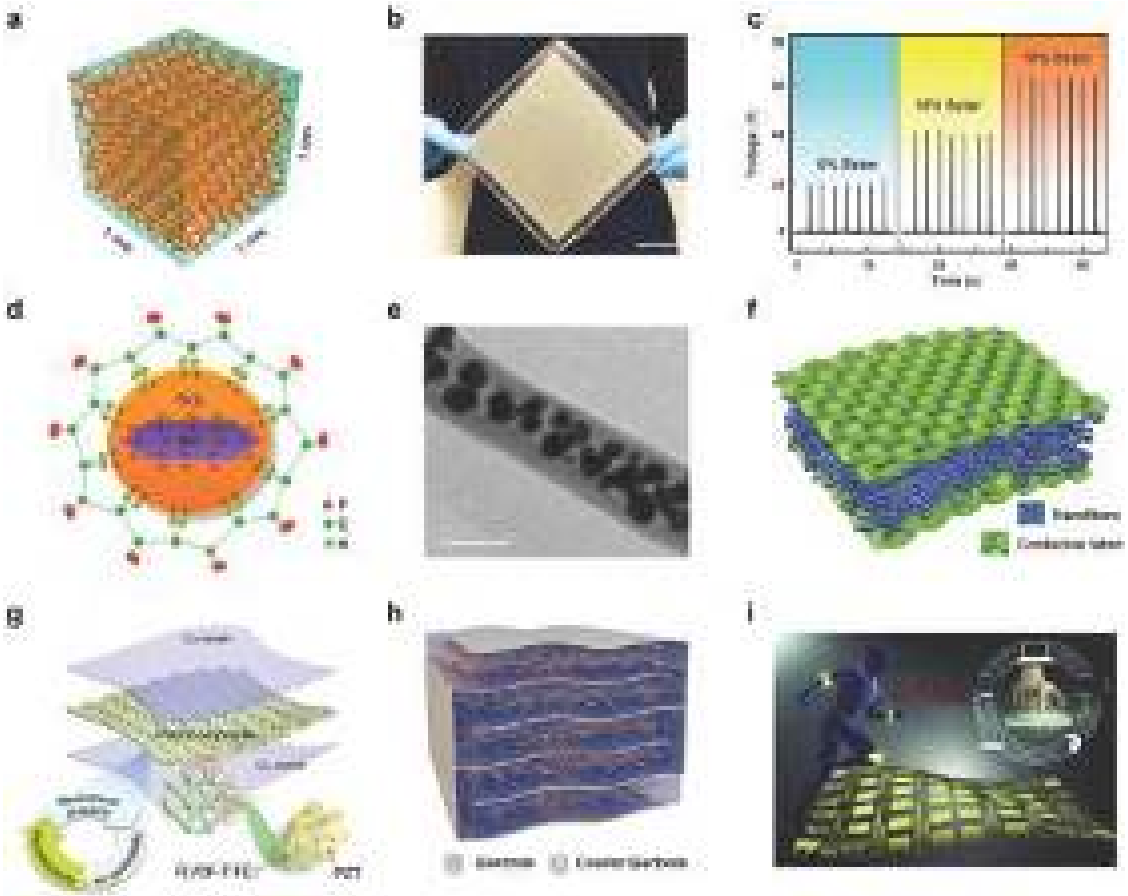

Fig. 6 Piezoelectric composites based energy harvesting. (a) Schematic diagram of a 3D interconnected piezoelectric ceramic foam-based PENG. (b) Photograph of a large-area 3D piezoelectric composite. Scale bar, 5 cm. (c) The output voltages of the 3D piezoelectric composite under different stretching strains. Reproduced with permission from ref. 218 Copyright 2018, Royal Society of Chemistry. (d) Schematic of the proposed β phase formation mechanism. Reproduced with permission from ref. 499 Copyright 2018, American Chemical Society. (e) TEM image of a core–shell piezoelectric nanofiber. Scale bar, 500 nm. (f) Schematic of a single unit of electronic skin. Reproduced with permission from ref. 500 Copyright 2020, Elsevier. (g) Schematic illustration of the HIPT composite based on a hierarchical PZT ceramic framework and a P(VDF-TrFE) coating film, with Cu mesh as the electrodes. Reproduced with permission from ref. 502 Copyright 2021, Wiley. (h) Schematic diagram of the Sm-PMN-PT-based PENG with a 3D intercalation electrode. Reproduced with permission from ref. 503 Copyright 2020, Nature Publishing Group. (i) Illustration of the proof-of-concept of fabricated electronic skin for joint motion energy harvesting. Reproduced with permission from ref. 500 Copyright 2020, Elsevier.

piezoelectric polymers, piezoelectric composites could simultaneously maintain decent electrical output and mechanical flexibility, allowing devices using these materials to exhibit better overall performance.

4.4 Natural piezoelectric materials-based PENGs

Compared to the previously discussed piezoelectric materials, natural piezoelectric materials are often compelling due to their high uniformity and degradability. Previous studies have shown that hierarchically structured natural materials (such as bones, tendons, viruses, polypeptides, or amino acids) can exhibit piezoelectric properties which can also be used for energy harvesting. For instance, Lee *et al.* demonstrated that the piezoelectric properties of M13 bacteriophage could be employed in energy harvesting applications.[253] Structurally, the M13 phage is rod-like in shape (Fig. 7a) and covered by major and minor coat proteins with a net dipole moment along its axial direction (Fig. 7b). Because its aligned protein coat structure lacks inversion symmetry, the phage film of ~100 nm thick demonstrated a d_{33} value of up to 7.8 pm V^{-1}. On this basis, Shin *et al.* assembled vertically aligned phages *via* enforced infiltration methods with a porous template to further enhance piezoelectricity, as illustrated in Fig. 7c.[240] With repeated infiltration processes, the phages were continuously captured in the channel of the template, leading to the formation of phage nanopillars (Fig. 7d) and a three-fold increase in d_{33} compared to film-type phages, suggesting feasibility for energy harvesting. Such protein-based materials are of great interest due to their abundant availability, ease of processing, and longitudinal piezoelectric sensitivity. For example, the piezoelectric response of spider silk at the molecular level was investigated by the Khatua group,[258] with the topography image of nature-driven spider silk illustrated in Fig. 7e. The aligned microfibrils of spider silk could induce piezoelectric

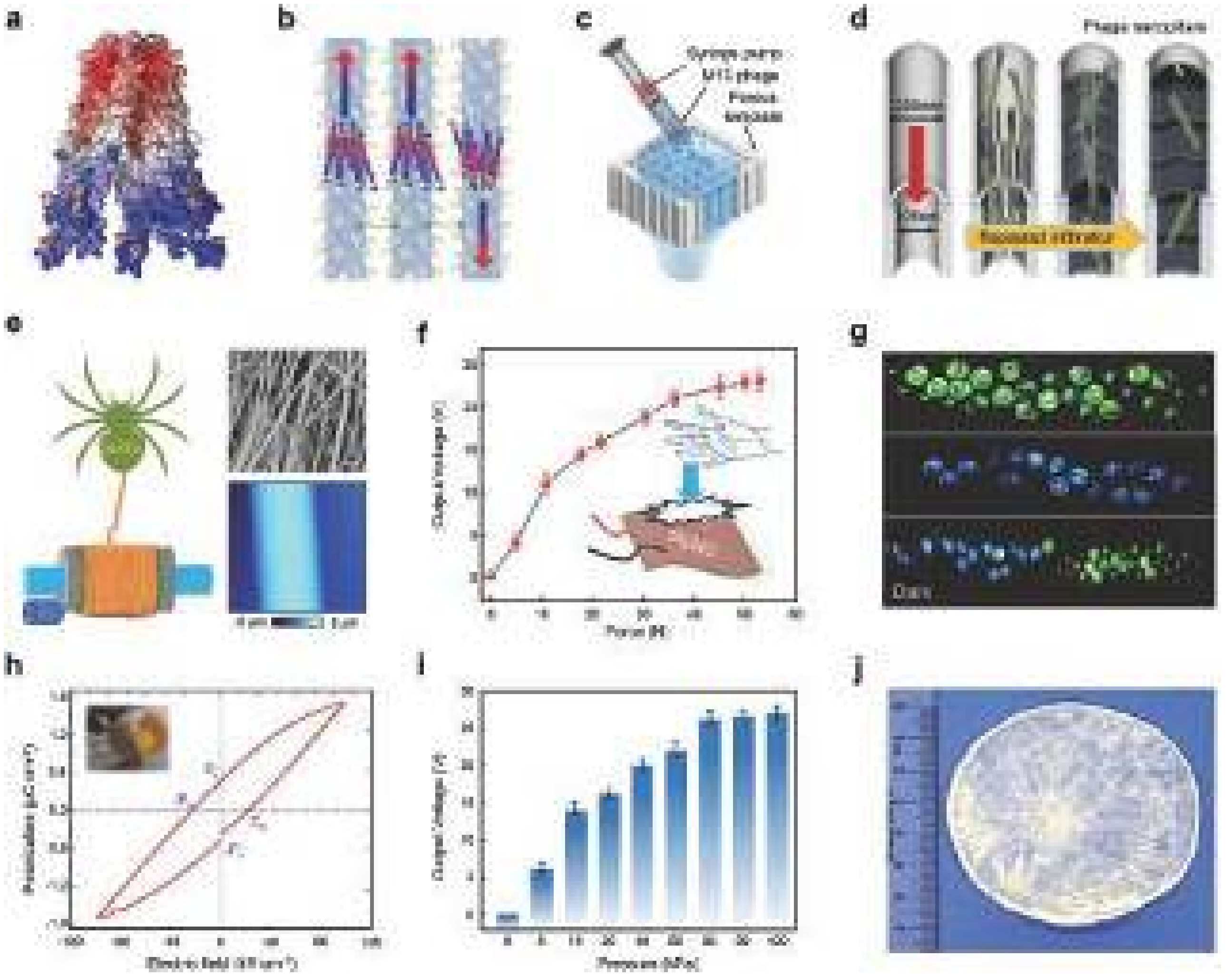

Fig. 7 Natural piezoelectric materials based energy harvesting. (a) Schematic of the piezoelectric M13 phage structure after the bioengineered modification with four glutamate amino acids. (b) Schematic depicting piezoelectric properties of phage with randomly mixed dipoles through the axial direction of the monolayer. Reproduced with permission from ref. 253 Copyright 2012, Nature Publishing Group. (c) Schematic of the enforced infiltration for the assembly of the vertically aligned M13 phages. (d) Schematic illustration of the phage nanopillar formation controlled by repeated infiltration in the porous template. Reproduced with permission from ref. 240 Copyright 2015, Royal Society of Chemistry. (e) Schematic of spider silk structure (left). SEM image of natural spider silk (upper right). Scale bar, 20 μm. Topography image of spider silk (lower right). (f) The output voltage generation of a PENG subjected to external applying force. (g) The power generated from 3 units in series instantly lit up green and blue LEDs. Reproduced with permission from ref. 258 Copyright 2018, Elsevier. (h) Polarization and electric field curves of unpolarized ESM. (i) Pressure-dependent output voltage. Reproduced with permission from ref. 242 Copyright 2018, Elsevier. (j) Digital photograph of a wafer-sized piezoelectric glycine-PVA film. Reproduced with permission from ref. 506 Copyright 2021, The American Association for the Advancement of Science.

properties under mechanical stress *via* crystal deformation processes. Experimental results showed that the energy harvesting performance of spider silk strongly depended on the applied mechanical force (Fig. 7f). For a more intuitive display, the authors connected the devices in series by assembling them layer by layer, and they were able to light up 55 LEDs (30 green and 25 blue) with the generated energy, as shown in Fig. 7g. Furthermore, Karan *et al.* demonstrated that bio-waste porous eggshell membrane (ESM) could also be utilized as an efficient piezoelectric material.[242] From the polarization-electric field hysteresis loop (Fig. 7h), the remnant polarization of ESM was about 0.46 μC cm^{-2}, with a correspondingly calculated piezoelectric coefficient of ~19.3 pC N^{-1}. The output voltage of the device showed a strong dependence on the applied pressure, as seen in Fig. 7i, and was able to perform electromechanical conversion effectively. Through hand punching, a capacitor (10 mF) was charged up to ~10.3 V within 105 s by the ESM-based PENG, demonstrating fast-charging capabilities. Alongside ESM, many other natural materials such as fish skin,[140] fish bladder,[504] prawn shell,[505] crab shell[394] have also been developed as energy harvesters. Recently, Yang *et al.* presented a wafer-scale approach to creating piezoelectric biomaterial thin films based on γ-glycine crystals (Fig. 7j), which showed great potential for commercialization.[506] These easily available, naturally driven, biodegradable and biocompatible materials are ideal for deep brain stimulators, pacemakers, and nerve stimulators that require continuous energy and hold huge application prospects in implantable medical electronics.

4.5 The integration of PENG and energy storage

Current energy harvesters have the potential of providing electrical energy in time by harnessing biomechanical energy from the human body. However, the random movement time and uncertain movement amplitude make the output of PENGs fluctuate, thus utilizing PENGs for continuous stable energy supply remains a challenge. In this regard, a common solution is to first store the generated electrical energy *via* independent energy storage devices (such as batteries or supercapacitors) and then release the electrical energy at the discharge plateau of the energy storage units to deliver a stable electrical output.[507–513] For example, Petritz *et al.* integrated the PENGs and organic diodes on ultrathin (1 μm) substrates (Fig. 8a), thus imparting them with excellent flexibility (Fig. 8b), which could harness energy from biomechanical motions if the multilayer devices were worn against joints, knees, and elbows.[514] In recent years, with the continuous development of micro-nano processing technology and the increasing demand for miniaturization, integrating these energy harvesters and energy storages into one system has been extensively explored.[515–517] Tong *et al.* reported an all-solid-state flexible piezoelectric high-k film functioning as both a generator and *in situ* storage unit.[518] This bifunctional piezoelectric film was made of polyvinylidene fluoride-*co*-hexafluoropropylene (PVDF-HFP) as matrices and reduced graphene oxide (rGO) as fillers to harvest the mechanical energy of finger bending motions as well as store the converted electrical energy, as illustrated in Fig. 8c. When the integrated system was fixed onto the finger of a disposable plastic glove, the output currents corresponding to

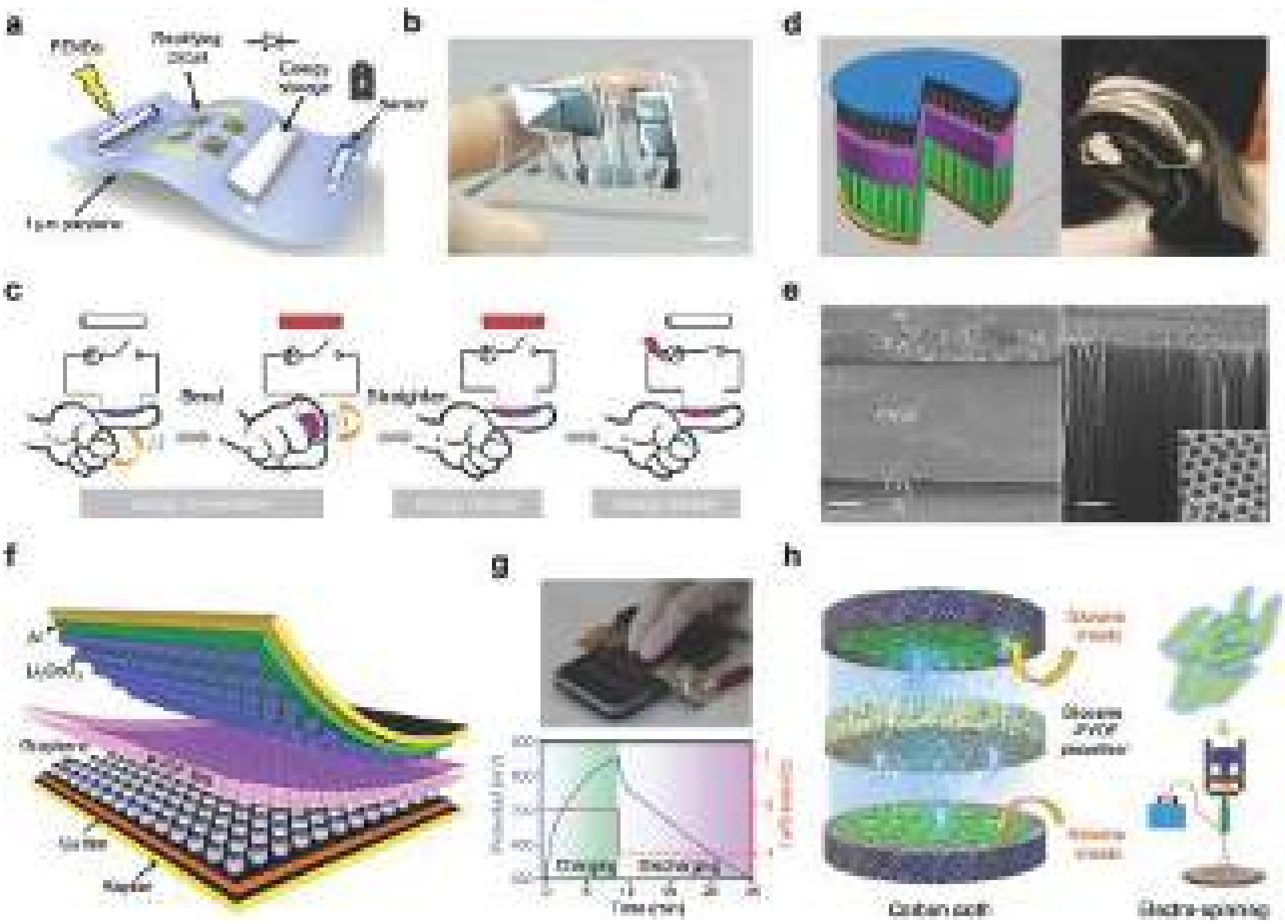

Fig. 8 Integration of energy harvesting and storage. (a) Scheme of a range of ultraflexible devices integrated on a 1 μm thin parylene substrate. (b) Photograph of the ultraflexible PENGs for harvesting biomechanical energy. Scale bar, 1 cm. Reproduced with permission from ref. 514 Copyright 2021, Nature Publishing Group. (c) Schematic illustrations of energy conversion and storage by nanocomposite films under biological motions. Reproduced with permission from ref. 518 Copyright 2015, Wiley. (d) Structure design of a self-charging power cell by hybridizing a PENG and a Li-ion battery. (e) Cross-sectional SEM image of a self-charging power cell (lower left corner). Scale bars, 30 μm. Enlarged view of the aligned TiO_2 nanotubes (lower right corner). Scale bar, 1 μm. The inset is a top view SEM image of the nanotubes. Scale bar, 200 nm. Reproduced with permission from ref. 519 Copyright 2012, American Chemical Society. (f) Schematic diagram showing the design of a flexible self-charging power cell. (g) Photograph of a flexible self-charging power cell charged by finger pressing (upper part). A typical self-charging and corresponding discharge process (lower part). Reproduced with permission from ref. 520 Copyright 2014, Wiley. (h) Schematic representation of a siloxane-based self-charging power cell. Reproduced with permission from ref. 524 Copyright 2020, Nature Publishing Group.

finger movements were observed, with stored electrical energy available to be exported after the finger motion stopped. Another viable integration strategy is to replace the separator of a traditional lithium battery with a piezoelectric film. For example, a PVDF film was used to replace the polyethylene film of a lithium battery to fabricate a self-charging power cell (Fig. 8d).[519] The piezoelectric potential generated by PVDF under mechanical strain acted as a charge pump to drive lithium ions from the cathode to the anode to achieve energy collection and storage. The cross-sectional scanning electron microscope (SEM) image of the self-charging power cell and the enlarged view of the aligned TiO_2 nanotubes are shown in Fig. 8e. It is important to note that this power cell was sealed in a rigid coin-type stainless steel cell that could not withstand large deformations, resulting in rather low efficiency in stress or strain conversion. To this end, Xue *et al.* further developed a flexible self-charging power cell using Kapton film as the supporting shell, flexible graphene material as the electrode, and piezoelectric PVDF film as the separator, which was sealed in a liquid electrolyte, as schematically shown in Fig. 8f.[520] Given the flexibility of the supporting shell, the as-fabricated self-charging power cell could be charged up to 832 mV *via* a 1 Hz frequency fingers pressure application, as shown in Fig. 8g, which proved that the designed device can simultaneously perform energy harvesting and energy storage. In addition, some similar hybrid cells have PVDF layers replaced by porous PVDF,[521] P(VDF-TrFE) film coated with PVA/H_3PO_4 gel,[522] PVA-KCl-$BaTiO_3$ gel membrane[523] to enhance the charging performance. Most recently, Krishnamoorthy *et al.* replaced conventional PVDF films with siloxene-based polymeric piezofibers (Fig. 8h), further improving the efficiency of the self-charging supercapacitor systems.[524] To summarize, this integral design of energy harvesting and energy storage devices allows direct conversion of mechanical energy into electrochemical energy without wasting energy on external circuitry and reduces energy conversion losses, effectively facilitating the practical applications of PENGs as sustainable power sources, especially in an implantable manner.

4.6 Outlook on PENGs for energy harvesting

As the most direct application of piezoelectric materials, research on PENG-driven energy harvesting has made considerable progress in recent years. Various prototypes for collecting passive human biomechanical energy have been designed and optimized on all fronts, from materials and structures to system integration. Although integrated systems for *in situ* storage of harvested energies have been successfully developed and validated, there is still a considerable gap between achieved and expected performance. Some critical issues should thus be addressed, and extensive efforts are required in many fields: (1) PENG exhibits the maximum output at the resonance frequency, usually in the high-frequency range, however, most human biomechanical movements fall in the low-frequency range.[68] Broadening the working frequency band and ensuring continuous device output, is always a challenge. In addition, most of the currently reported PENGs have been tested under simplified harmonic excitation and thus show considerable performance, but the real-life motion is heterogeneous with strong randomness and intermittency, the electrical output for practical applications may be even lower. (2) The subjected stress or strain of the device varies with the physical location, which directly determines its electrical output performance. For instance, the PENG could yield a maximal output voltage of $\sim$20 V when attached to the apex of the heart, and almost no output when it was attached to the posterior wall of the heart.[472] To obtain a higher output, there are thus still restricting physical location prerequisites for the piezoelectric device to ensure sufficient energy generation. (3) The overall output of PENGs is still low, thus powering electronic devices or third parties is contingent on there being enough charging cycles. The power management circuit is particularly important on this front as it acts as a bridge between the PENG and the energy storage equipment.[458,525–527] Efficient power management circuits with limited energy loss are desirable to permit an optimized extraction of harvested energy. (4) The integration of energy harvesting and storage is an important research direction, but higher requirements are placed on energy conversion efficiency, which is directly related to high-performance energy conversion and storage materials. In particular, the miniaturization process of integrated devices will lead to a reduction in the effective working area of functional materials and a decrease in energy transfer efficiency, which is of particular concern. (5) Most of the reported PENGs used as a self-charging system are still prototypes at the proof-of-concept stage. For implantable PENGs, most experiments are performed on small animals such as rats or mice, and *in vivo* experiments covering large animals and humans are urgently needed to further validate the research.

5. PENGs for sensing

In addition to energy harvesting, another important application area of piezoelectric materials is self-powered sensing. According to the piezoelectric equation, there lies a quantitative relationship between the electrical output of the piezoelectric material and the subjected strain or stress, indicating that PENGs can be directly used as sensors to detect and measure external force-related stimuli, such as pressure, strain, vibration, acceleration, acoustic waves, *etc.* Specifically, given the presence of positive and negative charge centers within piezoelectric materials, no external power source is required to generate electrical signals under an applied force, thus eliminating the need to include, manage, or replace a battery. On this basis, active self-powered sensors can be developed as a highly desirable solution for the next generation of wireless wearable and implantable devices. For on-body PENGs' use, various vital signs monitoring and application scenarios have been exploited extensively, promising several innovative solutions for personalized healthcare.

5.1 PENGs for human motion sensing

Human motion sensing has important applications in many fields, including healthcare, human-machine interaction, "intelligent" sport,[528–530] and can provide detailed information with regards to the motion quality and type in the form of a quantitative electrical signal which can be further analyzed. PENGs can be attached anywhere needed such as necks,[531] fingers,[233] wrists,[532,533] elbows,[462,534] or knees,[535] and can help monitor joint motions such as bending angle, deformation amplitude, and frequency.[536–538] Much like joints, smaller muscles such as those controlling the eye and eyelid can also be monitored using PENGs as well. Lee *et al.* developed an ultrathin PENG made of aligned ZnO nanowire compacted arrays that could work as an active sensor for eyeball motion tracking.[539] The high sensitivity of the fabricated flexible PENG enabled the measurement of the local deformation of the eyelid brought about by the eye movements, as shown in Fig. 9a. Similarly, Feng and his collaborators replaced the functional layer of ZnO nanowires with PZT nanoribbons to create a sensor only 10 μm thick, which could detect abnormal eye movements including overlong closure, high blinking frequency, and weak gazing strength, with the promise to provide timely feedback with which to assess parameters such as eye fatigue.[540] Beyond eyeball motion monitoring, Sun *et al.* attached a conformable piezoelectric device to the face, consisting of an AlN film on a compliant PDMS substrate, and achieved the decoding of

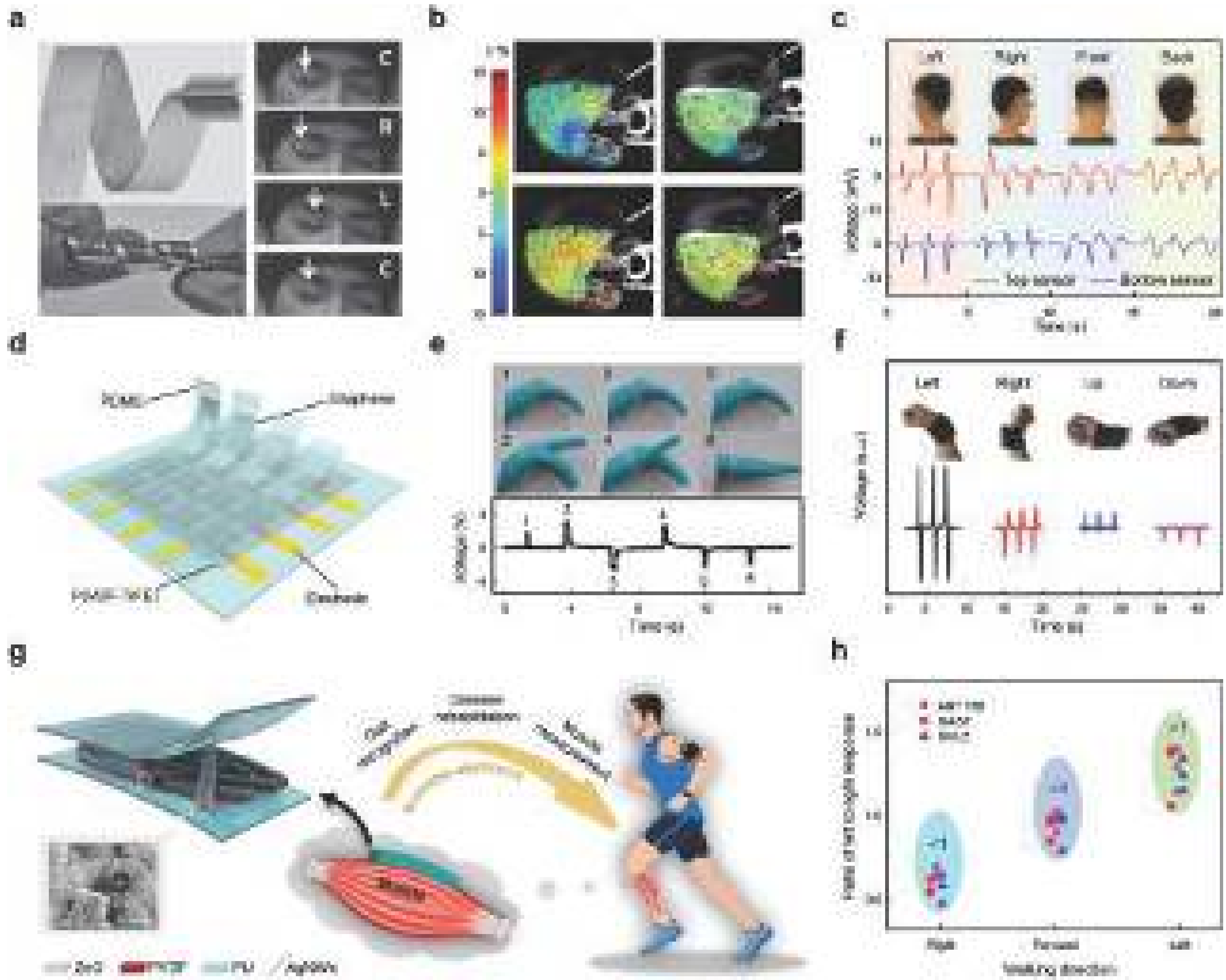

Fig. 9 PENGs for human motion sensing. (a) Ultrathin nanogenerators as self-powered skin sensors for tracking eyeball motion. Reproduced with permission from ref. 539 Copyright 2014, Wiley. (b) Skin strain was analyzed while participants performed facial motions, such as right cheek twitch. The images on the left show strain maps as an example of the results from a human participant with the sensor (left) and without the sensor (right), showing minimal principal strain (top) and maximal principal strain (bottom). Reproduced with permission from ref. 541 Copyright 2020, Springer Nature. (c) Experimental results of the electrical signals produced from neck motions based on piezoceramic kirigami. Reproduced with permission from ref. 542 Copyright 2021, The American Association for the Advancement of Science. (d) Schematic illustration of the multistage sensation matrix. (e) Photo image of the four-column multiple sensors attached in the palm to capture the motion of hand bending and fingers lifting (top panel) and the corresponding sensing signals (bottom panel). Reproduced with permission from ref. 543 Copyright 2018, American Chemical Society. (f) The output signals of a smart wristband subjected to various wrist movements. Reproduced with permission from ref. 47 Copyright 2018, Elsevier. (g) Schematic diagram of the three-dimensional hierarchically interlocked PVDF/ZnO fibers-based PENG for muscle behavior monitoring. Scale bar, 1 μm. (h) The ratio of calf response amplitude (Right leg) to the other calf response amplitude (Left leg) in different walking directions. Reproduced with permission from ref. 194 Copyright 2020, Elsevier.

facial strains as well as unique facial deformation features (Fig. 9b).[541] Recently, a kirigami-structured highly anisotropic piezoelectric network composite sensor was proposed to monitor multiple information of joint motions. As shown in Fig. 9c, as the neck moved in different directions, the top and bottom sensors generated different electrical signals due to the piezoelectric anisotropy, thus enabling the monitoring of neck motion.[542] Furthermore, based on a crossbar design, a flexible, self-powered multi-layer sensory matrix was also constructed to detect the complex motions of the human body from a fixed position.[543] As shown in Fig. 9d, sensing units in the matrix each contained a P(VDF-TrFE) sensing layer sandwiched between two graphene electrodes. When a four-column multi-sensor was attached to the palm of a hand, multiple movements including hand bending and finger lifting could be captured, the corresponding images and sensing signals of which, are illustrated in Fig. 9e. In this example, the distribution of strain and detaching areas could be detected by the sensation matrix and displayed in the form of 2D color mapping. To further improve the flexibility, a stretchable PENG based on a PZT/rubbery matrix was used as a smart wristband for joint monitoring.[47] As visible in Fig. 9f, the bending movement of the wrist in different directions (up, down, left, and right) can be monitored and represented *via* piezoelectric signals with different peak heights and widths. Yang *et al.* have recently proposed a flexible PENG using 3D hierarchically interlocked PVDF/ZnO nanofibers as piezoelectric functional layers, demonstrating ultra-high sensitivity and accurate detection of subtle muscle activity, as schematically illustrated in Fig. 9g.[194] Due to its decent flexibility, the fabricated sensor array could be conformally and firmly attached to the epidermis of the calf muscles, thus allowing the detection of slight deformations caused by muscle tension or relaxation. Gait monitoring could be moreover carried out by analyzing electrical signals specifically associated with different muscles during activities such as walking (Fig. 9h).

Combining flexible sensing, signal processing, and control execution, some researchers further enabled the recognition and synchronization of human actions using flexible PENGs.[468] For example, Deng and his colleagues successfully demonstrated the remote transmission of gestures to a robotic arm using a combination of PENG sensing, signal transmission, and executive control, as shown in Fig. 10a.[256] Here, the functional layer of the PENG was made of cowpea-structured PVDF/ZnO nanofibers (Fig. 10b), which better facilitated the mechanical flexibility as well as electrical output, enabling the

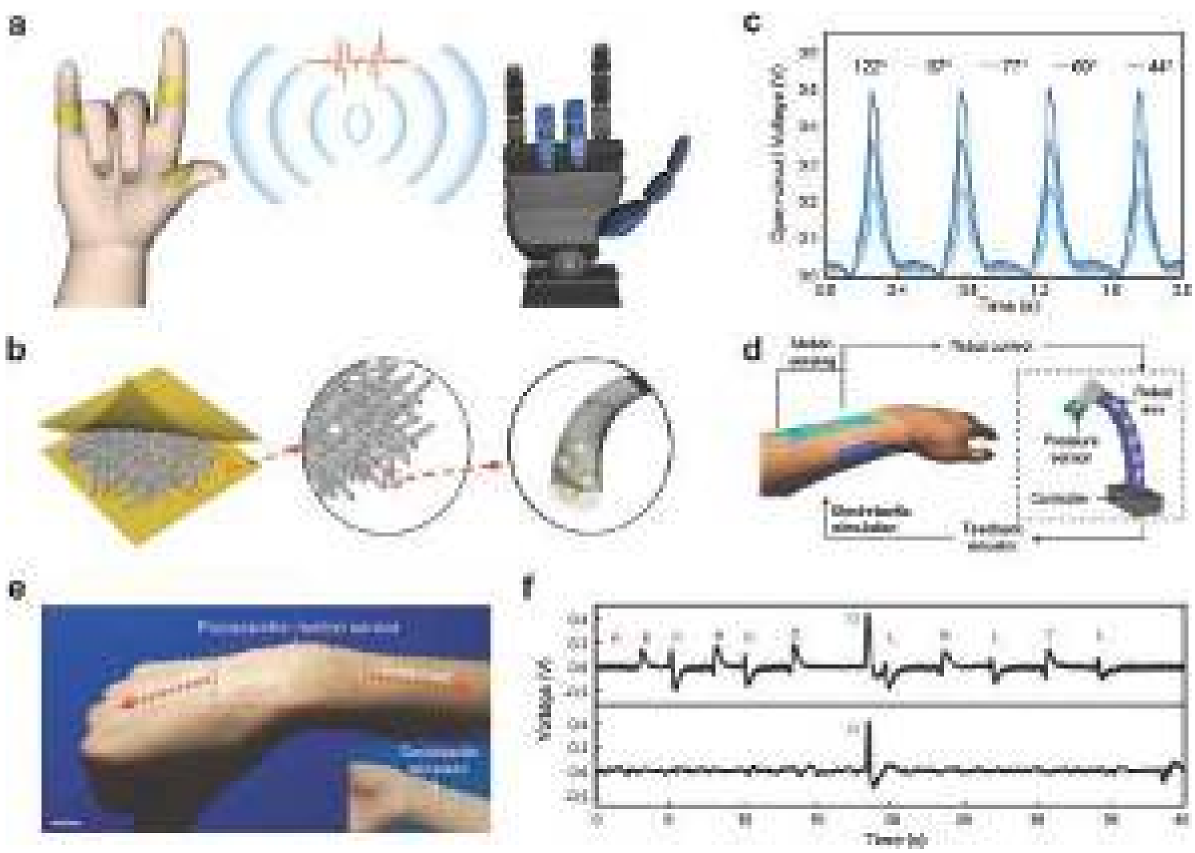

Fig. 10 PENGs for human machine interaction. (a) Schematic diagram of self-powered piezoelectric sensor based on cowpea-structured PVDF/ZnO nanofibers towards the remote control of gestures. (b) Sketch of the designed device comprising a nanofiber film, and a single nanofiber. (c) The open-circuit voltage generated by bending at different angles. Reproduced with permission from ref. 256 Copyright 2019, Elsevier. (d) Schematics of the closed-loop system of the interactive human-machine interface. (e) Images of a transparent piezoelectric motion sensor and the electrotactile stimulator (inset). Scale bar, 20 mm. (f) Detected signals acquired by the transparent motion sensor (first row) and by the pressure sensor on the gripper (second row). Reproduced with permission from ref. 544 Copyright 2014, Wiley.

designed sensors to quickly and sensitively respond to bending angle motions (Fig. 10c). When the sensor was fixed on the finger knuckle, the finger bending movement could be converted into electrical signals by the sensor, with the robotic hand remotely controlled to perform the same gesture as the human hand through a peripheral circuit module. Similarly, Lim *et al.* reported a closed-loop interactive human-machine interface system based on a designed sensor and actuator, as shown in Fig. 10d.[544] The designed transparent and stretchable piezoelectric motion sensor consisted of poly(L-lactic acid), single-walled carbon nanotubes, and silver nanowires (Fig. 10e), which could be used to monitor the wrist movements and convert the collected data into electrical signals to control a robotic hand. When the human wrist was bent, pressed, or relaxed, the robotic arm would mirror those movements. The corresponding representative data collected from the motion sensor is presented in Fig. 10f. Human movements tend to generally consist of relatively large deformations which can be easily detected *via* a wearable PENG. In summary, PENG-based human motion sensing has been widely adopted to monitor a variety of body sites, showing huge application potential.

5.2 PENGs for vital signs sensing

Within the promise of a personalized healthcare system, monitoring of vital signs is indispensable, to not only accurately assess the physiological state of an individual, but also provide

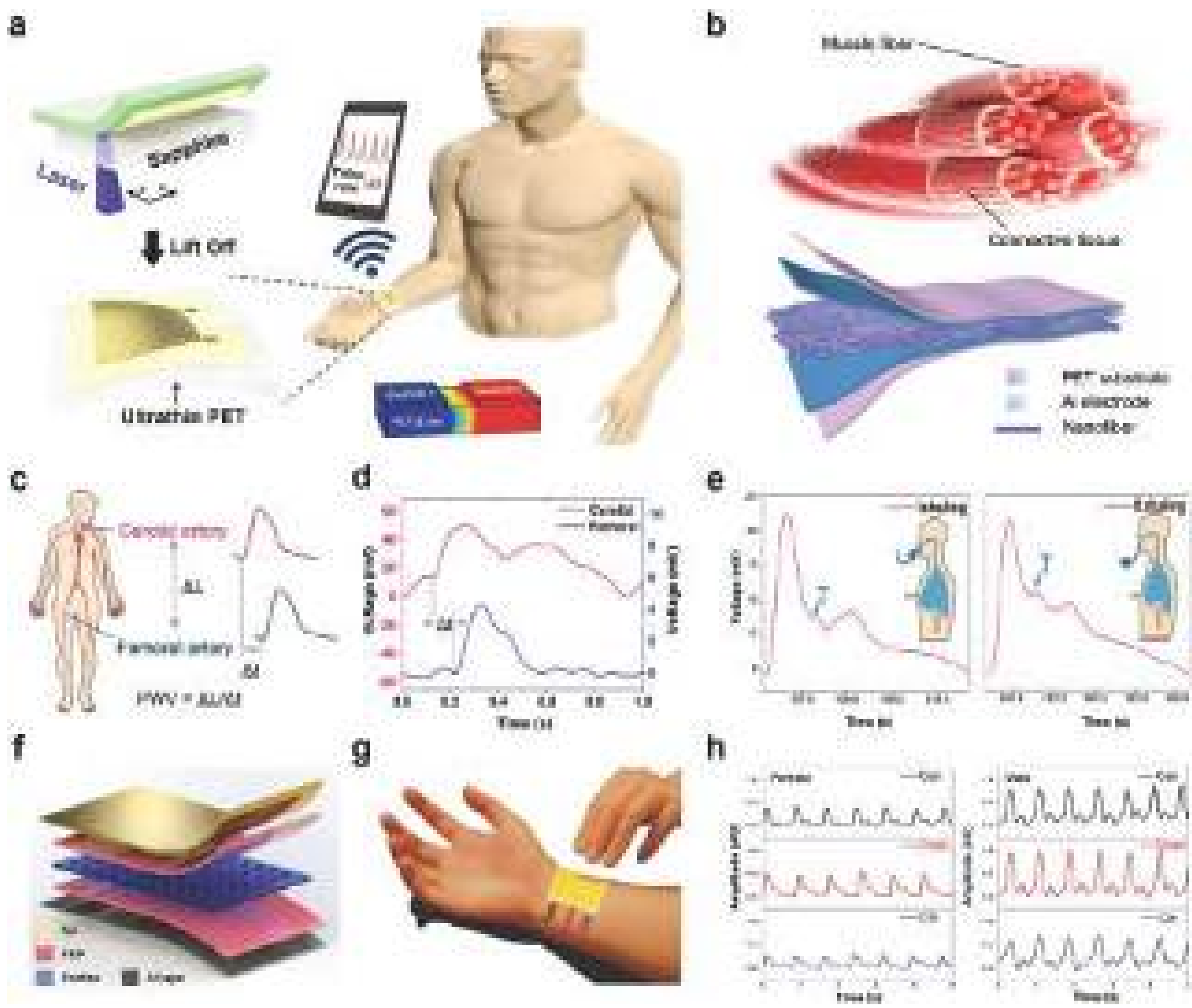

Fig. 11 PENGs for vital signs sensing. (a) Schematic illustration of the fabrication process for a self-powered pressure sensor based on PZT film. Reproduced with permission from ref. 138 Copyright 2017, Wiley. (b) Schematic of muscle fibers reinforced by connective tissue and the as-fabricated muscle fiber-inspired piezoelectric textile. Reproduced with permission from ref. 548 Copyright 2021, Wiley. (c) Schematic illustration of the calculation of PWV. (d) Carotid-femoral pulse waveform used for PWV calculation. (e) Brachial artery pulse waveforms during inhaling and exhaling periods in a deep breathing process. Reproduced with permission from ref. 149 Copyright 2019, Wiley. (f) The schematic diagram of the pulse sensing device using the FEP/Ecoflex/FEP sandwich-structured piezoelectret film. (g) Mimicking the Traditional Chinese Medicine pulse palpation acquisition scheme of a real doctor by using three fingers with the pulse sensing system at the Cun, Guan, and Chi positions. (h) Measured typical pulse waveforms at the Cun, Guan, and Chi positions for a female and male volunteer, respectively. Reproduced with permission from ref. 137 Copyright 2018, Wiley.

a reference for the diagnosis of related diseases.[4,52,545,546] PENG, as an active self-powered sensor, has been extensively developed to monitor human vital signs (including pulse frequency,[137–145] respiration rate,[146–151] blood flow,[154,155] *etc.*) in a continuous, real-time, and non-invasive manner. Among them, the most common application is pulse measurement, as continuous monitoring of arterial pulsation is important to detect the early onset of cardiovascular diseases and to assess a person's health. On this ground, Park *et al.* developed a PZT film-based PENG used for real-time pulse wave monitoring.[138] As shown in Fig. 11a, the PZT film was exfoliated from a rigid Sapphire substrate using the LLO technique, and then transferred to an ultrathin PET substrate. When the completed ultrathin PENG was attached to the human wrist, the pulse information could be detected and transmitted to a smartphone for diagnosis. So far, there have been many reports on the measuring of pulse frequency *via* a number of different piezoelectric materials. Indeed recently, novel piezoelectric materials have also been developed in this sense: Dai *et al.* reported a 2D layered van der Waals indium selenide device with a large piezoelectric response and long-term mechanical durability, which could be utilized to monitor pulse frequency and respiration rate in real-time by attaching the devices onto the wrist and chest.[547] Su and his collaborators developed a muscle-fiber-inspired nonwoven piezoelectric textile with tunable mechanical properties, which can be used for wearable physiological monitoring, as shown in Fig. 11b.[548] To extract more information from the pulse waveform, Chen *et al.* developed a flexible PENG based on a single-crystalline group III-nitride thin film.[149] The as-fabricated PENGs were attached at different pulse sites, each pulse waveform was recorded simultaneously allowing an extrapolation of pulse wave velocity (PWV), as schematically illustrated in Fig. 11c. From the test results, a time delay between the carotid and femoral artery of about 98 $\pm$ 5 ms can be extrapolated from the signal, representing a corresponding pulse wave velocity of 4.5 m s^{-1}, with an estimated path length of 44 cm (Fig. 11d). Alongside this application, given the high sensitivity of the III-nitride thin film, this sensor could also be used to monitor the breathing rate and detect subtle differences in pulse waveforms during inhalation and exhalation (Fig. 11e). Chu *et al.* constructed a high-precision PENG with a sandwich-structured piezoelectret, which could be used to detect weak vibration patterns of the human radial artery, even in close proximity.[137] Two layers of fluorinated ethylene propylene (FEP) film and a middle layer of Ecoflex film (with circular holes) were bonded together to form a sandwich structure, with two opposite electrodes connected to form a PENG, as schematically illustrated in Fig. 11f. After a corona charging process, the equivalent piezoelectric coefficient d_{33} of the PENG was characterized as about 4100 pC N^{-1}, equipping the sensor with ultra-high sensitivity. As a result, a three-channel pulse sensor array could be used to mimic the three fingers used by a Traditional Chinese Medicine doctor in Traditional Chinese Medicine and allow to record pulse waves at the corresponding mapping points on the radial artery, namely the Cun, Guan, and Chi positions (Fig. 11g). Using the recorded waveforms, distinct human organs could be differentiated by observing specific frequency, amplitude and shape differences, a helpful tool for medical diagnosis (Fig. 11h). In short, due to its outstanding sensitivity and ultrafast responsiveness, the PENG has shown great advantages in monitoring vital signs linked to dynamic weak physical deformation, of paramount importance to provide rich data insight for subsequent personalized medical diagnosis.

5.3 PENGs for biomechanics sensing

Human body-associated biomechanics signals could also provide essential healthcare information.[549] To date, biomechanical sensing has been monitored by various PENGs through materials design, micro/nano structural design, and various manufacturing approaches.[550] For example, Yao *et al.* recently reported a wearable PENG with high piezoelectric responsiveness and compliance consisting of 3D printable piezoelectric nanocomposites which were made through additive manufacturing.[386] As shown in Fig. 12a, the author inserted the 3D printed flexible piezoelectric lattice with microarchitectures into a boxing glove, to measure the punch force, with the obtained signals transmitted to the terminal through an embedded WiFi module. As a result, when the glove hit the wall with a direct hit or a right hook, the corresponding spatial distribution of the force could be plotted and displayed on the receiving device through the electrical signal obtained by the designated PENG electrode (Fig. 12b). This application demonstrated the ability of PENG to monitor the spatially resolved and time-resolved mapping of the reaction force applied to the hand during boxing activities. However, most of the reported PENGs can only detect normal pressure or bending strain, and almost no shear force. To this end, Chen *et al.* proposed a three-axial PENG device with which to detect both normal compressive force and horizontal shear force.[257] As illustrated in Fig. 12c, the proposed PENG was composed of imprinted P(VDF-TrFE) micropillars and a PDMS truncated pyramid bump. Since the bump was attached to the micropillars, when a shear force was applied to the bump, the torque would cause different forces in different areas of the micropillars (Fig. 12d). Consequently, the three-axial force exerted on the sensor could be estimated from the outputs of the PENG array. Further implementing an arrayed sensors approach, Fuh and his colleagues successfully demonstrated foot pressure mapping based on a sensor array composed of PENGs, as shown in Fig. 12e.[551] Most recently, a complex 3D piezoelectric microsystem was fabricated by converting 2D patterns of electrodes and thin films of piezoelectric polymers to 3D frameworks (Fig. 12f).[266] On this basis, many mechanical stimuli such as pressure, normal force, stretching, and bending could all induce output, making this 3D structure more responsive. When the device was subjected to pressure at different areas (Fig. 12g) and different levels (Fig. 12h), the device exhibited different performances. Compared with the 2D alternative method, the 3D geometry offered greater freedom of deformation and could respond to external mechanical stimuli more comprehensively, creating opportunities for an ever-growing

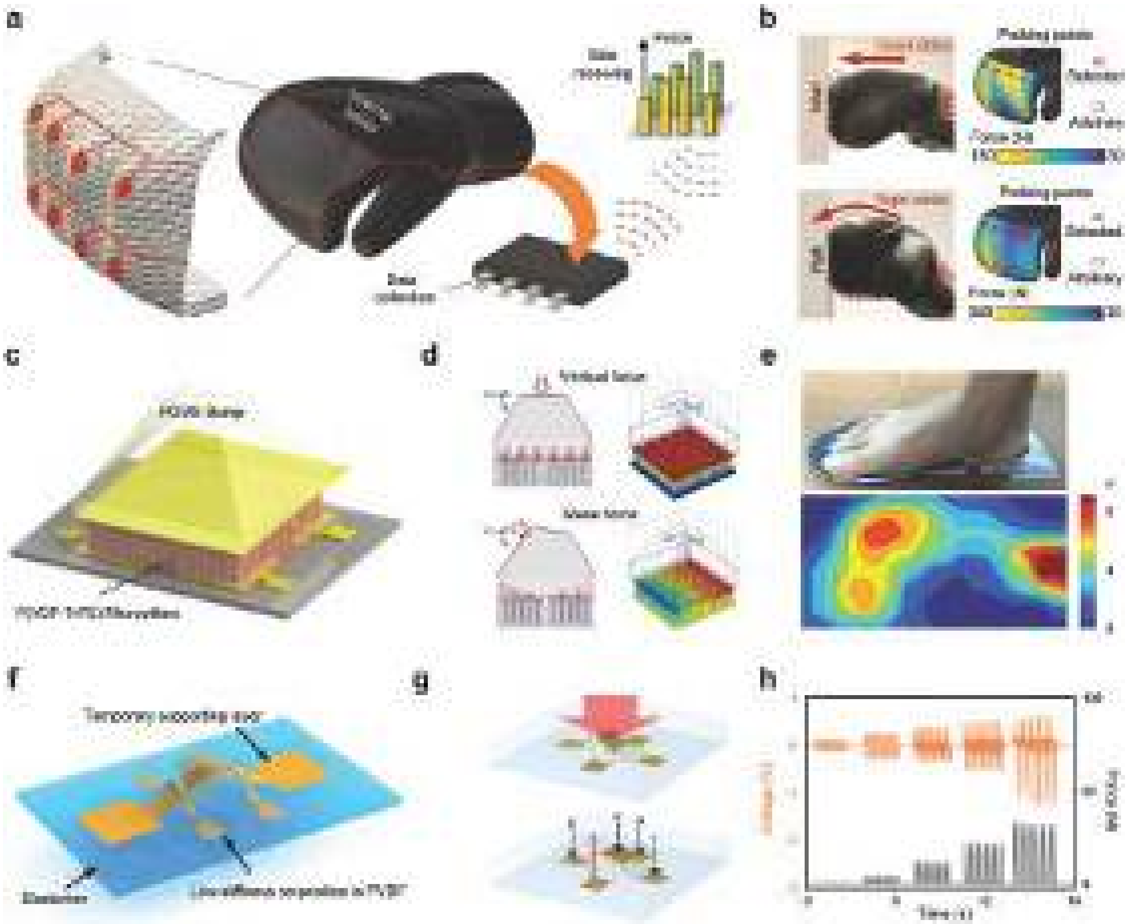

Fig. 12 PENGs for extracorporeal biomechanical sensing. (a) Schematics of a wireless self-sensing boxing glove based on a 3D printed flexible piezoelectric lattice with stretch-dominated microarchitectures. (b) Optical image of the glove and spatial distribution of force magnitudes when it hits a wall by a direct strike and right hook, respectively. Reproduced with permission from ref. 386 Copyright 2019, Wiley. (c) Schematic diagram of a flexible piezoelectric three-axial tactile sensor based on imprinted P(VDF-TrFE) micropillar array. (d) The simulation of the three-axial tactile sensor under normal force and shear force. Reproduced with permission from ref. 257 Copyright 2018, IOP publishing. (e) Photograph of integrated self-powered foot pressure sensor arrays and two-dimensional contour plot mapping of pressure potential from the objects. Reproduced with permission from ref. 551 Copyright 2017, Nature Publishing Group. (f) 3D mesoscale piezoelectric frameworks with ultra-low stiffness. (g) Schematic illustration of the device under pressure applied over a large area (top) and a small area (bottom). (h) Time-domain output voltage (orange curve) with different external forces (dark grey curve) applied over a squared area. Reproduced with permission from ref. 266 Copyright 2019, Springer Nature.

array of design scenarios. In addition, when the PENG was attached to the throat (skin), the faint vibration of the throat could be detected and used to detect deglutition and even its associated sound. Yan *et al.* adopted the serpentine mesh design concept to construct a stretchable micromotion piezoelectric sensor. Combining the detected sensed data with machine learning algorithms, the researchers devised a system that allowed human voice recognition.[552]

The mechanical evaluation of soft biological tissues and organs is another important application of PENGs, with a significant impact on the clinical diagnosis and treatment of diseases. Dagdeviren *et al.* introduced an ultra-thin stretchable network composed of PZT nanoribbons to measure the viscoelasticity in the near-surface regions of the epidermis.[152] As shown in the schematic diagram in Fig. 13a, the serpentine metal electrode linked to the PZT nanoribbon was supported by a thin elastomer. The resulting device could be directly coupled to the surface of the skin or other biological tissues through the mere passive use of van der Waals forces. When the device was attached at the site of a lesion on the forearm, as shown in Fig. 13b, the corresponding modulus mapping could be obtained (Fig. 13c). This approach provided a non-invasive and rapid measurement for dermatologic investigation. Recently, this ultra-thin piezoelectric microsystem was further improved by Yu *et al.* and engineered into a needle-shaped form (Fig. 13d), so that it could be injected or directly fixed onto a conventional biopsy needle to achieve real-time quantitative monitoring of tissue modulus (Fig. 13e).[153] As illustrated in Fig. 13f, the developed probe could be percutaneously inserted into various biological tissues, such as the spleen, liver, kidneys, lungs, and even cutaneous fat, providing information on the corresponding modulus and demonstrating the ability of such a miniaturized modulus-sensing device to harness the piezoelectric effect. Inspired by the kirigami concept, Sun *et al.*

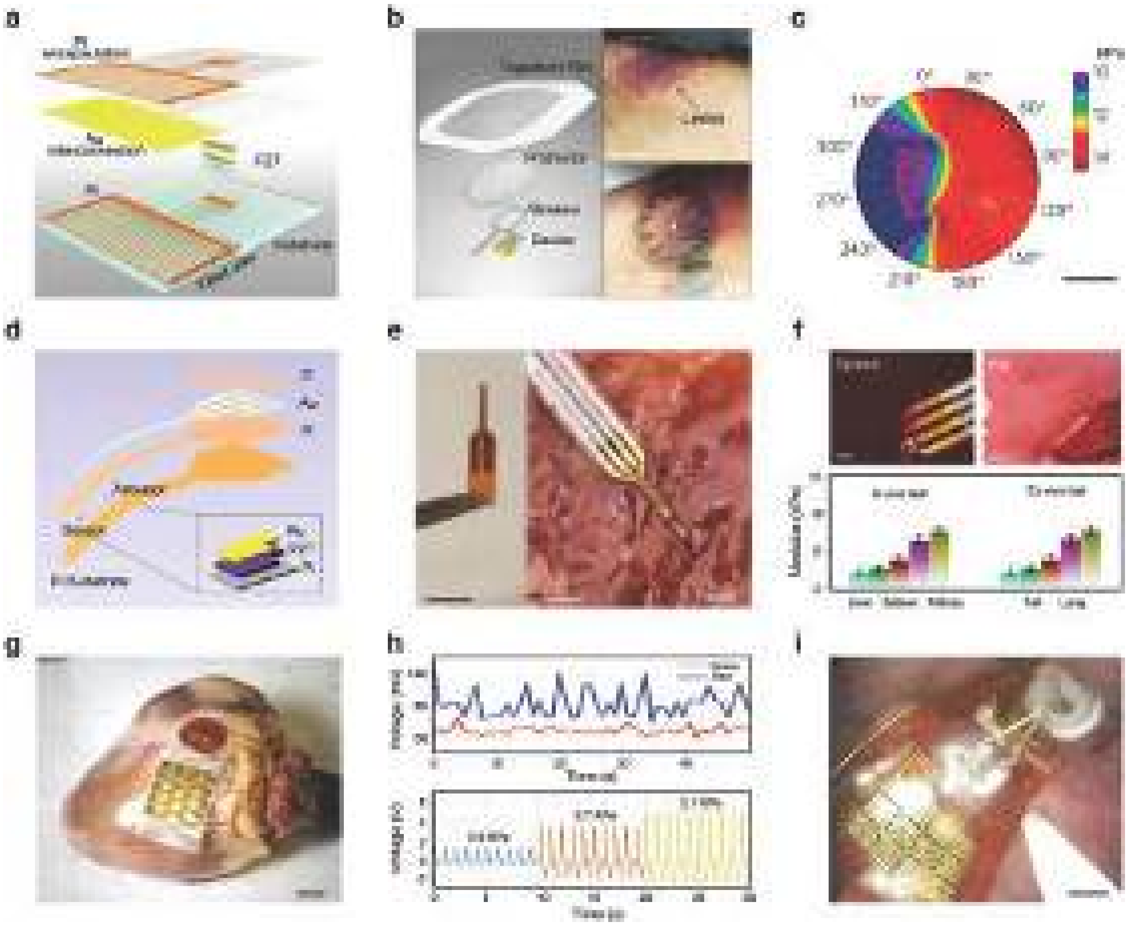

Fig. 13 PENGs for intracorporeal biomechanical sensing. (a) Exploded-view schematic illustration of the modulus sensor based on nanoribbons of PZT. (b) Schematic illustration of a system and photograph of a forearm without and with a mounted device. (c) Representative data from experiments. Scale bar, 2 cm. Reproduced with permission from ref. 152 Copyright 2015, Nature Publishing Group. (d) Schematic illustrations of a device based on ultrathin PZT actuators and sensors. (e) Optical images of said device (left, Scale bar, 5 mm) and the device placed on a biological tissue (right, Scale bar, 2 mm). (f) Optical images and results of *in vivo* modulus measurements on a live rat and *ex vivo* results of the same organs after explanation. Scale bars, 1 mm. Reproduced with permission from ref. 153 Copyright 2018, Springer Nature. (g) Potential application of implantable sensors on tissue (pig heart) surface. Scale bar, 1 cm. (h) The voltage output of the sensor on the pig heart under different pressures with the pulse and heartbeat-like inputs on the air-driven platform (bottom). Reproduced with permission from ref. 120 Copyright 2019, Wiley. Voltage output graphs of before and after milk ingestion from *in vivo* evaluation in a Yorkshire swine model (top). Reproduced with permission from ref. 553 Copyright 2017, Springer Nature. (i) Photograph of the PZT-based sensor with percutaneous endoscopic gastrostomy tube inside the stomach. Scale bar, 1 cm. Reproduced with permission from ref. 553 Copyright 2017, Springer Nature.

introduced a stretchable strain monitoring system based on a piezoelectric PVDF film used for self-powered cardiac monitoring.[120] When the device was mounted on a pig's heart (Fig. 13g), the sensor could monitor heart activity by yielding varying voltage outputs, as shown in the lower part of Fig. 13h. In another work, Dagdeviren and his colleagues developed a flexible piezoelectric device based on a PZT ribbon, which could be used to detect the mechanical deformation of the stomach.[553] As shown in Fig. 13i, the designed sensor was conformally attached to the stomach and responded to the stomach peristaltic action by yielding varying voltage outputs, linked to digestive activity, as depicted in the upper part of Fig. 13h. This approach demonstrated the great theragnostic potential of piezoelectric devices in gastrointestinal motility disorders. To sum up, PENGs have been extensively developed for mechanical sensing based on the advantages of excellent sensitivity, high signal-to-noise ratio, and good stability. Their fast response rates make them exploitable for the detection of dynamic mechanical-related stimuli. Meanwhile, their self-powered features provide a convenient implementation for *in vivo* sensing of implantable devices.

5.4 PENGs for sweat sensing

Sweat analysis can shed light on an individual's health. Obtaining health information through non-invasive and real-time continuous monitoring of sweat is one of the development priorities of wearable sensing.[554] In addition to the physical sensing described above, PENGs can also be used as chemical sensors, for instance, Saravanakumar *et al.* developed ZnO microwires with which to detect pH changes.[555] Furthermore, piezoelectric materials can also be used for sweat monitoring after being modified by related enzymes.[556] Han *et al.* presented a self-powered wearable noninvasive PENG array for sweat analysis based on enzyme/ZnO nanoarrays.[557] In this device, four piezo-biosensing units were modified with lactate oxidase, glucose oxidase, uricase, and urease, to respectively detect lactate, glucose, uric acid, and urea (Fig. 14a). Two types of device structures were developed, horizontally aligned (Fig. 14b) and vertically aligned (Fig. 14c), depending on the arrangement of ZnO nanorods.[558] Compared with the vertically aligned sandwich structure, the horizontally aligned structure was more likely to come into contact with sweat due to its larger exposed area. From the side view of ZnO nanowire arrays (Fig. 14d – left), the nanowires hold an average length of ~12 μm, and the enzyme-modified ZnO nanowires can be seen crossing on the interdigital electrodes (Fig. 14d – right). When the analyte appeared and the enzymatic reaction occurred, the surface carrier density increased, and then the strong piezoelectric screen effect caused the output piezoelectric voltage to decrease, as illustrated in Fig. 14e. As a result, when the designed PENG was attached to the forehead of a runner, the physiological status of human subjects including lactate, glucose, uric acid, and urea levels, could be continuously monitored by analyzing the runner's sweat (Fig. 14f). Recently, Liu *et al.* demonstrated a piezo-electrocatalytic sensor based on the charge transfer between the strained ZnO nanostructures and ascorbic acid,[559] promising for monitoring ascorbic acid in sweat, tears, and saliva. Another application of this technology can be envisioned using ascorbate acid oxidase and alcohol oxidase to modify ZnO nanowires; these device variations can be used to mimic taste buds and achieve an artificial gustatory system,[560] which could further extend the application of PENGs to the field of chemical sensing.

5.5 Outlook on PENGs for sensing

Due to the outstanding electromechanical coupling characteristics of piezoelectric materials, the PENG has been widely engineered to yield a variety of active sensors, with the feasibility of various scenarios having been proven both *in vitro* and *in vivo*. Recent advances made by PENGs exhibit tremendous potential in the field of flexible wearable electronics. To further promote the practical application of PENGs, several challenges need to be overcome: (1) piezoelectric potentials generated by piezoelectric materials are easily neutralized by the presence of free charge in the surrounding environment, making it difficult to detect static force and limiting the application of PENGs. Further performance improvements of PENGs in the realms of sensitivity, reliability, and detection range are also required; (2) most of the piezoelectric materials are also pyroelectric (*e.g.*, PZT, $PbTiO_3$, PVDF), meaning that when using wearable PENGs, the individual's body temperature could also affect the output of devices made with such materials, which in turn could affect their sensing performances. In subsequent studies, the effect of temperature on electrical performance needs therefore to be considered; (3) the majority of currently developed PENGs adopt a sandwich structure of thin-film and planner electrode or a laminated structure of thin-film and interdigital electrode. In both instances the functional layer or electrode comprises a dense structure, leading to poor air permeability of the devices. Improving the air permeability of such devices without damaging their performance requires further study; (4) three-axis force sensing is primarily achieved

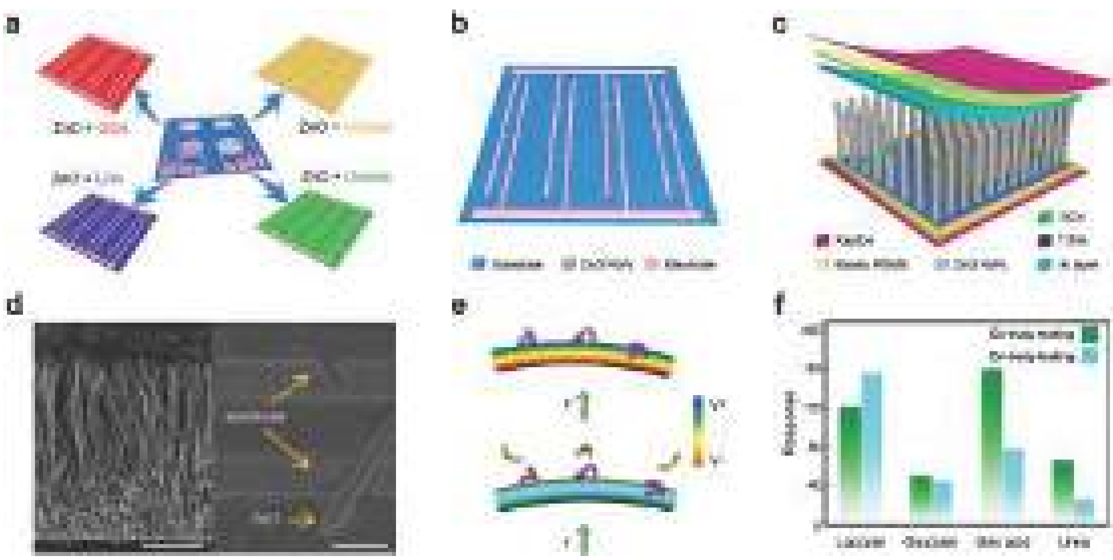

Fig. 14 PENGs for sweat sensing. (a) Four self-powered wearable noninvasive piezo-biosensing units for sweat analysis. (b) Lateral device architecture illustration of the self-powered wearable noninvasive electronic skin. Reproduced with permission from ref. 557 Copyright 2017, American Chemical Society. (c) Vertical device architecture illustration of the self-powered wearable noninvasive electronic skin. Reproduced with permission from ref. 558 Copyright 2016, Elsevier. (d) SEM image of ZnO nanowire arrays and single ZnO nanowire bridging two Ti electrodes. Scale bars, 10 μm. (e) The piezoelectric potential of modified ZnO nanowire in pure water created by applied deformation. (f) The comparison of the response between on-body and ex-body testing. Reproduced with permission from ref. 557 Copyright 2017, American Chemical Society.

using 3D structure PENG design, which represents a trade-off between device size and performance, and this limitation needs to be addressed. To achieve a flexible and stretchable device, the optimization of the material working mode may be a feasible solution to avoid sole reliance on 3D structure designs; (5) in the field of PENG use for chemical sensing, the device's limited lifetime remains a challenge given the rapid exhaustion of utilized enzymes. Moreover, the effect of temperature on enzyme activity and stability also needs to be further investigated in-depth; (6) at present, most of the reported PENG-based sensing performances are obtained under specific experimental conditions. To achieve accurate quantitative measurements of any object under any conditions, a reliable calibration system for self-powered PENG is urgently needed; (7) PENG can respond to almost all external-force related stimuli, so when there are multiple different stimuli at the same time, decoupling the output and detecting the source of each stimulus is of paramount importance; (8) contact electrification, including friction and contact separation, is often mistaken for piezoelectric output,[561] and within the field of PENG used in sensing applications, clearly distinguishing between the triboelectric output and the piezoelectric output is fundamental, especially in the presence of a state change during material contact and separation; (9) PENG itself is self-powered, but its signal acquisition circuit still requires an external power supply. To produce a fully self-powered sensing system that includes self-powered detection and signal processing, further research is needed to enable a full system integrating with PENGs being used as active sensors, energy harvesters, and energy storage components, all within a single system.

6. PENGs for therapy

In addition to energy harvesting and sensing, another important application of PENGs is therapy. Since electric fields have been shown to affect cellular activities including migration, proliferation, differentiation and mechanical transduction, exploring their potential therapeutic applications has been an important scientific focus in recent years.[562–566] As it happens, due to the direct piezoelectric effect, piezoelectric materials can generate an electric field when stimulated by human body motions, vibrations or ultrasounds. Moreover, these materials can also transform electrical signals into mechanical deformations *via* the inverse piezoelectric effect, thereby further enhancing their ability to modulate cellular activities. Hence, various piezoelectric materials have been widely explored for different biomedical applications, such as cell activity modulation, wound healing, drug delivery, neural stimulation, and so forth.

6.1 PENGs for cell activity modulation

As the basic unit of biological structure and function, cell activity regulates the expression of tissue-specific genes. Many efforts have been devoted to the regulation of cell activities, in which electrical or electromechanical stimulation is a proven and effective approach. The PENG has aroused a lot of attention in the field of cell behavior regulation,[567–574] as it is favored as a self-powered source and avoids risks associated with battery replacement and secondary implantation. For instance, Hoop *et al.* demonstrated that the piezoelectric charge generated by PVDF under ultrasonic wave stimulation could induce the generation of neurites (neuronal cell projections).[170] As illustrated in Fig. 15a, the effect on two different molecular signaling pathways of PC12 differentiation: the MAPK/ERK pathway and the cAMP pathway, were studied. When PC12 cells were seeded on the pre-treated β-PVDF membrane, the charge generated by PVDF in response to wireless dynamic stimulation of external ultrasonic transferred directly to the cells and affected their behavior (Fig. 15b). In detail, the piezoelectric charge seemingly activated the cAMP-dependent pathway in cells, resulting in increased differentiation of PC12 cells. A similar result was also confirmed by Liu and his colleagues.[184] As shown in Fig. 15c, piezoelectric $BaTiO_3$ nanoparticles were used to convert ultrasonic energy to an electrical signal *in situ*, inducing the differentiation of the targeted neural stem-like cells. Moreover, the surface nanotopography of biomaterials has been shown to promote cell adhesion, which in turn activates downstream cell signaling and regulates cell behavior through cell mechanical transduction pathways.[575] On these grounds, Zhang *et al.* showed the combinatory effect of nanotopography and bioelectricity in regulating cell behaviors through exposure to piezoelectric materials' activity.[162] In their experiments, a PVDF device with a nanostripe array structure was used as the substrate to culture living cells, as shown in Fig. 15d. Given the different cell traction forces, the PVDF nanostripe would generate different piezoelectric potentials, ranging from a few microvolts to a few millivolts (Fig. 15e), with both the nanoscaled stripe structure and the generated potential modulating the cell activities. Similarly, Murillo *et al.* reported a PENG based on 2D ZnO nanosheets, which could be used to stimulate macrophage motility.[161] As shown in Fig. 15f, the plasma membrane of the cultured cells was in close contact with the ZnO nanosheets, and the emitted projections were also firmly attached to the nanosheet. Alongside this research, Kim *et al.* designed a series of topologically structured piezoelectric scaffolds through electrospinning to determine the response of neural cells (Fig. 15g).[166] The results confirmed that the radially aligned pattern (p-RA) and repetitive gradient align/random pattern (rp-gAR) composed of PVDF, could preferentially induce neurite outgrowth. In general, PENGs can easily inherently respond to external stimuli and convert these stimuli into electrical signals to regulate cell behavior *in situ*, providing a therapeutic solution with a simple structure and easy operability. In addition to these advantages, the surface topography of the piezoelectric material can also be easily controlled according to specific needs, meaning that PENGs have bright prospects in the electrical self-stimulation arena and in inducing therapeutic cellular activities.

6.2 PENGs for wound healing

With the development of modern biomedicine and medical technology, electrical stimulation used to imitate endogenous

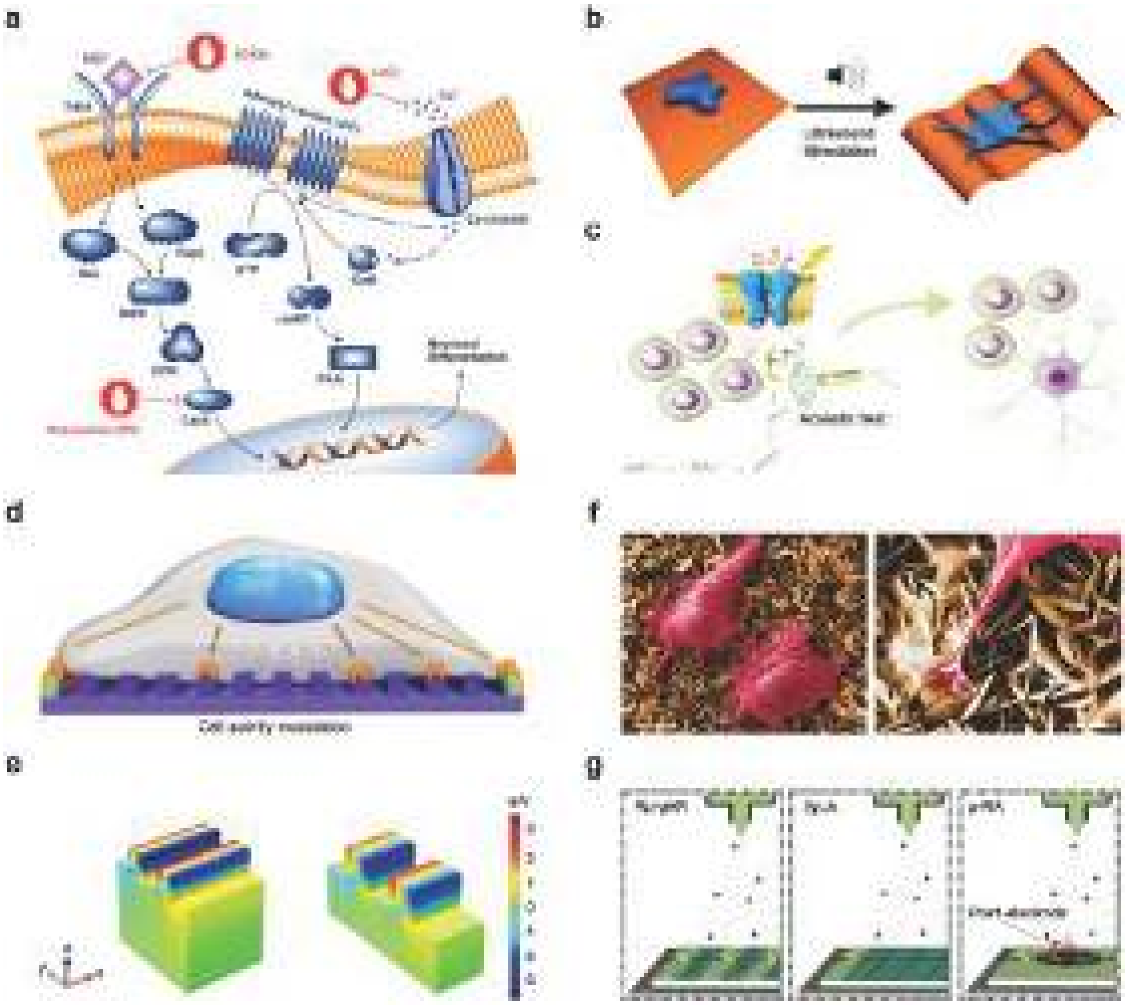

Fig. 15 PENGs for cellular activity modulation. (a) Illustration of intracellular pathways affecting PC12 differentiation. (b) Schematic illustration on the ultrasound stimulation of the piezoelectric β-PVDF membrane, inducing neuronal differentiation of PC12 cells. Reproduced with permission from ref. 170 Copyright 2017, Nature Publishing Group. (c) Schematic illustration of a highly controllable micromotor to induce the differentiation of the targeted neural stem-like cell. Reproduced with permission from ref. 184 Copyright 2020, Wiley. (d) Inherent cell forces of living cells grown on the surface of PVDF with nanoscaled stripe arrays. (e) Electrical simulation results of PVDF with different stripe arrays. Reproduced with permission from ref. 162 Copyright 2019, Wiley. (f) SEM images of morphology and NG-cell interaction. Scale bars, 5 μm (left) and 2 μm (right). Reproduced with permission from ref. 161 Copyright 2017, Wiley. (g) Simplified generation mechanism of differently spun membranes. Reproduced with permission from ref. 166 Copyright 2019, Wiley.

electric field and promote wound healing, has emerged as a novel and exciting therapeutic approach.[576–580] The basic schematic diagram of the electrical stimulation wound healing *modus operandi* is illustrated in Fig. 16a, the nanogenerator is attached to the wound, and the electric field generated between the two electrodes penetrates the dermis to imitate the endogenous electric field and promote wound healing. Recently, Wang *et al.* demonstrated the *in vitro* and *in vivo* wound-healing efficacy of a piezoelectric nanofibrous scaffold PENG.[176] The designed PENG made of polarized P(VDF-TrFE) nanofibers was cut into a 20 mm × 20 mm film and implanted in the subcutaneous thigh region of highly active Sprague Dawley rats (Fig. 16b). A linear motor system was used to gently pull the rat's leg to mimic its movement with the designed PENG able to generate a piezoelectric output of about 6 nA (Fig. 16c). The results of *in vitro* cell proliferation experiments further confirmed that piezoelectric scaffolds can be designed to provide the necessary electrical stimulation for wound healing (Fig. 16d). In another work, Bhang *et al.* developed a piezoelectric patch (PZP) composed of bidirectionally grown ZnO nanorods, and evaluated its therapeutic efficacy in dermal wound healing.[174] As schematically illustrated in Fig. 16e, the designed PENG consisted of aligned ZnO nanorods embedded in a PDMS matrix, with two Ag electrodes sandwiched on opposite sides. With a bending radius of 5 mm, the device could produce a 1.8 V average voltage (Fig. 16f). When the device was applied to a wound, the histological criteria of subcutaneous distance was then applied to quantitatively evaluate its therapeutic effect (Fig. 16g). To further verify the performance *in vivo*, the author attached the designed PENG

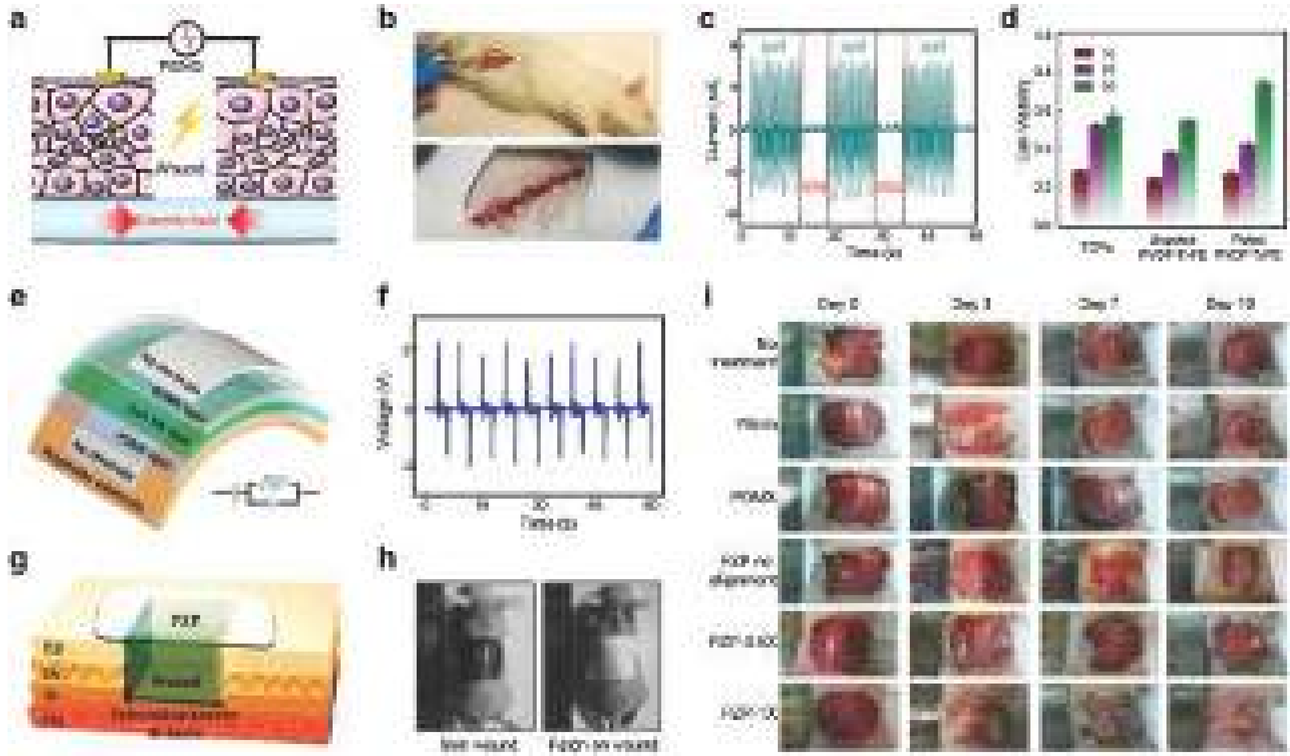

Fig. 16 PENGs for wound healing. (a) Schematic diagram of the wound-healing mechanism elicited under an endogenous electric field. (b) Image of electrospun P(VDF-TrFE) nanofiber scaffolds implantation in the subcutaneous thigh region of a rat (upper) and the implant site after suturing (lower). (c) Current output of the implanted scaffolds with intermittent pulling. (d) Bar chart of the proliferation of L929 fibroblast cells on excited tissue culture polystyrenes. Reproduced with permission from ref. 176 Copyright 2018, Elsevier. (e) Schematic illustration of the PZP modified with Ag electrodes to measure the piezoelectric voltage and current density generated by mechanical bending. (f) Quantification of piezoelectric voltage generated from a ZnO NR monolayer by mechanical bending. (g) Schematic diagram of the wound site with intersubcutaneous distance measurement. (h) Photographs of the skin wound on the back of a mouse (left), and a nine-layered PZP placed on the wound by dressing a transparent film over the PZP (right). (i) Representative skin wound photographs at 0, 3, 7, and 10 days after treatment. Reproduced with permission from ref. 174 Copyright 2017, Wiley.

to the wound on the back of mice as shown in Fig. 16h. From the results, it was obvious that the wounds with PZP-1X (9 layers of PZP, the filling rate is 95.2%) were almost completely closed after 10 days, which suggested that the designed PENG had the best effect on promoting wound closure and skin regeneration (Fig. 16i). In addition, many other piezoelectric materials such as electrospun PVDF/polyurethane membrane,[178] P(VDF-TrFE)/$BaTiO_3$ nanocomposites,[163] have also been confirmed to provide the effectiveness of piezoelectric electric fields in promoting wound healing and bone regeneration.[581,582] Recently, Liu and his colleagues have demonstrated that the piezoelectric PLLA nanofiber scaffold under applied force or joint load could promote chondrogenesis and cartilage regeneration due to the controllable piezoelectric charge.[583] The approach combining a biodegradable piezoelectric scaffold with controlled mechanical activation holds significant promise for applications in tissue regeneration and wound therapy.

6.3 PENGs for drug delivery

Driven by the huge clinical demand for targeted therapies, drug delivery systems have become a research hotspot in the field of biomedicine, hoping to bring more survival benefits to patients. In recent years, piezoelectric devices have proved to be an effective means of drug delivery, mainly including two working modes: one working mode is based on the inverse piezoelectric effect, which can control the precise deformation of piezoelectric devices through peripheral circuits, and enable quantitative release of drugs.[185,584–587] However, this model requires complex peripheral circuits and is mainly used for transdermal drug delivery, and thus difficult to be applied *in vivo*. The second mode is based on the direct piezoelectric effect, which can assist with drug release through electrical stimulation. Due to the small device size and easy control, this approach can be very conveniently used *in vivo*, especially for targeted drug release, with the help of a magnetic field. Recently, Chen *et al.* successfully developed a hybrid core-shell composite nanowire composed of P(VDF-TrFE) and FeGa for targeted drug delivery.[187] As shown in Fig. 17a, the nanowire was synthesized through a template-based wetting technique and electrodeposition method, in which P(VDF-TrFE) nanotubes were the shell, and FeGa was the core. This structure was also confirmed by the characterization using Atomic Force Microscopy (AFM) (Fig. 17b). Under the external magnetic field, the strain of magnetostrictive FeGa was transferred to the piezoelectric P(VDF-TrFE) shell through interfacial coupling, resulting in a change in the polarization state of P(VDF-TrFE),

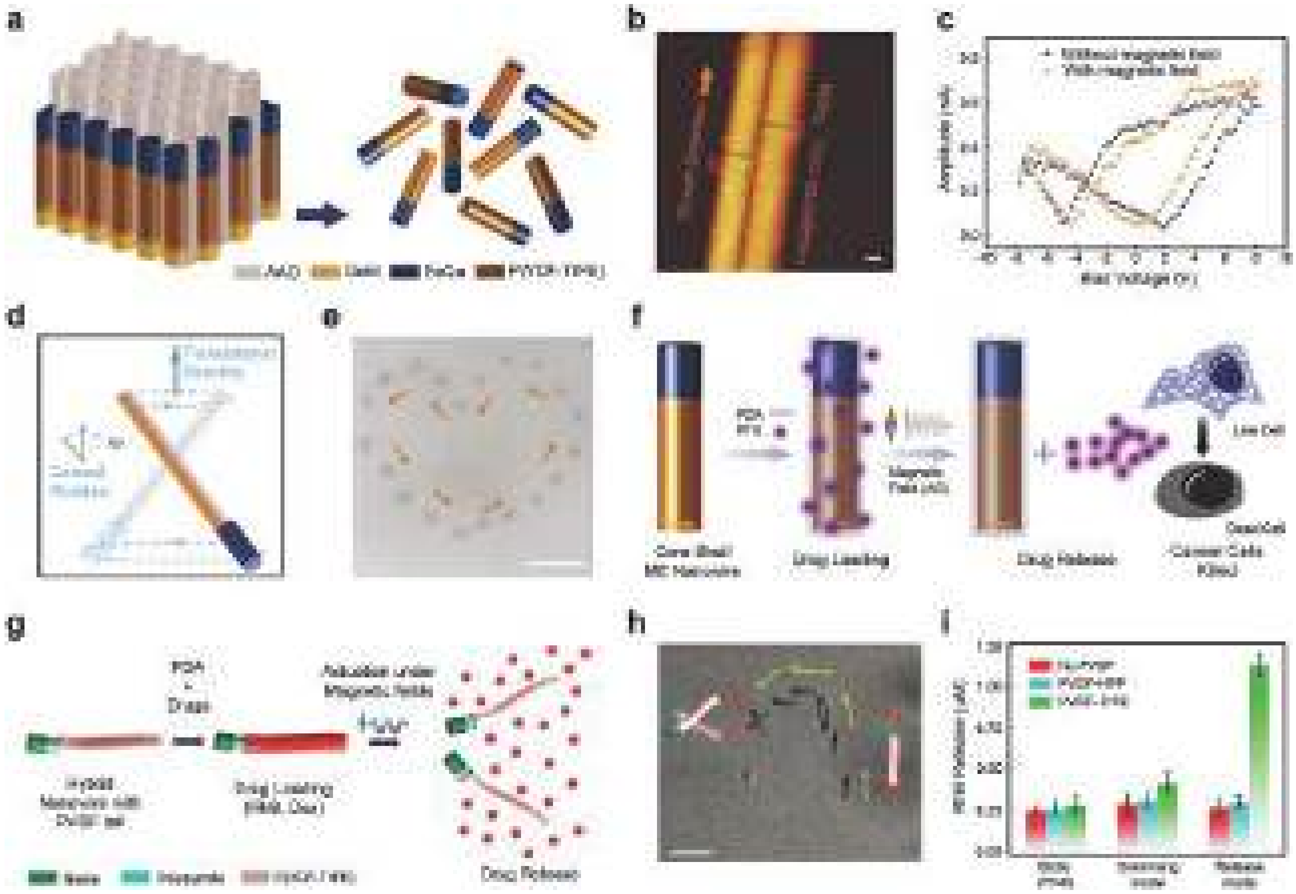

Fig. 17 PENGs for drug delivery. (a) Schematic diagram of FeGa@P(VDF-TrFE) core–shell nanowires. (b) AFM image of two nanowires with FeGa grown out of P(VDF-TrFE) nanotubes. Scale bar, 200 nm. (c) Piezoresponse amplitude loops obtained from the FeGa@P(VDF-TrFE) core–shell nanowires with and without magnetic field exposure. (d) Schematic illustration of the motion of a core–shell nanowire in a conical rotating magnetic field. (e) Manipulated nanowire to follow the shape of a heart. Scale bar, 20 μm. (f) Scheme showing on-demand drug delivery experiment. Reproduced with permission from ref. 187 Copyright 2017, Wiley. (g) Scheme depicting functionalization of hybrid nanoeels with PDA and drugs, followed by magnetically triggered drug release. (h) Time-lapse image showing a single hybrid nanoeel transition from a surface-walking swimming mode to a wobbling motion upon changing the parameters of magnetic fields. Scale bar, 15 μm. (i) Plot showing the release of RhB from NWs without PVDF, with P(VDF-HFP), and with P(VDF-TrFE), under different magnetic actuation. Reproduced with permission from ref. 186 Copyright 2019, Wiley.

which was verified by the butterfly curve (Fig. 17c). The generated transient changes of polarization state could be used as a trigger for drug release. At the same time, the asymmetry of the upper and lower parts of the nanowire leads to the translational force on the nanowire as illustrated in Fig. 17d. This approach could enable precise control of locomotion and maneuvering of the nanowires by varying the external magnetic fields. The authors demonstrated this by precisely steering the nanowire along a predefined trajectory, which could be used for targeted motion (Fig. 17e). On this basis, after the drug was adsorbed onto the surface of the core-shell nanowires, a rotating magnetic field could be used to manipulate the nanowires to the targeted lesion, and the drug was then released under an alternating current magnetic field (Fig. 17f). Similarly, inspired by the workings of electric eels, Mushtaq *et al.* proposed a multifunctional piezoelectric tailed nanoeel composed of P(VDF-TrFE), polypyrrole (Ppy), and nickel rings to use for drug release (Fig. 17g).[186] Unlike previous studies, Ppy nanowire heads decorated with nickel rings could provide magnetic actuation, thereby enabling more flexible control. As shown in Fig. 17h, the designed nanoeel could switch from a tumbling motion to a wobbling motion by simply changing the external magnetic field, and subsequently reaching the target position. The results of the drug release experiment clearly demonstrated that the amount of released drug can be effectively controlled by altering the applied magnetic field parameters (Fig. 17i), showing great potential for developing an efficient and controllable drug delivery platform.

6.4 PENGs for assistive physical therapy

Voice recognition can help people with hearing impairments to communicate and enable smart devices to bilaterally communicate with people, extremely important in our daily lives. Recently, due to their high sensitivity and fast response capability, flexible piezoelectric acoustic sensors[588] that mimic the basilar membrane of the human cochlea have attracted much attention as promising candidates in the field of voice recognition.[126,589] A typical diagram of such a sensing system is depicted in Fig. 18a, whereby a piezoelectric sensor converted sound signal into electricity which was subsequently analyzed *via* a machine learning process to finally implement sound recognition.[590] Han *et al.* reported a basilar membrane-inspired piezoelectric acoustic sensor, which exhibited four to eight times higher sensitivity than the conventionally used condenser sensor.[591] Fig. 18b illustrates the overall concept of the human cochlea, able to detect many resonance frequencies *via* the wide and thick nature of its basilar membrane which varies along the cochlear spiral. Inspired by this structure, the author designed a multi-channel asymmetric trapezoidal-shaped piezoelectric acoustic sensor made with PZT films, capable of resonance-based sensing and frequency tuning capabilities (Fig. 18c).[592] Compared to the reference sensor, the designed piezoelectric acoustic sensor exhibited a higher electrical signal amplitude and wider frequency band response, as shown in Fig. 18d. Utilizing this method, the original sensed voice was successfully reproduced as the testing results demonstrated in Fig. 18e. Furthermore, Kim *et al.* developed a high-performance PENG based on biodegradable chitin polymer, able to provide a high-fidelity paper-type speaker, and doubling in use as a microphone.[393] As shown in Fig. 18f, the chitin polymer material derived from squid pen material exhibited high optical transmittance (71%), and good crystal orientation (Fig. 18g), allowing the device to detect sound. The short-time Fourier transform spectrogram obtained from the voltage of the fabricated device further demonstrated its ability to replicate the original sound modulations, as displayed in Fig. 18h. In another work, an acoustic sensing platform composed of a P(VDF-TrFE) nanofiber mesh prepared by dynamic near-field electrospinning technology, showed a wide sensitivity bandwidth of 200–5000 Hz at hearing-safe sound pressure levels.[121]

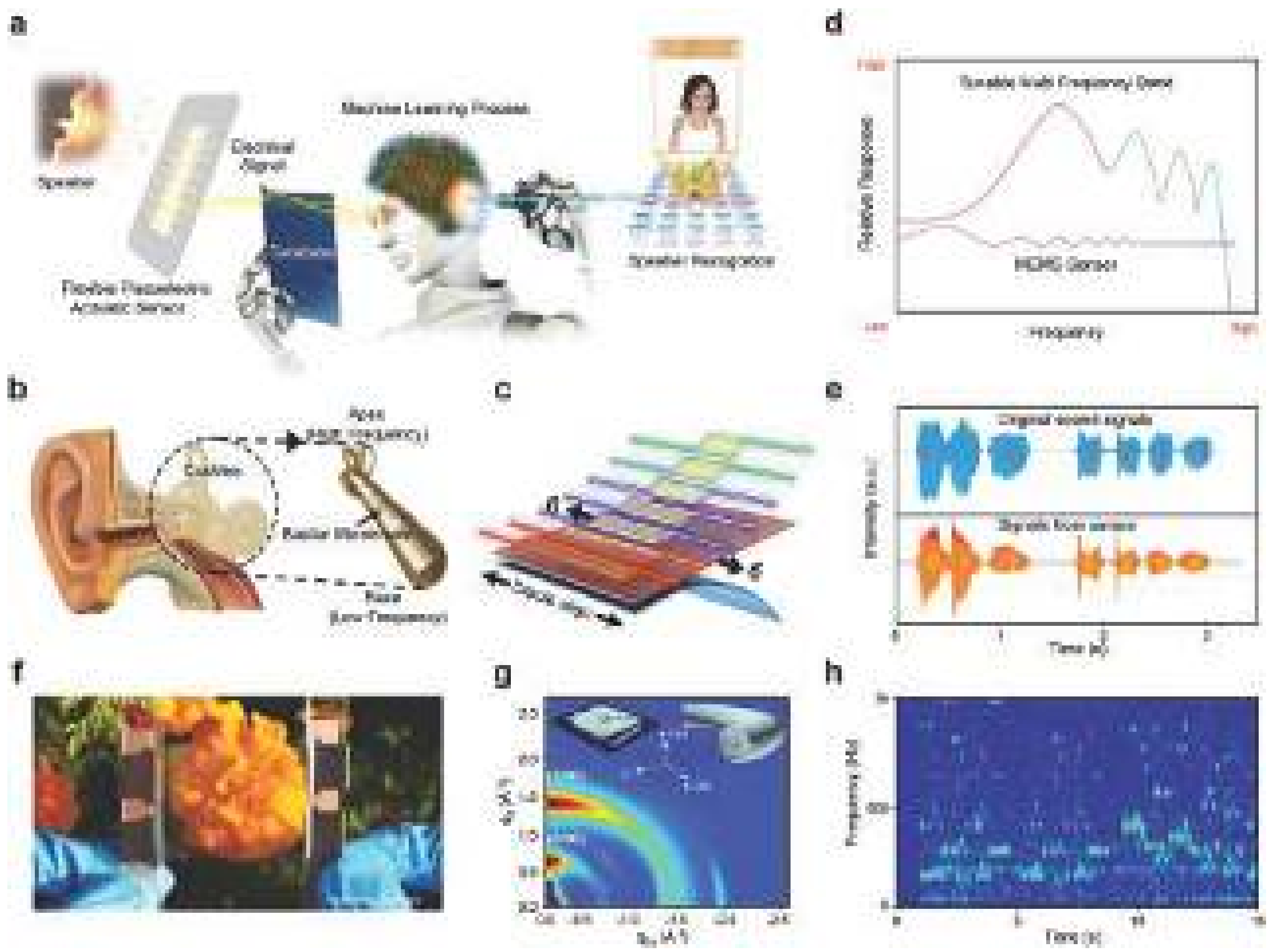

Fig. 18 PENGs for assistive physical therapy. (a) Overall schematic of the machine learning-based speaker recognition system. Reproduced with permission from ref. 590 Copyright 2018, Elsevier. (b) Schematic illustration of the basilar membrane-inspired flexible piezoelectric acoustic sensor. Reproduced with permission from ref. 591 Copyright 2018, Elsevier. (c) Schematic illustration of biomimetic multifrequency band control and mobile biometric authentication of miniaturized PMAS inspired by the resonant structure of the basilar membrane. Reproduced with permission from ref. 592. Copyright 2021, The American Association for the Advancement of Science. (d) Schematic graph of the highest electric signals yielded, selected from multi-channel, over the voice frequency band. (e) The sound signals of standardized female speech and corresponding signals recorded by the multi-channel flexible piezoelectric acoustic sensor. Reproduced with permission from ref. 591 Copyright 2018, Elsevier. (f) Digital photograph of the transparent chitin speaker sandwiched between two Ag NWs electrodes. Scale bar, 3 cm. (g) Crystal pattern of chitin thin film on Si substrate. (h) Short-time Fourier transform spectrogram of the transparent chitin speaker. Reproduced with permission from ref. 393 Copyright 2018, Elsevier.

In particular, with the integration of machine learning and recent advances in algorithms, the performance of voice recognition has been significantly improved, further promoting the practicality of PENG acoustic sensing applications.

6.5 PENGs for neural stimulation

Electrical stimulation has also been widely used to treat neurological diseases and restore lost functions as an important means of rehabilitation treatment and training. Examples of neurostimulation therapy include deep brain stimulation to relieve Parkinson's disease, cochlear nerve stimulation to restore hearing, and electrical potentials or currents cardiac applications to treat arrhythmias.[125,593–595] For implantable neurostimulation devices, PENGs can not only provide energy to extend the lifespan of the otherwise equivalent batteries but can also provide a directly independent power supply, attracting much attention. Generally, PENGs provide energy in two ways: one, collecting mechanical energy in the body, and the other, converting energy transmitted by ultrasound.[159,160,596] Hwang *et al.* demonstrated a self-powered brain stimulator made of indium modified crystalline $Pb(In_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PIMNT) thin film (Fig. 19a).[188] The designed flexible PENG could generate a maximum open-circuit voltage and short-circuit current of 11 V and 283 μA under bending motion. When the obtained electrical signal was connected into the M1 cortex in the brain to produce muscle contraction of the forelimb, the movement of the treated mouse's paws and forelegs could be clearly observed under electrical stimulation (Fig. 19b). To remove the external wiring, a wireless, leadless, and battery-free implantable neural stimulator was developed by Piech *et al.*[157] As the conceptual overview shown in Fig. 19c, the designed system incorporated a piezoceramic transducer, an energy-storage capacitor, and an integrated circuit, and directly fixed on a rat's sciatic nerve to achieve electrical stimulation. In this system, since the

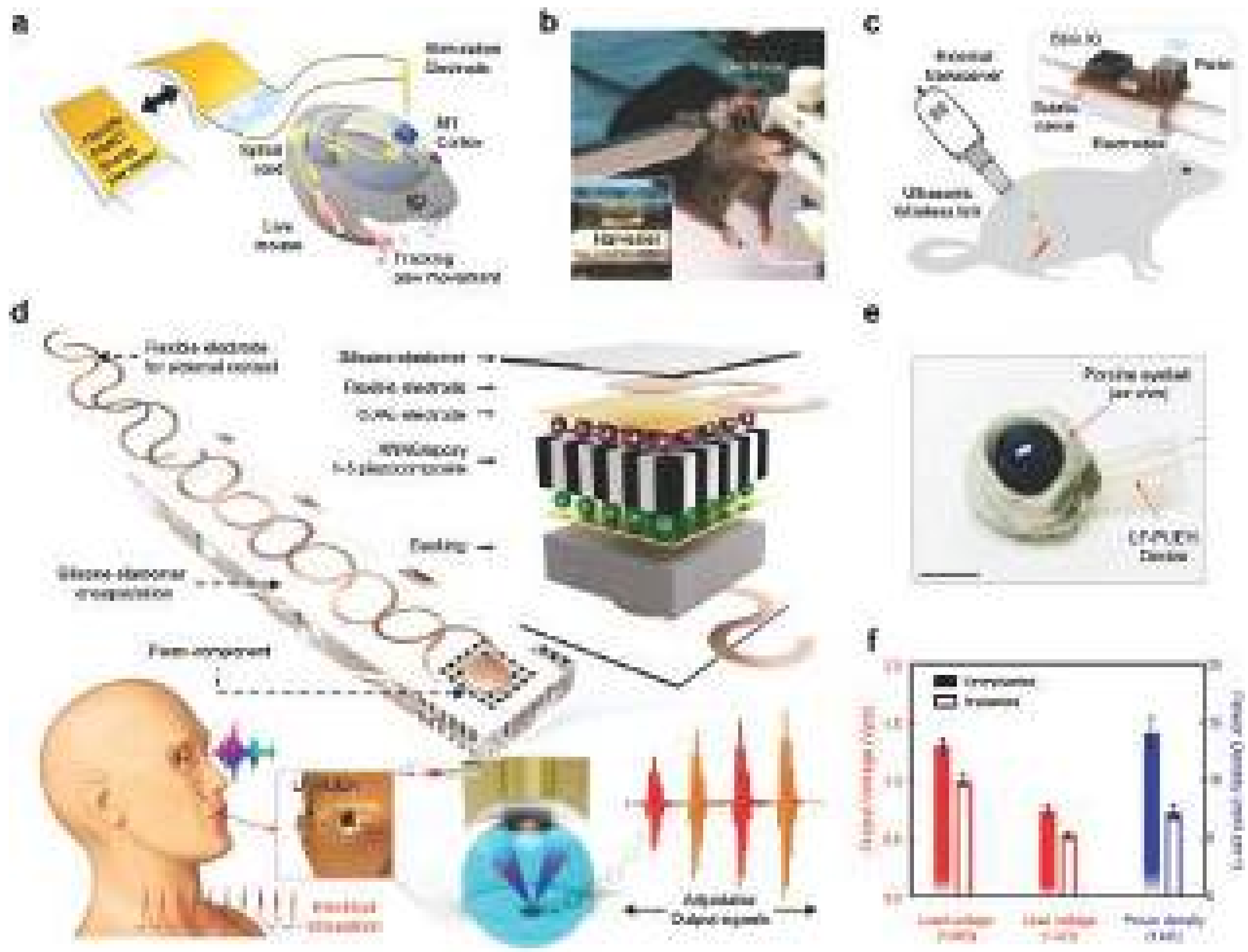

Fig. 19 PENGs for neural stimulation. (a) An illustration of the animal experiment involving brain stimulation of mice using a flexible PIMNT harvester. (b) A photograph of the flexible PIMNT stimulator connected to a bipolar stimulation electrode that is localized in the M1 cortex for electrical stimulation. Scale bar, 1 cm. Reproduced with permission from ref. 188 Copyright 2015, Royal Society of Chemistry. (c) Diagram of the battery-free implantable system used to stimulate the sciatic nerve of a rat. Reproduced with permission from ref. 157 Copyright 2020, Springer Nature. (d) Schematic and design of the mm-scale flexible ultrasound-induced wireless PENG for retinal electrical stimulation. (e) Optical image of the device implanted into an excised eyeball to mimic an implant scenario. Scale bar, 1 cm. (f) Comparison of output voltages and power densities of the device in implantation and non-implantation scenarios. Reproduced with permission from ref. 158 Copyright 2019, Wiley.

piezoelectric ceramic transducer could convert externally transmitted ultrasonic energy into electrical energy, electrical stimulation could be achieved without any external wires. Recently, Jiang *et al.* also fabricated an ultrasound-induced wireless PENG for potential retinal electrical stimulation through a modified dice-and-fill technique (Fig. 19d).[158] The core component of the designed PENG was an anisotropic 1–3 piezocomposite composed of high piezoelectricity lead-free ceramic $(K_{0.48}Na_{0.52})(Nb_{0.95}Sb_{0.05})O_3$-$(Bi_{0.4}La_{0.1})(Na_{0.4}Li_{0.1})ZrO_3$ pillar array, of parallelepipeds shape, as well as an insulating polymer filler (EPO-TEK 301). The entire device was encapsulated in silicone elastomer, with good flexibility and considerable output performance, reaching a maximum power density of 45 mW cm^{-2}. As shown in Fig. 19e, the as-fabricated PENG was implanted into an excised porcine eyeball to prove its feasibility of embedding into the body. Comparing the electrical output performance of *in vivo* and *in vitro* experiments, the voltage amplitude of the device implanted in the body decreased slightly (Fig. 19f), but the output current easily exceeded the average threshold required for retinal electrical stimulation, by cutting the electrode area. Overall, the designed ultrasound-induced wireless PENG showed great potential in the electrical stimulation of implantable biomedical devices.

6.6 Outlook on PENGs for therapy

In recent years, piezoelectric materials have gained significant attention in therapeutic fields due to their excellent mechanoelectrical properties, and many functional applications such as tissue regeneration, wound healing, drug delivery, and nerve stimulation have been developed in novel and interesting ways. It is noteworthy that the piezo-potential induced by strain or stress can induce a giant electric field at the micro-nano- scale, which has proven to be an effective means for piezo-catalysis. In combination with ultrasonic stimulation, piezo-catalysis has been successfully developed for sterilization,[597] tooth whitening,[598] and tumor sonodynamic therapy[599–605] *via* the generation of reactive oxygen species. However, many of the current experiments are either proof-of-concept or limited to *in vitro* experiments. To further advance the progress of implantable PENGs, more efforts should be targeted at resolving the remaining issues: (1) most of PENGs used in electrical therapy need to be implanted in the body, and biological safety is of top priority. Implantable devices have direct contact with interstitial fluids and biological tissues, so the toxicity of functional materials and metal electrodes, as well as the degradability of the entire device, are issues that need to be considered; (2) most current implantable piezoelectric devices are stimulated by ultrasound as this can penetrate biological tissues at a considerable depth, and the stimulation parameters can also be easily controlled by adjusting external ultrasound emitter. However, ultrasound also interacts with the device's surrounding tissues, which could be damaged by the heat caused by the cavitation effect. Hence, the relationship between ultrasonic-related parameters such as center frequency, output power, output time, and the overall performance of the device is worth studying; (3) in general, implantable devices are required to have a long enough working life to avoid the risk of secondary operations. There is no doubt that the lifespan of the device needs to be extended as long as possible, but further research is still needed to detect the effectiveness of the implanted device during use; (4) in the application of targeted drug release, understanding how to ensure that all drug-loaded particles reach the targeted area under the external control field is also another issue worthy of study. (5) Most recently, Piech *et al.* demonstrated a wireless neurostimulator that can accept external programmable current control commands to achieve programmed electrical stimulation and can also send feedback about whether electrical stimulation occurs to an external transceiver.[157] Such a bidirectional communication function is another important development direction of implantable devices.

7. Summary and perspectives

Over the past decade, PENGs have received extensive and in-depth research attention, and have been widely developed and applied in various applications. In this review, recent advances in wearable and implantable PENGs have been comprehensively summarized and discussed, including their material selection, structural design, and applications in human body associated energy harvesting, self-powered sensing, and electrical stimulation therapy. Given the latest progress made by PENGs, we schematically depict an intelligent closed-loop system to construct an (IoMT) paradigm for the next-generation personalized healthcare, as illustrated in Fig. 20. This smart system includes high-performance PENG-based sensors that can detect a variety of physiological signals, such as pulse, respiration, temperature, blood oxygen, *etc.* These physiological signals could be then directly transmitted to the hospital or medical center through 5G network. After data analysis is completed, the clinician could send feedback to the patient to guide treatment and even remotely control therapy through the patient's terminals, thereby realizing personalized healthcare in a closed-loop manner. It is anticipated that the IoMT paradigm could thoroughly change the existing medical model, comprehensively improve the efficiency of medical service, and provide access to a brand-new lifestyle to the masses. Despite the advancements of flexible PENGs in recent years, there are still many challenges and opportunities which lie ahead and that need to be addressed to achieve such an intelligent system.

(1) Flexible PENGs with excellent comprehensive performance need both outstanding mechanical flexibility and electrical performance. Numerous studies have shown that piezoelectric composites can, not only exhibit excellent electrical properties like piezoelectric ceramics but also show compelling mechanical flexibility like those found in piezoelectric polymers, which are ideal candidates for the flexible PENGs. However, the current understanding of the mechanism of piezoelectric composites is lacking and insufficient. Continued efforts need to be committed here to develop flexible PENGs with better performance from a material innovation perspective.

Fig. 20 PENGs for internet of medical things. This smart system includes high-performance sensors that can detect and wirelessly transmit a variety of physiological signals to the doctors over a distance through the 5G network. After data analysis is completed, the doctor can send feedback to the patient or even guide treatment through the patient's in- and on-body terminal and devices, thereby realizing personalized healthcare in a closed-loop manner.

(2) There are multiple interfaces present within PENGs themselves, including the interface between the organic phase and the inorganic phase in the composite, the interface between the material and the electrode, the interface between the electrode and the packaging materials, and the interface between the device and the human body. These interfaces play a vital role in not only the devices' biomedical performance, but also their flexibility, stability, and durability. However, research on these interfaces is usually ignored, which hinders the improvement of the overall performance of the device.

(3) In both sensing and energy harvesting applications, PENGs must closely adhere to the surface of the skin or tissue to extract small biomechanical pressure. Although many studies have proved that PENGs can form conformal contact with the targeted tissue surface, fixing and anchoring of the device itself is rarely mentioned. If the device is directly covered with tape for fixing, for instance, the altered stress and strain distribution can affect its output performance. Especially in the case of a sensor, extra stress or strain placed on PENGs can lead to inaccurate measurement results. To ensure accurate transmission of deformation, advanced packaging and effective fixing of devices are the important research avenues.

(4) With the miniaturization of devices, there is also a desire to integrate more functions on the same device to achieve simultaneous detection of various stimuli, such as temperature, stress, strain, sound, light, atmosphere, and so on. It has been demonstrated that piezoelectric devices can respond to multiple stimuli,[113,117,606] which will further promote its wide applications. However, if the responses of multiple stimuli are all based on electromechanical conversion, it will inevitably cause crosstalk in the output signals, as a result, how to decouple the output to enable accurate detection of individual stimulus is of utmost importance.

(5) Except for the piezoelectric effect, other working principles such as triboelectric, flexoelectric, magnetoelastic, and electromagnetic effect can also be applied to convert mechanical energy into electrical energy with different output characteristics. Especially, the giant magnetoelastic effect was discovered in the soft polymer systems in 2021 from the Chen Group at UCLA and was applied to convert biomechanical activities into electrical signals, which paves a new way to construct biosensors and bioelectronics with a collection of compelling features, including intrinsic waterproofness, decent biocompatibility, high current output, and low inner impedance.[88] Furthermore, many studies have proved that

hybrid generators which rely on a joint mixture of different energy harvesters provide a more effective way to improve energy utilization efficiency.[607–611] Moreover, a sensor with a hybrid mechanism can also offer complementary and improved performance.[612–614] For example, when piezoelectric sensing is coupled with piezoresistive sensing, a wider detection range can be obtained.[615–617]

(6) In addition to developing sensors with excellent performance, signal identification and signal analysis are key steps to implement when using those devices towards practical applications.[618] In recent years, machine learning has shown its ability to extract feature values from a large amount of sensing data,[619–621] remove interference signals to make measurements more accurate, and show important application prospects in data analysis and pattern recognition. Consequently, deep integration of multi-sensor data fusion and machine learning is imperative in their synergistic use for both wearable and implantable bioelectronics.

(7) The application of wearable and implantable PENG in the human body involves multiple disciplines, including physics, chemistry, material science, computer science, mechanical engineering, electrical engineering, and bioengineering. Scientists in various fields should work closely together to ensure that they are tackling specific topic areas using their optimized know-how. In addition, establishing a strong relationship between industry and academia is important to advance the process of transition of these devices from the lab to commercially viable products, and thus to industrialization. Therefore, interdisciplinary cooperation is another important trend in this field.

Piezoelectric nanogenerators have greatly seized people's interest and imagination because of their excellent electromechanical properties, with extensive research progress having already been carried out. However, there is still a long way to go to achieve practical applications of these devices. With more extensive and in-depth research, it is foreseeable that flexible piezoelectric devices can have broad prospects in the fields of wearable and implantable bioelectronics and will exert a significant impact on future personalized healthcare.

Author contributions

J. C. initialized and supervised the whole project. W. D. and Y. Z. contributed equally to this work. The manuscript was written through the contributions of all authors. All authors have approved the final version of the manuscript.

Conflicts of interest

The authors declare no competing financial interest.

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