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Review

1D nanomaterial based piezoelectric nanogenerators for self-powered biocompatible energy harvesters

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ABSTRACT

Wearable applications require power sources that are flexible, affordable, compact, and easily accessible. However, conventional power electronic devices are impractical due to their heaviness and rigidity. Additionally, batteries and external power sources have limited lifespans and applications, posing challenges for implanted and wearable devices that require consistent energy supplies. To overcome these challenges, 1D energy-related technologies have been developed, with nanogenerators emerging as an ideal power source. They can generate biomechanical energy from physical activities like muscle contraction and heartbeat and convert it into electrical signals for various applications, including biological indicator detection, cardiac pacing, nerve stimulation, and tissue repair. This review provides an overview of piezoelectric nanogenerators (PENGs) and recent progress in their development, with a focus on biomaterial-based PENGs for healthcare monitoring. Furthermore, the review discusses the future prospects and challenges of optimizing PENGs.

1. Introduction

Technological gadgets such as sport wristbands, apple watches, and google glass have become an integral part of our daily lives, allowing us to connect with others and monitor our health. [1–3]. However, the development of these devices is hindered by the limitations of conventional power supplies, such as bulky and rigid lithium-ion batteries. To address this challenge, researchers have turned their attention to self-powered systems, particularly nanogenerators, which offer durability and biocompatibility in wearable applications. Before nanogenerators can be used in the human body, factors like biocompatibility and non-toxicity must be carefully considered to ensure user safety [4–7]. Additionally, the structure of the nanogenerator must be designed to fit within the limited and asymmetrical spaces both inside and outside the body, while maintaining high sensitivity and efficiency.[8,9].

There are various methods to convert biomechanical energy into electrical energy, including piezoelectric, triboelectric, magnetoelastic, and electromagnetic effects [10,11]. Among them, PENGs based on piezoelectric effects have gained significant attention since their invention in 2006 [12]. Unlike their electrostatic and electromagnetic counterparts, PENGs can convert mechanical energy into electrical energy and vice versa, enabling simple architectures for energy harvesters [13]. PENGs have proven useful in monitoring weak physiological signals and find applications in healthcare, such as wound healing detection, respiration monitoring, alcohol consumption detection, and more [2,3,14].

Recent developments in PENGs have focused on fabricating high-performance piezoelectric materials using different synthesis approaches and characterizing their properties. One-dimensional (1D) bio-piezoelectric energy harvesters (BPEH) are particularly attractive due to their high piezoelectric coefficient, which is an intrinsic property of the nanomaterial arising from its internal structure and composition. This property is independent of any external sources and device configuration, resulting in enhanced energy conversion efficiency [15]. These devices typically consist of a piezoelectric material coated on a flexible substrate, which is then attached to a biological system, enabling energy harvesting from natural movements. These harvested energies can power healthcare monitoring devices, eliminating the need for battery replacement. The energy harvested by the 1D BPEH is typically in the form of AC (alternating current) output due to their piezoelectric nature. To make this energy usable for powering healthcare monitoring devices and other electronics that require stable DC (direct current) power, a rectifier is used. The rectifier converts the AC output into DC, enabling efficient energy storage in batteries or capacitors and eliminating the need for frequent battery replacements [16]. This approach ensures a continuous and sustainable power source for various applications.

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However, challenges remain, such as low output voltage and current, as well as the stability and reliability of these devices in the human body [16–18]. Despite the challenges, 1D BPEH holds great promise for healthcare monitoring applications. Ongoing research aims to improve their energy conversion efficiency and reliability, with the potential to revolutionize the healthcare industry through continuous, non-invasive monitoring of physiological parameters in real-time [16,18,19]. This article provides an overview of recent advancements in PENGs for biomedical applications, wearable electronics, and human motion monitoring. The focus is on the material structures and designs of 1D PENGs, their applications in self-powered medical electronics, and practical considerations for in vivo and in vitro investigations. Fig. 1.

2. Piezoelectric nanogenerator

Piezoelectricity was discovered in 1880 by French scientists Pierre and Jacques Curie on Rochelle salt, quartz, and cane sugar. The “direct piezoelectric effect” is the ability of piezoelectric materials to generate electric charges when they are put under mechanical stress (Fig. 2.). In the same way, the “indirect piezoelectric effect” [20–23] describes the mechanical stress observed when an electric field is applied. When an electric field is applied, the polarization of ferroelectric materials changes. This is a subclass of the piezoelectric family. Therefore, to enhance the ferroelectric properties, apply a high electric field, which leads to the alignment of dipoles. In the poling process, the randomly aligned dipoles tend to align in the same direction with respect to the applied direction [24–26]. When an outside force is applied to a crystal structure, it breaks the center of symmetry, which creates a piezoelectric potential. For successfully converting mechanical energy to electrical energy in ZnO nanowires, Wang [12] proposed the first PENG. Next, polyvinylidene fluoride (PVDF) and its copolymers [27–30] were found to have piezoelectric, pyroelectric, and ferroelectric properties. As diverse mechanical energy exists in diverse situations, such as human motion, rolling vehicles, and sound waves, using PVDF as a piezoelectric material has received global attention in recent years. To date, the most popular technology for self-powered systems has been piezoelectric material-based technology, compared to triboelectric, pyroelectric, electromagnetic, and electrostatic systems [15,31]. Despite its ability to typically generate a relatively high output, the triboelectric nanogenerator (TENG) exhibits instability in its output property due to its susceptibility to wear and moisture. In contrast to TENG, PENG possesses several advantages such as compact size, extended durability, and reduced susceptibility to environmental factors. These benefits arise from the electrical output being generated through the change of internal polarization state caused by structural cell deformation. However, PENGs further progress has been hindered by its relatively lower

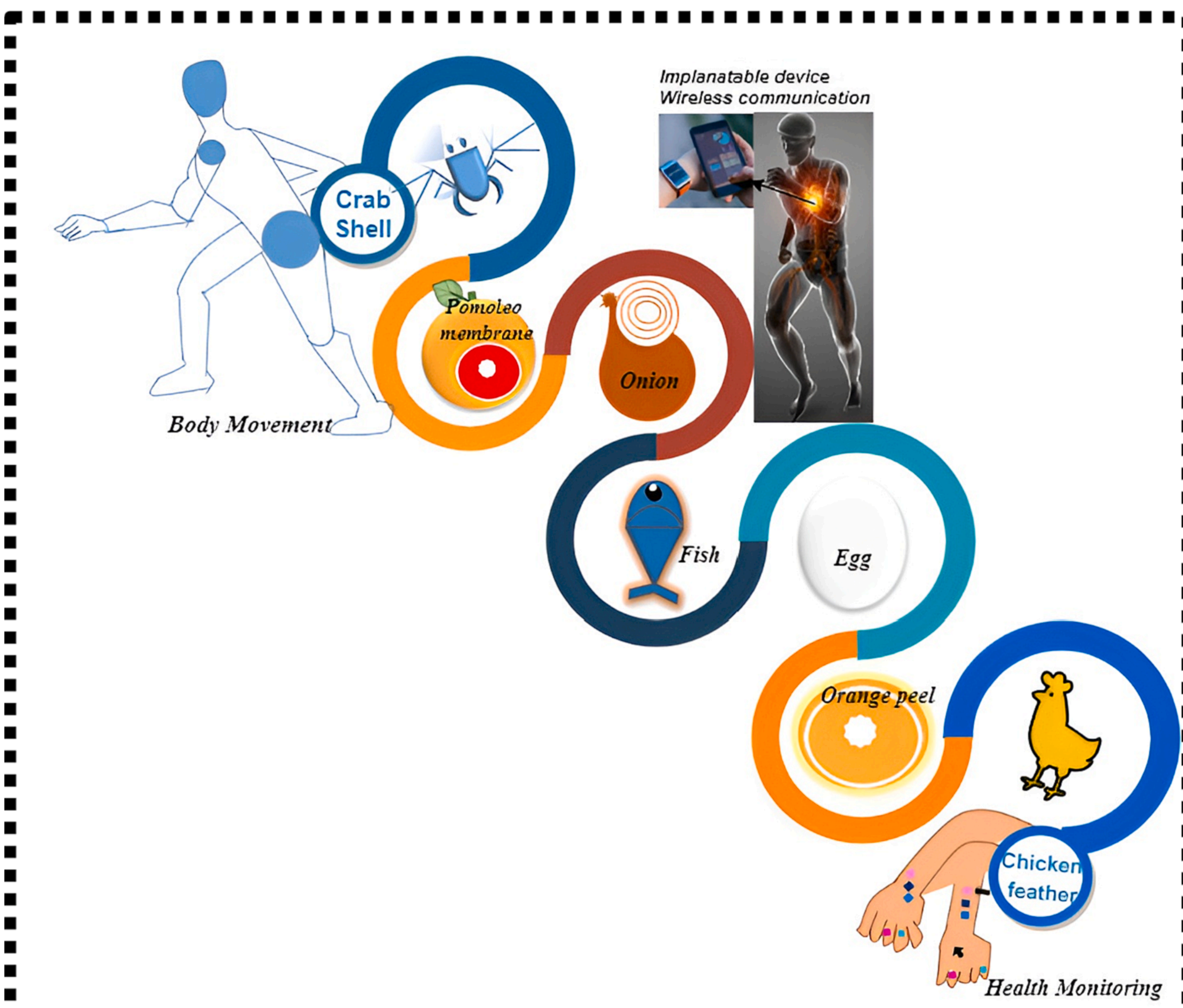

Fig. 1. A schematic diagram of nanogenerators based on natural materials and their applications in vitro and in vivo for biomedical applications.

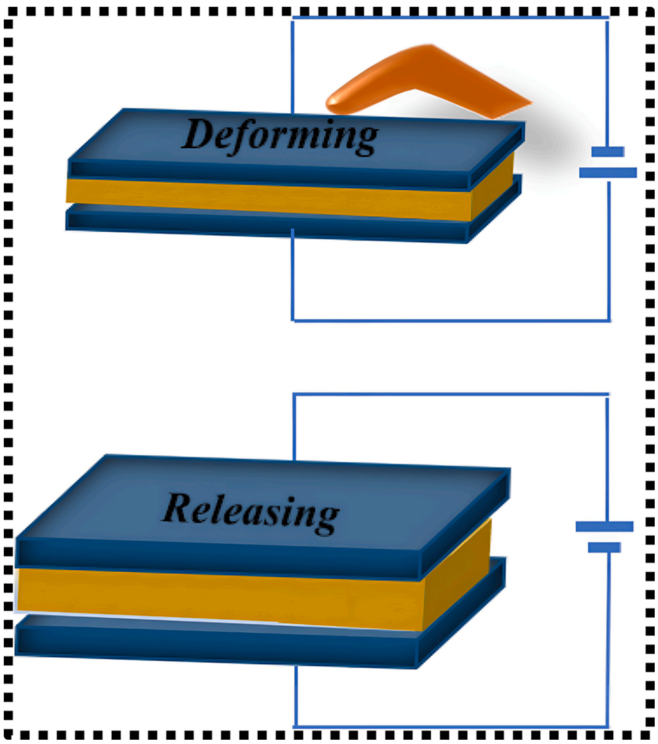

Fig. 2. A Schematic diagram of piezoelectric nanogenerator.

electrical output. As a result, if the output performance of PENG can be enhanced through thoughtful design, it retains significant potential for applications in micro-nano mechanical energy harvesting and self-powered sensing systems [32,33].

2.1. Material selection

Selecting a piezoelectric material for energy harvesting depends on the material's piezoelectric properties and its application. It can be classified into the following: ceramics such as barium titanate ($BaTiO_3$) [34], lead zirconate titanate (PZT), and potassium niobate ($KNbO_3$) [35]; single crystals such as rochelle salt, lithium niobate, and quartz crystals; polymers such as PVDF, PVDF-terpolymer, and PVDF-copolymer, polylactic acid (PLLA) [36–39]; nature biomaterials such as fish scales, onion, crab shell, pomelo, and chicken feather and their composites [40–45]. The most significant aspect is that harvesting less mechanical energy from the human body would lead to reduced efficiency. On the other hand, if the device took in more mechanical energy, it would need to be more efficient. This would mean that the user would have less energy that could be recovered and would have to use more energy to do the same amount of work. Additionally, there is a possibility of physical discomfort while wearing the device. Therefore, the challenge that remains is to develop an efficient device that can operate on its own power without restricting body movements or negatively impacting the user's comfort.

2.2. Material structure and design

With the rapid growth of wearable technology, wearable bioelectronics are under increasing pressure to continuously improve their mechanical and electrical performance. These devices need to be both flexible and stretchable while maintaining high electrical output performance, which requires careful control of the material structure and design. To achieve these goals, researchers are exploring two main approaches: material engineering and structural design, as illustrated in Fig. 3. One-dimensional nanomaterials, such as organic–inorganic lead halide perovskite materials, carbon-based additives, metal materials, lead-free piezoelectric ceramics, and hydrated ionic salts, have been reported as promising options for improving the performance of wearable bioelectronics.

2.3. Synthesis of 1D PENGs

2.3.1. One-dimensional (1D) nanostructures

Due to their superior physical and chemical characteristics, 1D nanostructures such as nanofibers (NFs), nanowires (NWs), nanorods (NRs), nanobelts (NBs), and nanotubes (NTs) are the best candidates for a variety of applications, including biomedicine, drug delivery, nanoelectronics, gas sensors, nanodevices, and perovskite solar cells [46–52]. NFs, a subset of these 1D nanostructured materials, have drawn a lot of interest because of their unique properties when compared to other nanostructured materials used in energy storage devices. NFs can be synthesized using a variety of methods, such as the hydrothermal method, electrospinning (ES), phase separation technique, and template-assisted method.

2.3.1.1. Hydrothermal method. Continuous-flow hydrothermal and solvothermal synthesis techniques have gained significant attention in the last 20 years as highly promising one-step procedures for the synthesis of inorganic nanomaterials. They offer the potential to produce a diverse range of nanoparticles at large scales required to meet rising industrial demand in a cost-effective and efficient manner. These techniques involve heating precursor solutions in a sealed vessel at temperatures beyond the boiling point of the solvent, along with pH, reagents, and additives, enabling the design of sophisticated reaction systems. As these reactions occur at relatively low temperatures, they can create metastable phases that might not be produced by other methods, and highly nanocrystalline products can be synthesized without additional heating steps. However, long reaction times and uneven heating can lead to

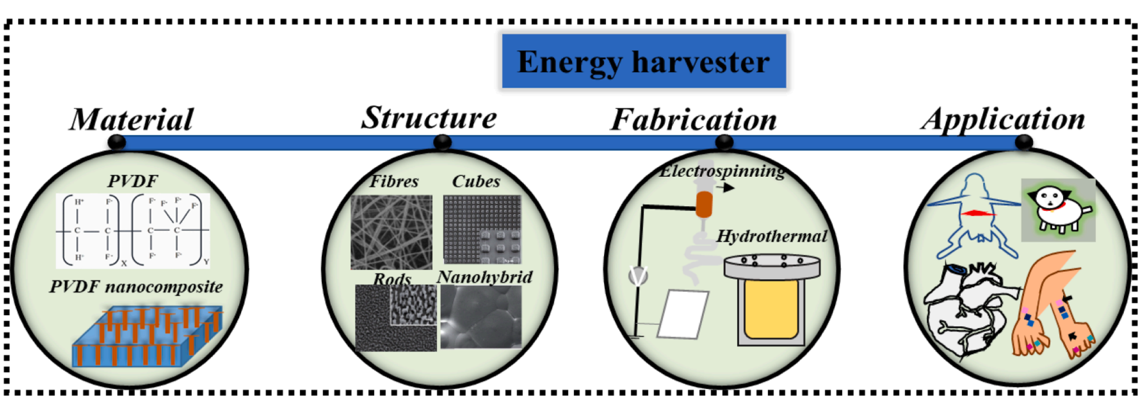

Fig. 3. A Schematic illustration of material structure and design.

complications such as phase separation and polydispersity [53,54], and the physical conditions inside the reaction vessels are not yet fully understood.

Despite these challenges, more research efforts have led to the development of nanofiber-based PENG using the hydrothermal method. For example, Yao et al. demonstrated that a PENG device made of barium titanate (BT) nanocrystals embedded in a PDMS matrix can generate an output voltage and current of 13 V and 200nA, respectively, at a high concentration of 30 wt% of BT under periodic bend-release motion. At its ideal resistance of 35 MΩ, the maximum output power of the PENG device can reach 2.6 W. This PENG device can also convert biomechanical movements into electrical energy and light up four green LEDs without any external energy source [55]. Additionally, Zhao et al. improved the performance of PENG by fabricating BTO/HBP/PMMA nanowires into PVDF, resulting in an output voltage and current of 3.4 V and 0.32 A, respectively [56].

2.3.1.2. Electrospinning technique. An important method for processing nanofibers is electrospinning, which was first proposed and used in the 1930s and began to receive significant attention in the late 20th century as a result of growing industrial and academic interest. To meet commercial requirements, the fabrication parameters still need to be improved and enhanced. For example, getting electrospun nanofibers without clogs is still difficult. The fabrication of nanofibers with improved strength, more controllable morphology, and novel functionalities is the result of extensive research [57]. The low crystallinity, random alignment, and orientation of nanofibers contribute to their generally low mechanical strength. Electrospinning has been used to fabricate a variety of nanomaterials directly or indirectly, including organic polymer nanofibers, carbon nanofibers, metal nanofibers, ceramic nanofibers, and inorganic hybrid nanofibers. Their use in soft electronics, including transparent electrodes, conductors, transistors, optoelectronics, sensors, and energy devices, has been growing. Incorporating nanofiber into a polymer matrix is another successful method for increasing the mechanical flexibility of PENGs. For instance, through the use of finger knocking, Ichangi et al. embedded a flexible piezoelectric nanofiber made of PVDF/$BiFe_2O_3$ to generate an output voltage and power density [58]. On this basis, under the influence of finger imparting, a self-poled PVDF based PENG on Bi_2O_3 nanostructure has been developed by Biswas et al. The output voltage and current are 3.6 V and 2.4 μA respectively [59]. Han et al. fabricated a P(VDF-TrFE)/PEDOT:PSS core shell nanofiber-based piezoelectric energy that could be folded, doubling the performance of voltage and current compared to its unfolded state. When a PENG composed of 10-fold and 5-fold could reach output higher than ~ 8.76 V and ~ 547 nA respectively [60]. Chen et al. fabricated a $BiCl_3$-PVDF nanofiber-assisted PENG through an electrospinning technique [61]. At 2 wt% of $BiCl_3$-PVDF nanofiber, the output voltage was found to be 1.1 V, which was four times higher than that of pure PVDF. Furthermore, a key factor in improving piezoelectric response is sample thickness. When struck by a falling ball, the output of PENG was found to be 38 V. It suggests that piezoelectric materials show great potential for electronic devices [62]. Uddin et al. discussed the fabrication of yarn-PVDF PENGs through casting (SC-PENG) and touch spun nanofibers (NFC-YPENG), which are used in low-power electronic applications [63]. Resonant frequencies, a range of forces, and related energy outputs were applied to the Y-PENGs. In comparison with SC-PENG, the output voltage of NFC-PENF was found to be 1.24 V. It is proved that it is possible to light an LED by electricity storage in a 4.7 F capacitor and power wearable electronics by converting electrical output from AC to DC. So, the coated yarn could be used in sensor and actuator applications as well as single-fiber microdevices [64,65].

2.3.1.3. Phase separation technique. It is considered that this synthesis approach is the most effective for the fabrication of 1D nanofibers with diameters ranging from 50 to 500 nm. It makes use of a variety of solvents and is backed up by a freezing and drying process, which is carried out using a method of thermally induced liquid–liquid phase separation. This technique for making fibers requires the following steps: gelation of the polymer at a low temperature, phase separation, exchanging the solvent by drowning it in water, polymer dissolution in the solvent, and finally freeze-drying the solution. A wide variety of polymers, including PLGA, PLLA, and PDLLA, are utilized in the production process of porous NFs. Phase separation allows scaffold morphology to be evaluated by adjusting polymer type and concentration, freezing temperature, and porous framework. This method can also be useful in creating scaffolds of varying shapes in accordance with the requirements while preserving uniformity from phase to phase. This technique has a number of drawbacks, including the fact that it is difficult to maintain a consistent liquid level and presents challenges in producing clean sand, wax, and paraffin [66–69]. Abolhasani et al. discussed the performance of PVDF-HFP/DMF/THF nanofiber PENG and exhibit higher output voltage and current of PVDF-HFP/DMF than the PVDF-HFP/THF nanofiber. The use of porous nanofibers, though, is not restricted to nanogenerators. This insight can be used to improve battery design using polymer membranes [70]. Huang et al. developed a PVDF/graphene@Fabric composites to monitor the speaking and body motions and could generate the sensitivity of 34 V/N [71].

2.3.1.4. Template-assisted method. The template-assisted method is used for the synthesis of nanomaterials and mainly includes two classes, namely, hard-template and soft-template methods. For the hard template-assisted method, porous templates, such as mesoporous silica, zinc oxide, and anodic aluminum oxide, have been utilized to fabricate 1D polymer nanomaterials, such as Polypyrrole (PPy), Polyacrylonitrile (PAN), poly(p-phenylenevinylene) (PPV) derivatives, and etc. [72–74]. In contrast, soft template-assisted methods involve different block copolymers, surfactants, aggregates, liquid crystals, biomolecules, and polymer nanofibers as templates that can be utilized as sacrifice scaffolds to fabricate 1D polymer nanomaterials. Ponan et al. reported that the magnesiochromite in PVDF was prepared using a template coprecipitation approach and designed a PENG output voltage and power density of 1.05 V and 0.51 $\mu W/cm^2$ at 1 MΩ and 50 kΩ of load resistance, respectively, which are significant for wearable electronics and energy harvesting applications[75]. Calahorra et al. obtained a shear piezoelectric response along the axial direction in the self-assembled cellulose nanofibers through a template-assisted method [76]. Table 1. summarizes recent advancements in PENG made of various materials. Indeed, a large number of studies have adopted a similar method of discretizing traditional bulk piezoelectric materials. Electrospinning is a versatile and flexible method for preparing 1D nanomaterials with diameters ranging from tens of nanometers to several micrometers. It can be used to create nanomaterials from a variety of materials, including polymers, inorganic materials, and composites.

2.4. Characterization

In section 2.3, the synthesis method for 1D nanostructures has been discussed. Next, the ferroelectric, piezoelectric, and mechanical properties can be studied through characterization techniques.

2.4.1. Ferroelectric properties

Two phenomena, including (1) nonlinear and spontaneous electric polarization that can be reversed by the applied electric field and (2) ferroelectric to paraelectric phase change when the material is heated above the Curie temperature, Tc, indicate that some nonconductive materials are ferroelectric. To comprehend the nature of ferroelectric materials and the polarization (P) versus electric field (E) loop measurement, it is essential to study their unique ferroelectric properties such as polarization reversal, coercive field, remnant, and saturation

Table 1
Summarizes recent advancements in 1D-piezoelectric nanogenerators made of various materials.

Materials	Method	Voltage (V)	Current (μA)	Power density ($\mu W/cm^2$)	Application	Ref.
PVDF-TrFE nanofiber	Electrospinning	21	1.2	5700	Wearable electronic devices	[126]
PVA nanofiber	Electrospinning	2.6	–	–	Wearable electronic devices	[127]
PVDF-TrFE nanofiber	Electrospinning	150	0.7	8750	Human motion	[128]
PVDF nanofiber	Electrospinning	0.8	0.390	–	Human motion	[129]
BT-PVDF-GO nanofiber	Electrospinning	2.5	0.10	–	Joint motion monitoring	[130]
Carbon fibers/ZnO nanowires	Two-step method	0.02	0.02	–	Human motion	[131]
PVDF-BZT-0.5BCT	Electrospinning	6.37	–	–	Respiration monitoring	[132]
PEDOT:PSS-PVP nanofiber membrane	Electrospinning	0.08	0.16	–	Electronic textile	[133]
Calcium phosphate nanorod	In-situ	47	1.8	–	Electronic devices	[134]
PDMS-ZnO nanoflakes	Chemical vapor deposition	122	16	6220	Finger tapping	[135]
PMN-PT	Hydrothermal	4 V	0.4	1.75	Wearable electronic devices	[136]
BiCl3-PVDF	Electrospinning	1.0	2	0.2	Electronic devices	[137]
Ce-PVDF-graphene	Electrospinning	11	600	6.8	Energy harvesting	[138]
PVDF-SM-KNN	Hydrothermal	21	33	115.5	Electronic devices	[139]
Vanadium-ZnO nanosheets	Hydrothermal	32	6.2	–	Energy harvesting	[140]
PEDOT:PSS	Oxidative polymerization	1.54	0.63	0.59	Human motion	[141]

polarization. These parameters play a crucial role in understanding the unique behavior and characteristics of ferroelectric materials, which are essential for their various applications in electronic devices [77]. Batra et al. discussed the performance of dielectric, ferroelectric, and piezoelectric properties and used them for the application of PENG [78]. The ferroelectric properties are enhanced by the addition of Nd into ZnO through the wet chemical co-precipitation method, and the values of P_r and E_c are observed to be 0.064 $\mu C/cm^2$ and 6.04 kV/cm, respectively, at 600 V (Fig. 4 (A)). If the P_r value is not zero, it means that the Nd-ZnO nanorods have ferroelectric domains. This shows that it can be used to make nonvolatile ferroelectric storage devices [79,80]. Motion detection is accomplished using the wearable device for the $Bi_{0.5}Na_{0.5}TiO_3$-polycaprolactone film [81]. Raj et al. compared the ferroelectric properties of the films, with the remnant polarization (P_r) measured at 1 Hz observed to be 34 $\mu C/cm^2$ which is higher than that of the film measured at 0.05 Hz (46 $\mu C/cm^2$) (Fig. 4 (B)). This may account for the time needed for dipole rotation at high frequencies [82]. According to a recent report, PENG is used for computer–human interfaces with metal-free single-crystal films. Specifically, P_s and E_c values of 13.3 $\mu C/cm^2$ and 30 kV/cm, respectively, were found to be attainable in MDABCO-NH_4 films, demonstrating their ferroelectric properties.

2.4.2. *Piezoelectric properties*

It is essential to measure the piezoelectric coefficients in order to assess the performance of piezoelectric materials. These coefficients provide a quantitative measure of the change in volume that occurs in a piezoelectric material when it is subjected to an electric field. As a result, this section reviews the recently developed methods for piezo force microscopy (PFM) and atomic force microscopy (AFM) for measuring the piezoelectric properties of 1D nanostructures [83]. Zhao et al. were the first to use the PFM to measure the piezoelectric coefficient of a ZnO nanobelt [36]. Since then, the PFM has been utilized on a regular basis to investigate the piezoelectric response of PZT nanofiber [84,85], and CdS nanowire [86]. For the fabrication of PENG, PVDF-$CsPbBr_3$ fiber is employed as the piezoelectric material [87]. Fig. 5 (A) shows the square-shaped hysteresis loop of the phase and the butterfly-shaped amplitude loop, which indicate the existence of a well-defined polarization. The electrostatic field between PVDF and $CsPbBr_3$ causes a high self-bias effect and non-symmetric hysteresis curves. This shows that the composite fiber has an effect of self-polarization [88]. Kaur et al. designed PVDF-PU PENGs for use in body motion applications such as the knee, elbow, and foot by merging them with wearables [89]. PFM, which represents the reverse piezo response achieved in the tapping mode, was used to study the piezoelectric behavior in terms of d_{33}. This was done with the intention of better understanding piezoelectric behavior. Fig. 5 (B) shows that the piezoelectric coefficient d_{33} value of pure PVDF (3.02 pm/V) is greater than that of PVDF-PU nanofiber (7.64 pm/V). The dipoles tend to align themselves in the direction of the electrical field that is being applied, and it is expected that these dipoles will change direction when the electric field is reversed. The point at which the direction of voltage begins to change is known as the coercive voltage, and its value is higher than that of pure PVDF. In order to capture mechanical energy, Bharti et al. created a transparent and stretchable PENG employing graphene and zinc silicate (Zn_2SiO_4) nanorods [90]. The tetragonal crystal structure of the Zn_2SiO_4 nanorods was achieved by a hydrothermal technique. The piezoelectric charge coefficient (d_{33}) of Zn_2SiO_4 nanorods was measured using piezoelectric force microscopy with the nanorods dispersed in acetone and deposited on a conductive substrate. After annealing, PFM amplitude and phase images were obtained, confirming the piezoelectric property. PFM phase-voltage hysteresis loops and displacement-voltage curve were also obtained, with a calculated d_{33} value of 117 p.m./V (Fig. 5 (c)). A slight positive shift in displacement values was observed, potentially due to the presence of an internal built-in electric field and surface potential effects.

2.4.3. *Mechanical properties*

For 1D nanostructures to maintain their electrical characteristics while being subjected to mechanical stress, including bending, folding, and stretching, long-term operation and durability under environmental effects must surpass the lifetime and performance of the device, particularly in the case of PENGs operated in high-frequency rotating mode. There are several factors that can affect the performance of PENGs, including: (1) high temperature, which can cause the piezoelectric material to expand and contract, thereby affecting the output voltage and current; (2) high humidity, which can cause the piezoelectric material to absorb water, thereby affecting its piezoelectric properties and reducing its electrical output; (3) changes in mechanical stress on the device, such as bending or twisting, which can also affect its output; (4) environmental vibrations, such as those caused by wind or traffic, which can affect the output of the device; (5) high levels of radiation, which can affect the electrical properties of the piezoelectric material and reduce its output; and (6) exposure to chemicals, such as acids or solvents, which can affect the electrical properties of the piezoelectric material and reduce its output. Therefore, it is crucial to consider these environmental factors when designing and using PENG to ensure optimal performance and reliability [85,86]. The mechanical properties of PENG make them useful for a variety of applications that require energy harvesting, sensing, and self-powered operation [87,88]. Rasel et al. developed a wearable hybrid device that combines one PENG and two TENGs into a single structure (Fig. 6 (A)). Both piezoelectric and triboelectric effects were simultaneously induced by the mechanical force of hand clapping [91]. Through structural optimization, the

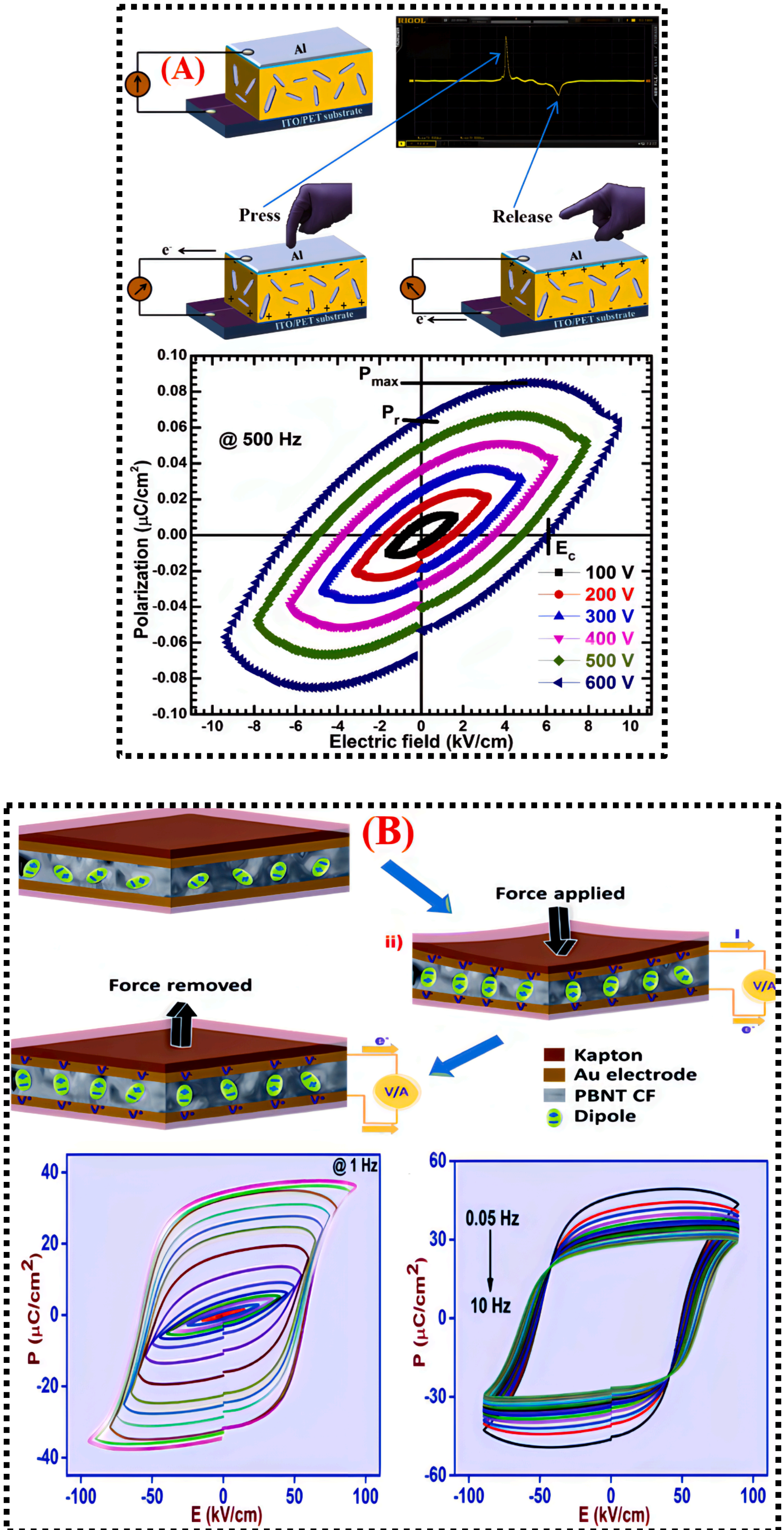

Fig. 4. Ferroelectric properties (A) P-E loops of Nd-ZnO NRs at various applied electric fields (@ 500 Hz). Reproduced with permission from ref. [78] Copyright 2018, Elsevier. Fig. 4 (B) Ferroelectric hysteresis behavior of the BNT system measured under varied electric fields at different frequency (1 Hz, 0.05 Hz, and 10 Hz). Reproduced with permission from ref. Copyright 2018, RSC [81].

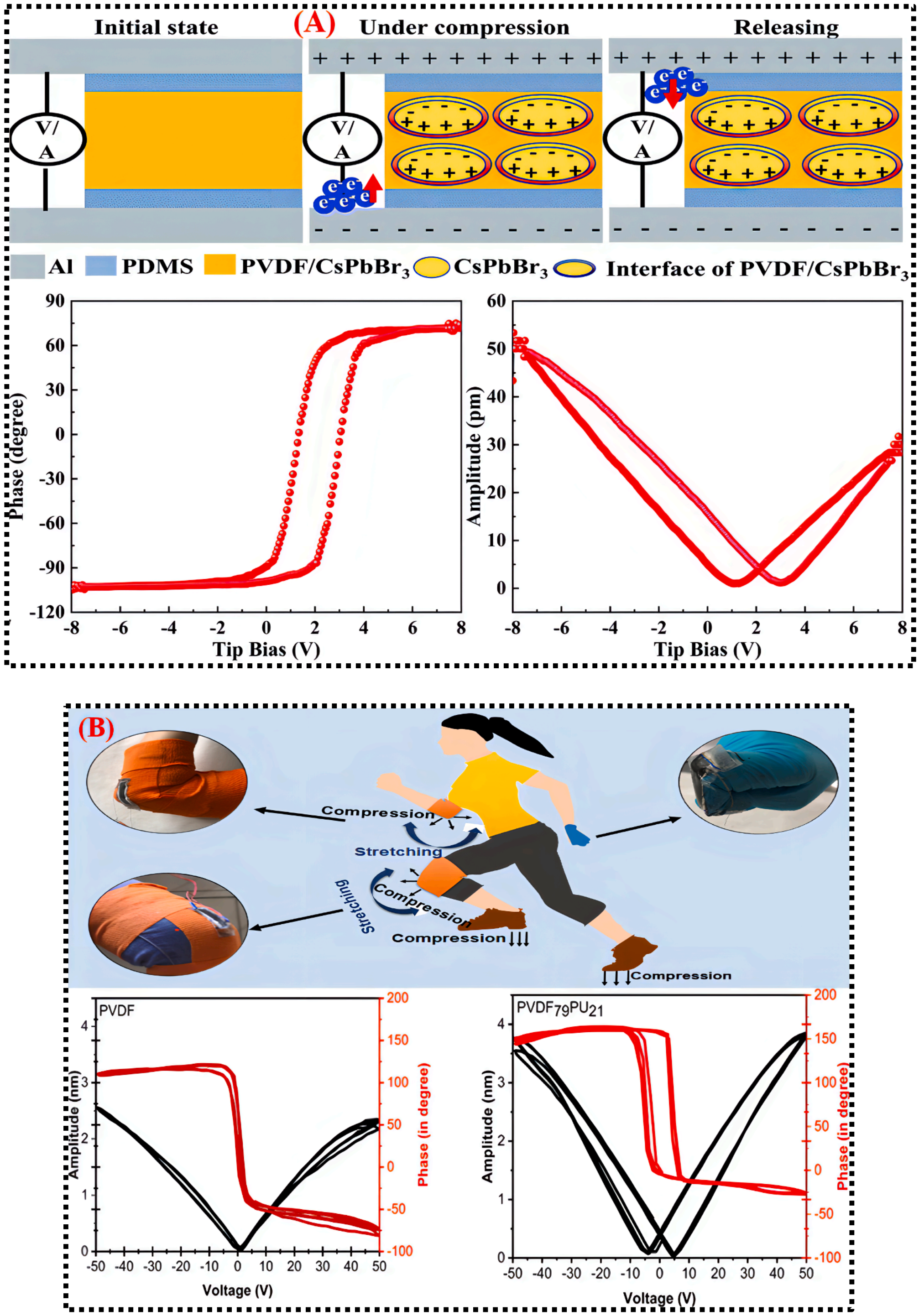

Fig. 5. Piezoelectric properties (A) phase and amplitude hysteresis loop of the PVDF-4-CsPbBr3 film. Reproduced with permission from ref [95] Copyright 2022, RSC (B) PVDF-Polyurethane nanofiber. Reproduced with permission from ref [96] Copyright 2022, ACS and (C) amplitude & phase images, phase versus voltage and d_{33} constant of Zn_2SiO_4 nanorods. Reproduced with permission from ref [98] Copyright 2020, Elsevier.

combined devices had the highest output voltage and current, reaching 1140 V and 890 A, respectively, with a peak power density of 3.7 W/m^2. The combined nanogenerator has been used in an electric trimmer, a mobile phone, a tracker, and a portable Wi-Fi router. Deng et al. designed a shaft-like hybrid nanogenerator that consists of two components: one is PVDF as a piezoelectric material, and the other is a PDMS-nitrile baffle as a triboelectric material, as shown in Fig. 6 (B). The device can generate electrical energy using low-frequency tension. The output voltage of the PVDF can reach 157 % of its original value when pulsed stimulation is applied, whereas the open-circuit voltage of the PDMS-nitrile baffle can reach 138 V. The hybrid structure of PENGs and TENGs is used in the application of low-frequency energy harvesting and nonlinear movement [92–94].

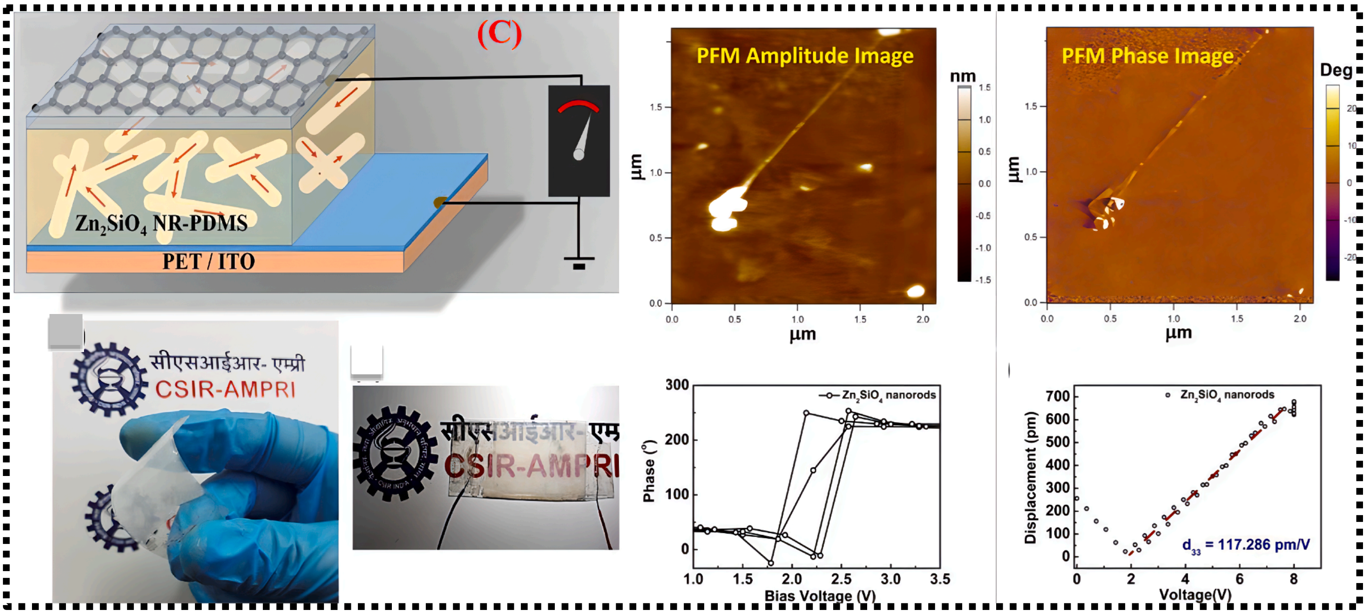

Fig. 5. (*continued*).

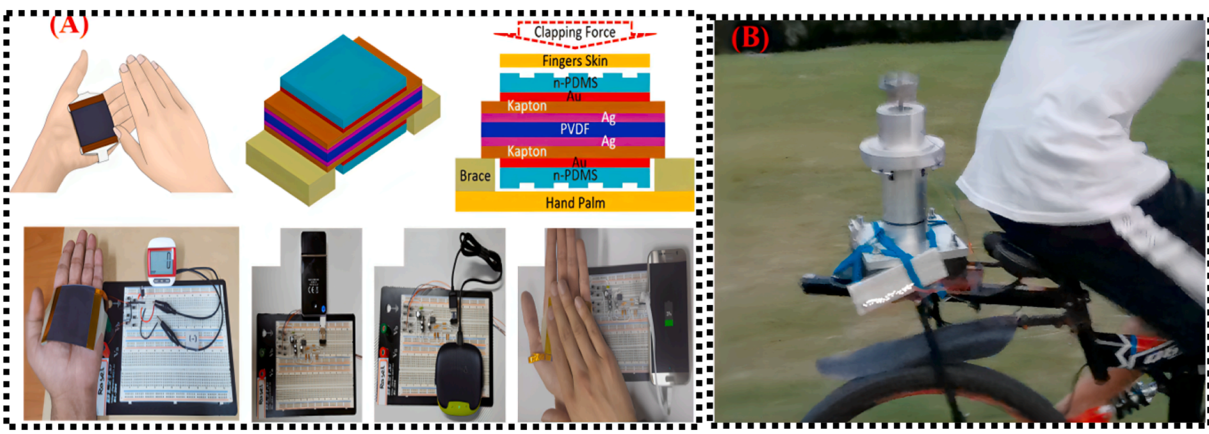

Fig. 6. (A) the digital still-frame images of hybrid triboelectric nanogenerator. Reproduced with permission from ref [99]. Copyright 2019, Elsevier and (B) installation of Bistable triboelectric linear generator. Reproduced with permission from ref [100] Copyright 2019, Elsevier.

3. Recent development in PENG

Researchers are investigating the potential applications of flexible and wearable PENGs. These devices, which can be embedded in clothing, shoes, or other wearable items, can generate energy from the wearer's movement or body heat. Recently, efforts have been made to increase these devices' adaptability, robustness, and effectiveness. Conventional PENGs often have a low power output, which makes them hard to use in many situations. However, most recent advancements have concentrated on increasing these devices power output. Researchers have come up with new materials, designs, and ways to make PENGs so that they have more power, are more efficient, and can be used in a wider range of situations. PENGs can be used as self-powered sensors that do not require external power sources. Self-powered PENG sensors are currently being developed for a variety of applications, such as environmental sensing, healthcare monitoring, and structural health monitoring. Researchers are exploring the use of PENGs as multi-functional devices that can harvest energy, sense mechanical motion, and perform other functions. For instance, PENGs can sense the motion of the waves and collect energy from them for environmental monitoring purposes. Additive manufacturing and nanofabrication enable complex and precise PENGs. These techniques allow for the fabrication of PENGs with improved performance and functionality. These breakthroughs in PENG technology will allow for novel energy harvesting, sensing, and self-powered devices. This section discusses the most recent studies on PENGs that harvest passive biomechanical energy from human activity using various piezoelectric materials.

3.1. Piezoelectric composites

Piezoelectric composites, composed of inorganic and organic materials, offer a balanced combination of mechanical flexibility and excellent electrical properties. Researchers have explored various concepts, including different filler morphologies, to enhance their performance. Recent work by Durga and Hemalatha focused on the development of PVDF-based nanocomposites for energy harvesting, specifically through the use of mechanical force. By incorporating zinc ferrite and nickel ferrite fibers into the PVDF matrix, they were able to increase the open circuit voltage and output power of the resulting nanogenerators. A schematic illustration of the PVDF-$ZnFe_2O_4$ composite film nanogenerator was shown in Fig. 7 (A) [90]. The compression and subsequent release exerted on the top of the nanogenerators by a mechanical

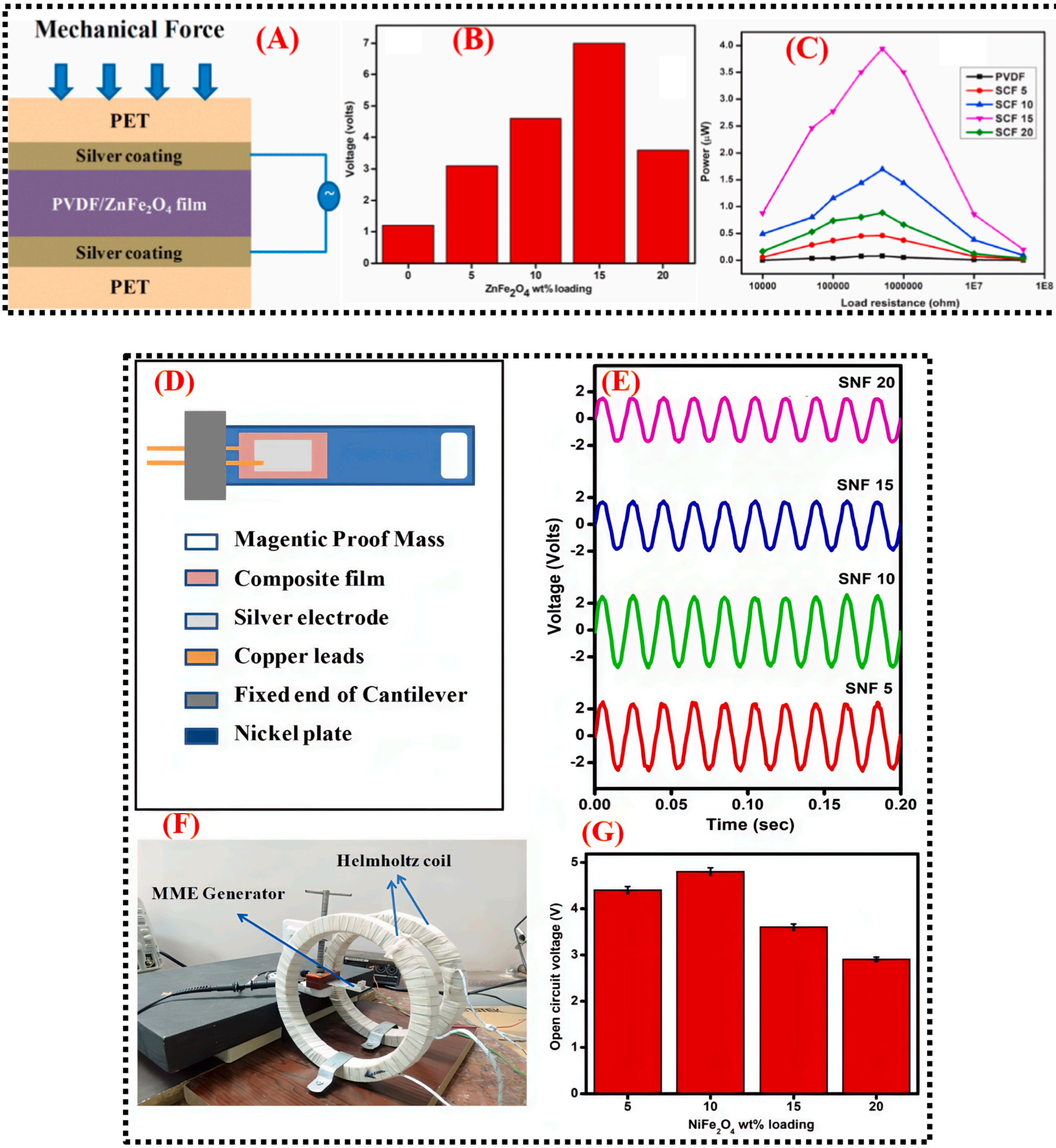

Fig. 7. (A) Representation of PVDF nanocomposite nanogenerators, (B) output voltage driven by nanogenerators, (C) voltage response histogram, Reproduced with permission from Ref [101] Copyright 2021, Elsevier. (D & F) illustration and image of MME generator, (E & G) open circuit voltage and histogram of PVDF-$NiFe_2O_4$ composite films with Reproduced with permission from Ref [102] Copyright 2021, Elsevier.

oscillator imparts a force of 1.5 N. The open-circuit voltage response of the composite film nanogenerators was shown in Fig. 7 (B), which displayed repeated changes in the output voltage as a result of force application. The output power of the SCF sample exhibited a maximum value of 4 μW, as shown in Fig. 7 (C). PVDF-$NiFe_2O_4$ exhibits a maximum open circuit voltage of 10 V, which is six times higher than that of plain PVDF. Fig. 7 (D) illustrate the schematic diagram of the PVDF-NiFe2O4 composite of the magneto-mechano-electric (MME) generator and the energy harvesting performance, respectively [91]. The open-circuit voltage obtained by applying a magnetic field of 10 Oe and the histogram of the peak-to-peak voltage response as a function of $NiFe_2O_4$ loading are shown in Fig. 7 (E), respectively. Overall, the development of these nanocomposites and composite film nanogenerators has the potential for energy harvesting applications, and the incorporation of nickel ferrite fibers into PVDF shows promise for higher open circuit voltage.

3.2. Lead-Free based PENGs

In PENGs, inorganic piezoelectric materials have gained attention for their excellent electrical output profiles, including ZnO, AlN, $BaTiO_3$, PZT, PMN-PT, MoS_2, WSe, and SnS. However, their bulkiness and brittleness hinder their application in wearable bioelectronics. Research is ongoing to optimize the bending and stretching capabilities of PENGs for increased flexibility and efficient electromechanical coupling. For example, Lead-free 0.5Ba($Zr_{0.2}Ti_{0.8}$)O_3 and 0.5(Ba0.7$Ca_{0.3}$)TiO_3 (BZT-

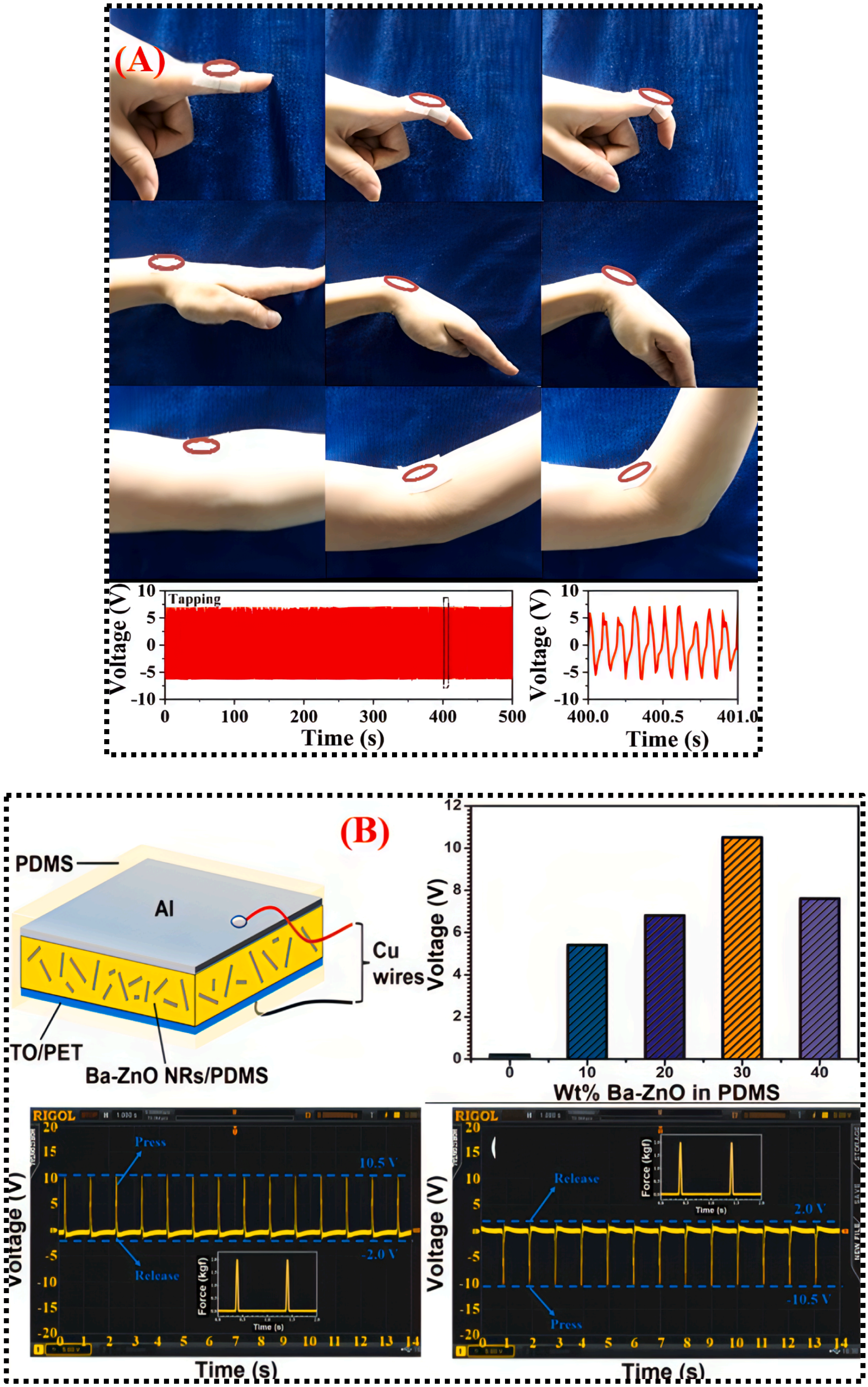

Fig. 8. (A) Piezoelectric nanogenerator of BZT-BCT/P(VDF-TrFE) nanofibers. Reproduced with permission from Ref [103] Copyright 2020, Elsevier. (B) Ba doped ZnO flexible nanogenerator. Reproduced with permission from Ref [104] Copyright 2021, Elsevier. (C) hybrid nanogenerator for mechanical energy harvesting. Reproduced with permission from Ref [116] Copyright 2019, Elsevier.

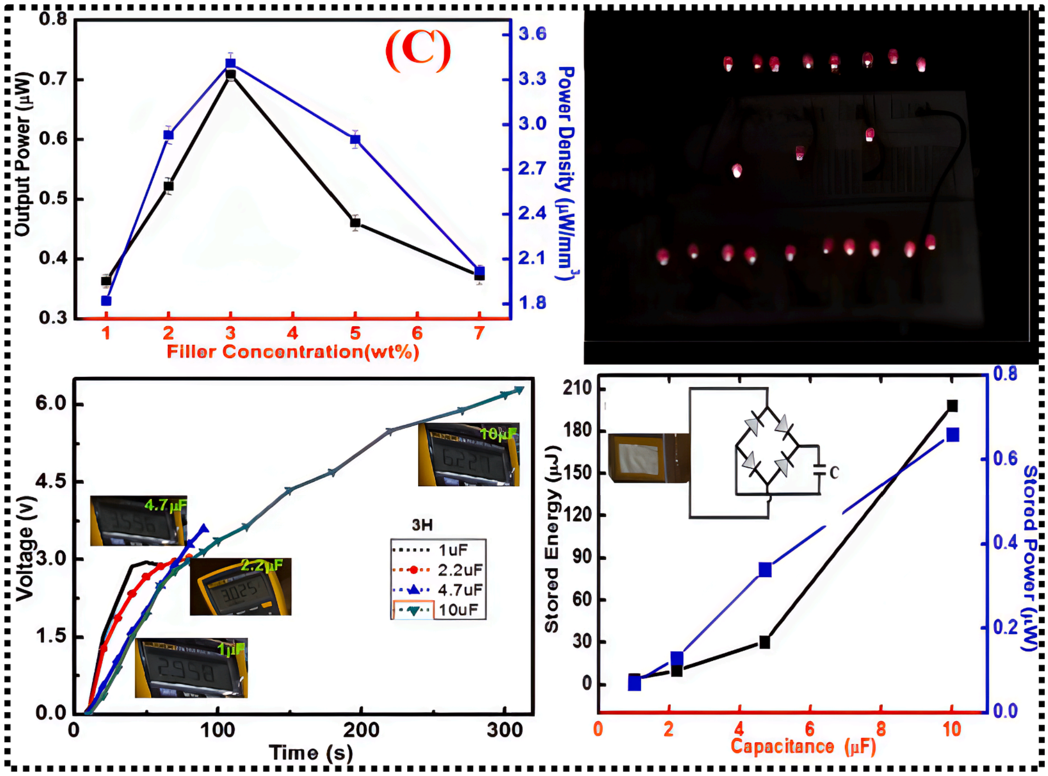

Fig. 8. (*continued*).

BCT) with P(VDF-TrFE) nanofiber were prepared by electrospinning, and the resulting film was placed on finger joints, wrist joints, and elbow joints [95]. The output performance of the nanogenerator was approximately 1.3 V during ten bending cycles, with the finger position producing the highest value. A reliability test showed that the generator maintained a constant output voltage after 5000 cycles (Fig. 8 (A)). Ba-doped ZnO nanorods were also used to fabricate a PENG, which produced an open-circuit voltage of up to 10.5 V containing 30 wt% Ba-ZnO (Fig. 8 (B)) [96]. The reverse bias connection provided an output signal opposite to and of the same magnitude as the forward signal, indicating that the generated signals are due to piezoelectric phenomena rather than noise or triboelectric contribution. These devices have potential for use in mechanical energy harvesting and to drive portable electronics [96–98]. These studies demonstrate the potential of lead-free flexible PENGs for energy harvesting applications. In addition to being environmentally friendly, they also have the advantage of being biocompatible, making them suitable for use in biomedical devices. The use of bio-flexible materials such as polylactic acid and polydopamine [99–102] in combination with piezoelectric ceramics such as BCZT and $BaTiO_3$ nanotubes [103] has shown promising results in generating high open-circuit voltages and short-circuit currents. The development of lead-free PVDF with potassium sodium niobate rollable nanocomposites has also shown excellent performance in terms of generating high electric output [104]. These PENGs can consistently provide high output even under repeated agitation, making them suitable for various mechanical energy harvesting applications. Overall, the development of lead-free flexible PENGs is an important area of research that has the potential to lead to the creation of environmentally friendly and biocompatible energy harvesting devices that can be used in a variety of applications [105].

3.3. The integration of PENG and energy storage

Current energy harvesters face challenges in providing continuous stable energy output from the fluctuating biomechanical energy of the human body. To address this, storing generated electrical energy in independent devices (e.g., batteries or supercapacitors) and releasing it at a steady rate has been a common solution. For instance, The development of various PENGs using different nanocomposite materials, such as P(VDF-HFP)-$NiFe_2O_4$ fibers, cerium-doped $NiFe_2O_4$-P(VDF-HFP) composite nanofibers [106,107], ferrite nanorods/PVDF/polyaniline nanochains [108], and Zinc Ferrite-Rod-P(VDF-HFP) nanocomposite films [109]. These materials aim to improve the piezoelectric output voltage, address the disadvantages of low short-circuit current and high internal resistance in nanocomposites, and enhance the flexibility and non-toxicity of the devices [110–112]. The generated power from these nanogenerators can be used to power self-powered devices and sensors, and can even charge capacitors up to 10 μF (Fig. 8 (C)) in a short amount of time. Overall, these studies demonstrate the potential of piezoelectric composites for developing high-performance and environmentally friendly energy harvesters.

4. Application of biomaterial-based PENGs for health care monitoring

Biomaterial-based PENGs have the potential to revolutionize health care monitoring by providing a self-sustaining power source for health monitoring devices. They can harvest energy from the body's movements and convert it into electrical energy to power health monitoring devices such as implantable sensors, wearable devices, health monitoring patches, prosthetic devices, and drug delivery systems. This technology has the potential to improve patient care by enabling continuous and non-invasive monitoring of vital signs and physiological parameters, thereby facilitating early detection of diseases and timely intervention. Additionally, biomaterial-based PENGs are eco-friendly and sustainable, as they can generate energy from natural sources and do not rely on batteries or external power sources. Overall, biomaterial-based PENGs have the potential to transform the healthcare industry by providing efficient, reliable, and self-sustaining power sources for health monitoring devices.

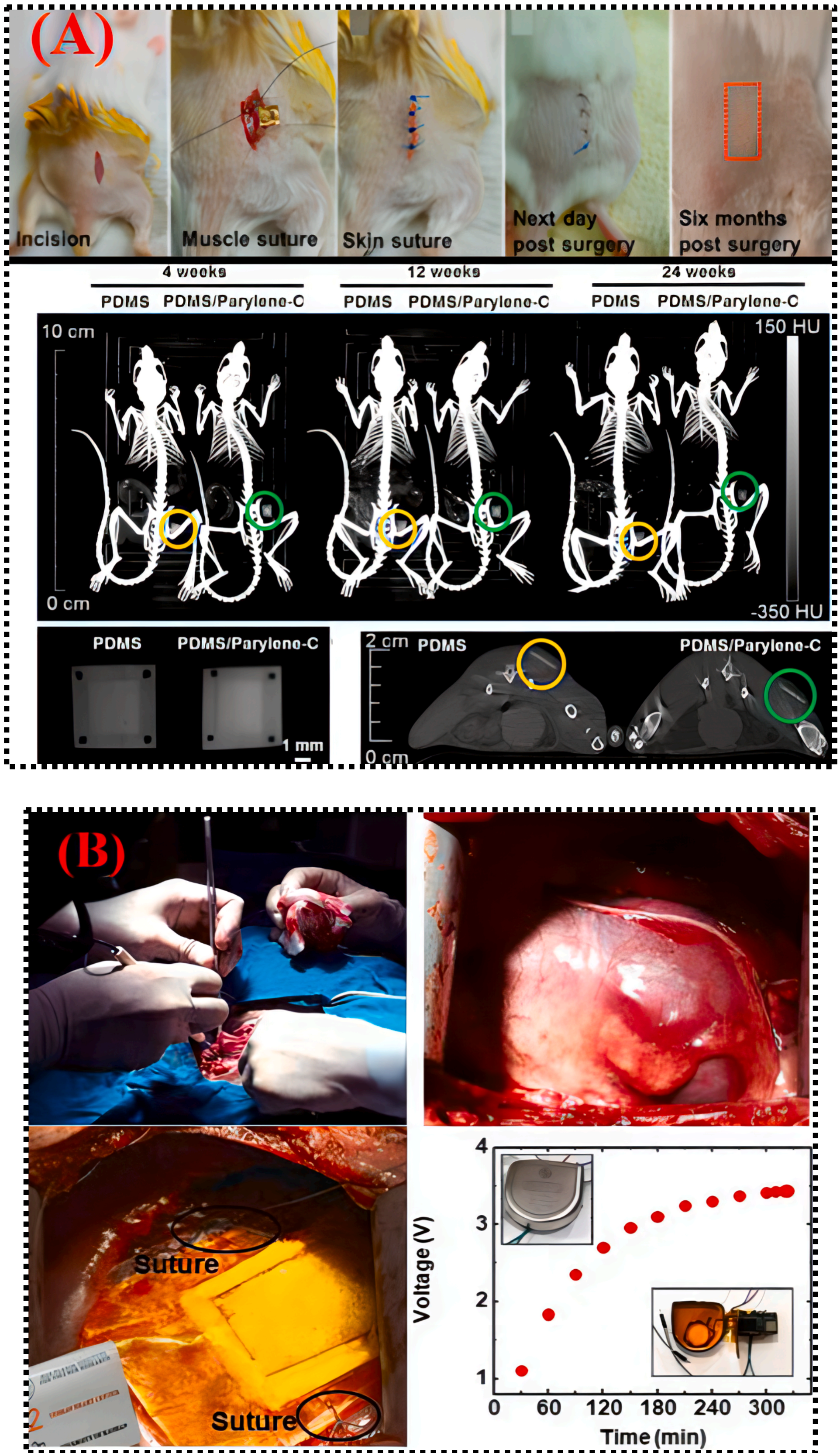

Fig. 9. (A) Photograph of the nanogenerators implantation surgery process. Reproduced with permission from Ref [120] Copyright 2018, Elsevier, (B) output voltage response with respect to time. Reproduced with permission from ref [122] from Elsevier. (C) battery free pacemaker. Reproduced with permission from Ref [123] from ACS.

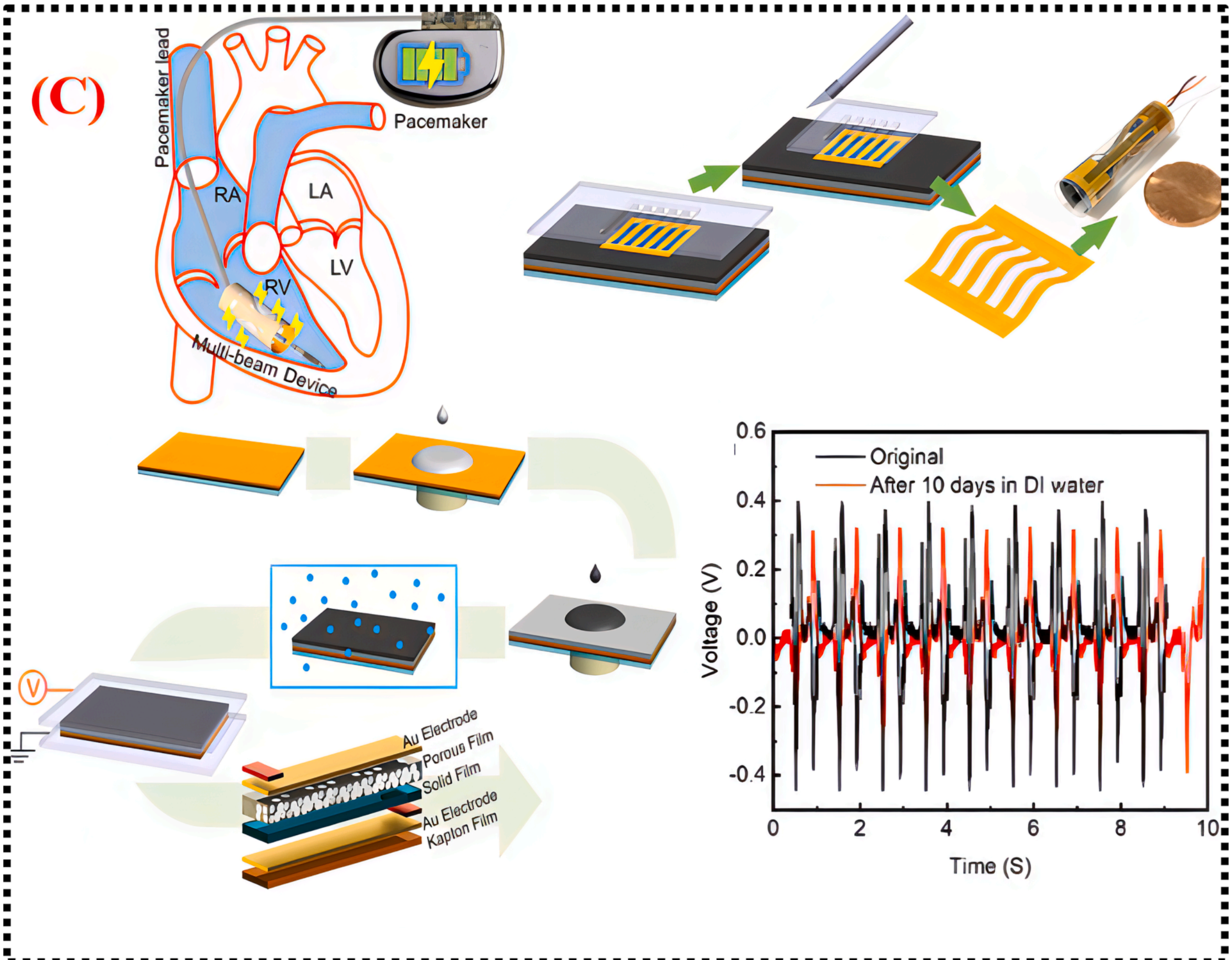

Fig. 9. (*continued*).

4.1. Biomechanical energy harvesting

Biomechanical signals from the human body can offer crucial healthcare information. Biomechanical sensing has been accomplished through diverse PENGs, utilizing materials design, micro/nano structural design, and various manufacturing techniques. For example, Lie et al. [113] implanted PDMS and PDMS/parylene-C-packaged PVDF nanogenerators in mice and studied their performance for up to six months using imaging techniques for in vivo small-animal imaging and shown in Fig. 9 (A). The implantable nanogenerators remained stable in the subcutaneous tissue throughout the implantation period and showed steady output, confirming biomechanical energy harvesting. In the same way, the sponge-like mesoporous PVDF film nanogenerator that was implanted in rodents had an open circuit voltage (V_{oc}) of 200 mV (Fig. 9 (A)). Using piezoelectric composite films, such as BZT-BCT nanowires and PVDF, an ultrasound-active thin-film nanogenerator was made [114]. The electrical pulses produced by the implanted piezoelectric thin-film nanogenerator can be programmed using distant ultrasound excitation, enabling variable input power and waveform. Azimi et al. studied a polymer-based nanogenerator made of PVDF, ZnO, and rGO to create a battery-free pacemaker [115]. The implanted nanogenerator produced 0.487 μJ of energy (Fig. 9 (B), enough to power the pacemaker via in vivo energy harvesting, allowing the battery to be removed. Xu et al. [116] developed a cardiac energy harvester using P(VDF-TrFE) composite films embedded in PDMS, which was implanted in a human body. The device showed excellent reliability, maintaining a constant output voltage even after immersion in distilled water. The output voltage was consistent in air, phosphate-buffered saline, and simulated blood, and the device retained a constant output voltage over an extended period. The device charging performance was also tested using a capacitor connected to a full-bridge rectifier, indicating potential use in implantable biomedical applications.

4.2. Human motion sensing

The fabrication of the next generation of PENGs is a great challenge in biomedical applications for the growth of non-toxic, super-sensitive, and flexible bioinspired PENGs [117]. PENGs play a vital role in comprehensive human motion sensing, providing detailed information about motion quality and types for diverse applications in healthcare, human–machine interaction, and sports. These flexible devices can be conveniently attached to body parts such as necks, fingers, wrists, elbows, or knees, facilitating the monitoring of joint motions, bending angles, deformation amplitudes, and frequencies. Recently, Maiti et al. developed a biowaste-based PENG using naturally occurring, untreated onion skin [40]. The output voltage, current, power density, and efficiency of piezoelectric energy conversion of the fabricated onion skin bio-PENG were 18 V, 166nA, 1.7 μW/cm^2, and 61.7 %, respectively, and 30 green LEDs were turned on by a single device under a repeated compressive stress of 34 kPa and a frequency of 3.0 Hz, respectively. In addition, Fig. 10 (A) shows that a maximum output voltage (106 V) was achieved when six units were connected in series, which turned on 73 combined LEDs (30 green, 25 blue, and 18 red). Therefore, PENG can be used in biomedical energy devices with a long-life span. Kumar et al. [118] investigated using human finger and other vibrations (twisting, bending, walking, and tapping the foot) to generate energy. Adding P-FS

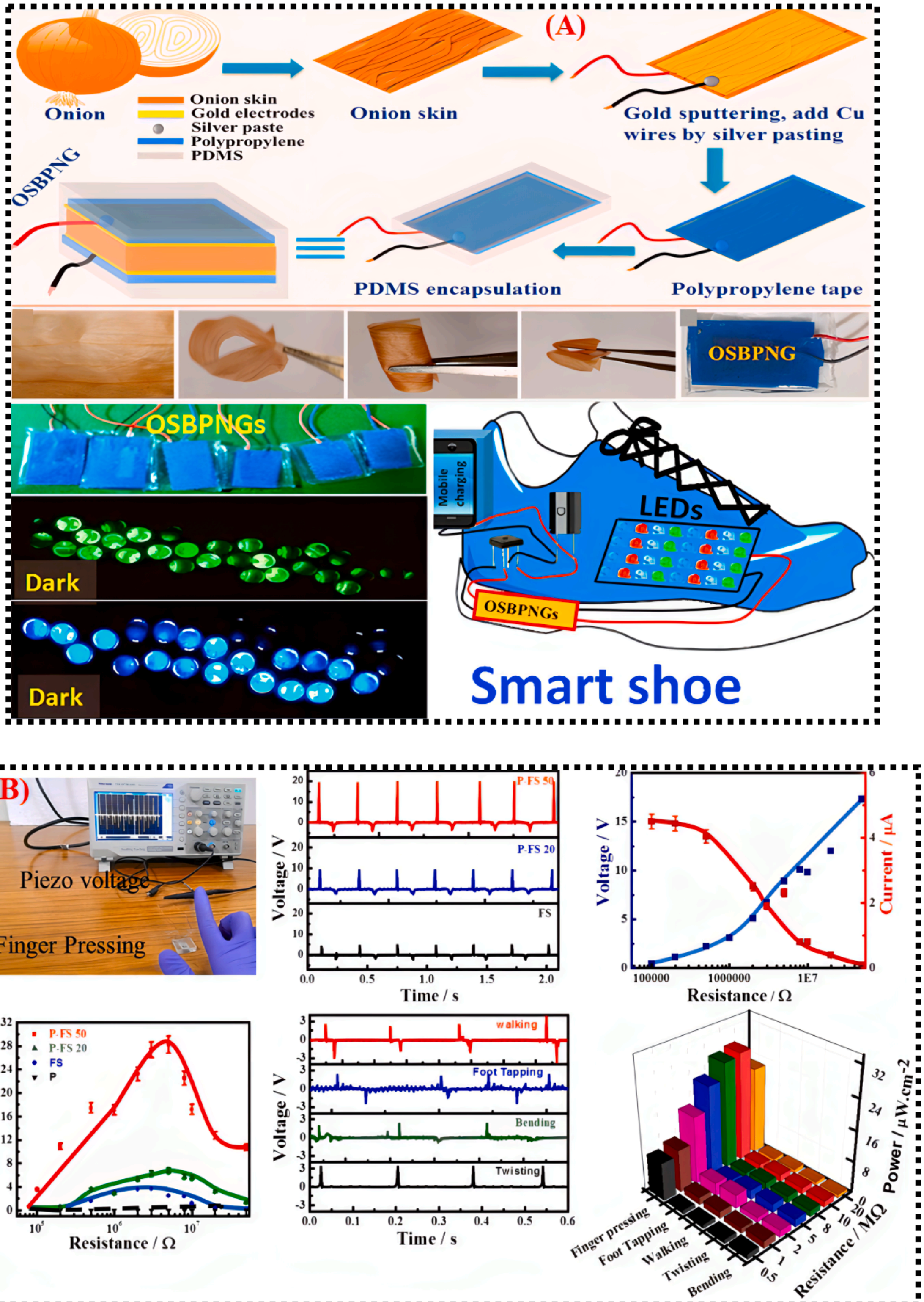

Fig. 10. (A) Combined LEDs (green, blue and red) under dark and light conditions-Reproduced with permission from Ref [34] Copyright 2017, Elsevier, Biowaste hybrid piezoelectric energy harvester. Reproduced with permission from Ref [125] Copyright 2019, Elsevier.

50 (50 wt% fish scale) resulted in a 20 V output voltage, higher than other weights and pure fish scale. Output voltage, current, and power densities were measured for different resistances to test nanogenerator feasibility. Voltage increased while current decreased with resistance. P-FS 50 achieved a maximum power density of 28.5 μW/cm^2 at 5 M, higher than pure fish scale (Fig. 10 (B)). Twisting and bending produced 2 V and 3.5 V, respectively, higher than other human activities. A three-dimensional view showed walking and foot tapping voltage and power were as high as twisting and bending, indicating the device's potential use in miniaturized devices, including bio-implants. Hoque et al. developed two types of PENGs using biowaste chitin nanofibers: chitin nanofiber-based PENGs (CPENGs) and a PVDF/chitin composite-based

PENG (PCPENG) [119]. The chitin nanofibers were extracted from crab shells and incorporated into PVDF to induce the electroactive β-phase. The entire system was encapsulated with PDMS. When a human finger pressed and released the device with a pressure of 27.5 N (Fig. 11 (A)). The positive value is due to the time of applied strain, and the negative value appeared when the device returned to its original position. The device can be used as a mechanical energy harvester in mobile devices [120,121]. Similar to this, Gaur et al. used PVDF to develop an energy harvester from biowaste orange peels [43]. The piezoelectric effect in orange peel, caused by cellulose and proteins, was studied to evaluate its energy harvesting performance. The devices with V_{oc} values of 3, 58, 70, and 90 V showed superior performance to pure PVDF (Fig. 11 (B)). The power density achieved was 135 $\mu W/cm^2$ for various applied resistances. Human movements such as bending, twisting, and walking generated 5, 4, and 9 V respectively, and the finger-tapping method charged the capacitor up to 1.2 V in 40 s, with discharge in the same time frame. A household sliding door was used to confirm the efficiency of the hybrid device in LED lighting. This biowaste-based hybrid has potential use in energy harvesting and as a green energy source for electronic devices. Bairagi et al. [45] discussed a biocompatible and self-poled piezoelectric energy harvester made from pomelo fruit membrane. The nanogenerator sensitivity was tested on various parts of the human body, including the wrist, elbow, finger, throat, jaws, and leg. Applying a small force to the device caused mechanical deformation, which induced dipole displacement and reorientation in the membrane, generating a potential difference between the two electrodes. The output voltages of front and back wrist movement were 205 and 481 mV, respectively (Fig. 11 (C)). The back wrist showed a higher maximum value than the front wrist, which may be due to the high stress induced in the generator. Bending and stretching of the fingers generated a peak-to-peak voltage of 536 mV by the PENG fruit membrane. The nanogenerator's overall performance in human motion can be used in various biomedical applications, including gadgets and implantable devices [122–125].

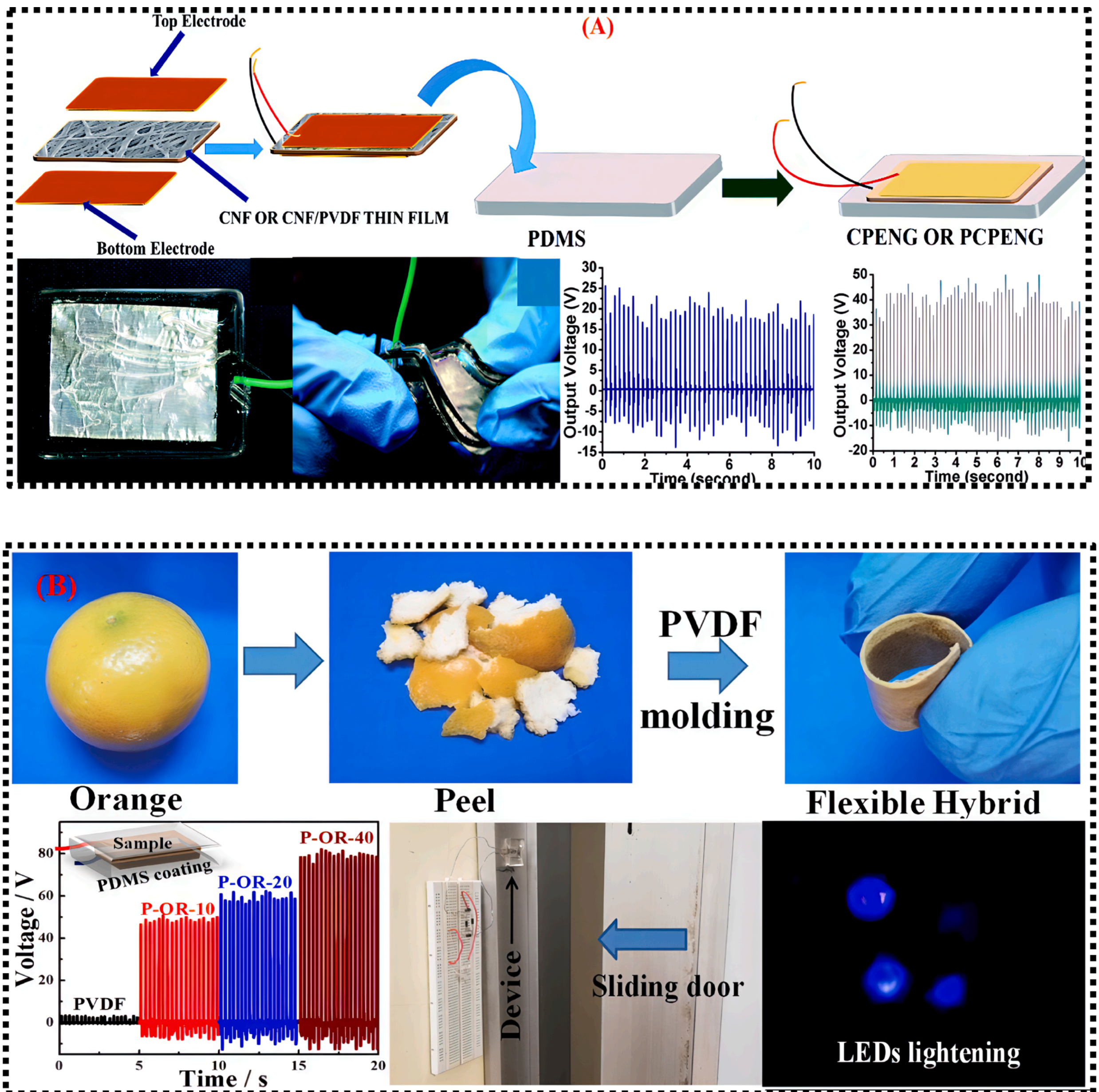

Fig. 11. Piezoelectric energy harvester (A) output voltage of crab shell-PVDF. Reproduced with permission from Ref [126] Copyright 2018, from RSC (B) output voltage and LED of orange-PVDF. Reproduced with permission from Ref [37] Copyright 2020, from Elsevier (C) output signal voltage of pomelo fruit Reprinted with permission from Ref [39] Copyright 2020, from Nature reports.

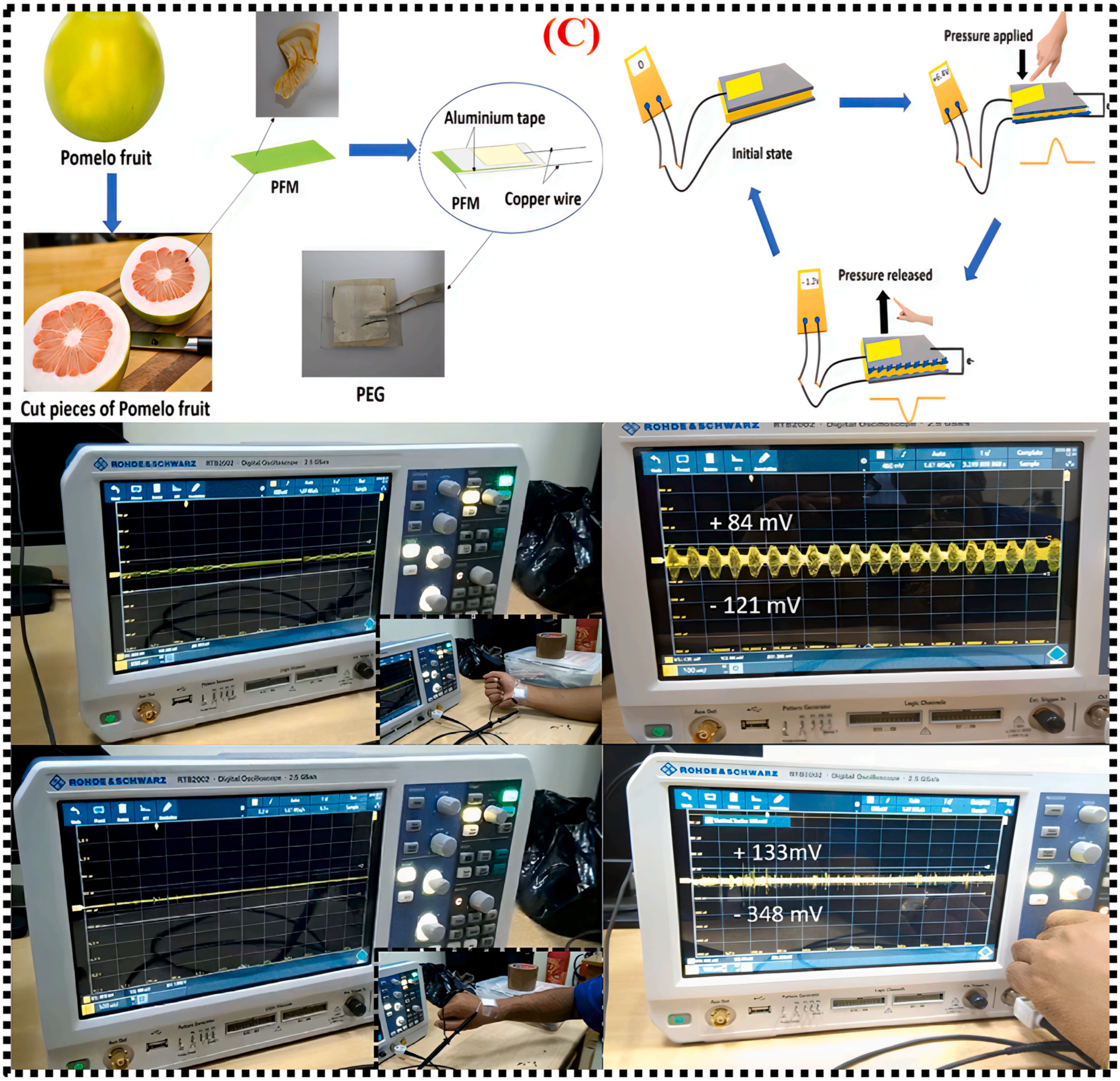

Fig. 11. (*continued*).

4.3. Health care monitoring

Healthcare monitoring involves continuous tracking of physiological parameters and health data using advanced sensors and wearable devices to improve patient care and enable early detection of health issues. For instance, Gaur [36] designed a PENG that harnesses waste mechanical energy from human body movements using a green composite of pomegranate peel powder with PVDF. The P-PM40 sample exhibited the highest voltage output compared to pure PVDF, with its maximum power density obtained due to the added filler that induces the electroactive β- phase of PVDF. The nanogenerator showed a time delay of 5 ms between bending and releasing modes, which was not present in pure PVDF. The influence of human body movements on the device's output voltage was demonstrated, with walking and bending resulting in output voltages of 3.8 V and 4 V, respectively. Fig. 12 (A) demonstrates the capacitor charging and discharging, as well as the LED lighting achieved through finger imparting. Therefore, human movement generates sufficient power to illuminate LEDs and store energy in capacitors. Kar et al have utilized chicken feather fiber (CFF) to design a biocompatible, flexible bio-piezoelectric energy harvester (BPEH) that demonstrates excellent piezoelectric output voltage, power density, and current density when exposed to periodic biomechanical pressure [41]. The piezoelectric behavior of the BPEH is attributed to the keratin-enriched CFF, while its mechanical strength is provided by disulfide bonds. Real-life applications, including charging capacitors, illuminating LEDs, and monitoring human pulse rate, have confirmed the potential of the BPEH to power wearable electronics, bio-implantable devices, and healthcare monitoring tools (Fig. 12 (B)). Energy harvesting using biowaste materials presents a promising avenue for sustainable and eco-friendly power generation. By utilizing waste materials such as fruit peels and membranes, which are often discarded, these materials can be repurposed for energy production instead of being treated as waste. Additionally, the use of biowaste materials is an attractive alternative to traditional materials because of their abundance, low cost, and biodegradability. Moreover, the development of biowaste-based energy harvesting technologies has the potential to reduce the dependence on non-renewable resources, ultimately contributing to a more sustainable future. Overall, these advancements in biowaste-based energy harvesting

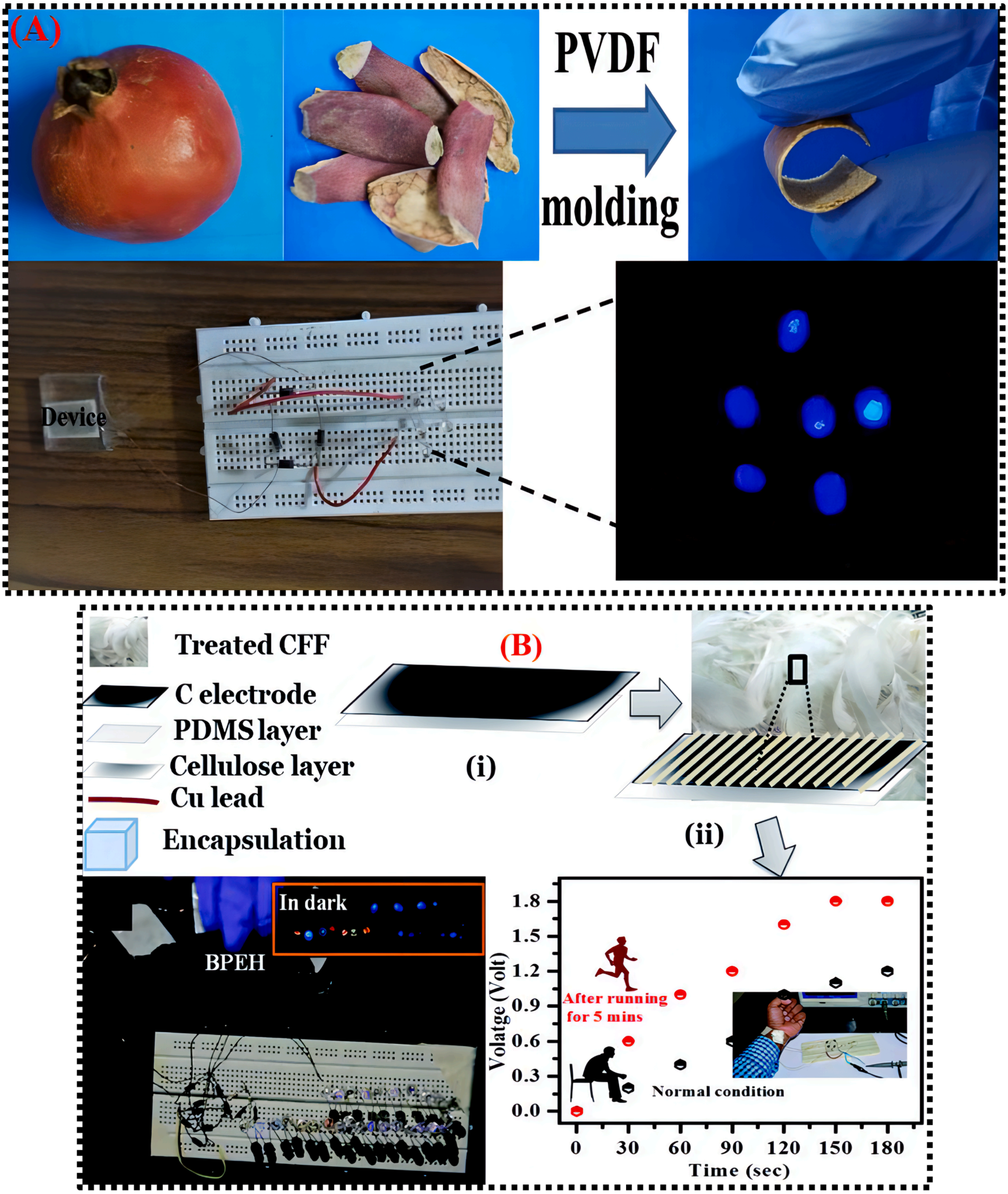

Fig. 12. Piezoelectric energy harvester (A) LED lightening of pomegranate-PVDF. Reproduced with permission from Ref [36] Copyright 2018, from RSC (B) LED lightening of chicken feather. Reproduced with permission from Ref [35] Copyright 2020, from Elsevier.

technologies hold significant promise for sustainable and environmentally conscious power generation.

5. Conclusion and future aspects

Living humans and animals have a variety of power sources, including chemical, thermal, and mechanical energies. Utilizing these energies could aid in the development of self-powered health care electronics and long-term wearable or implantable medical devices. To harvest the various types of energies, many ways have been developed. In this review, we summarized and discussed the overview of PENGs, followed by material selection, structure and design, characterization, and recent developments in PENGs. The development of implantable and wearable electronic devices on human and animal bodies has been studied intensively. According to the literature, polymer-based PENGs are rapidly growing and have made significant contributions to the medical field. However, specific issues still need to be addressed in the future.

5.1. Device packaging and biocompatibility

The most essential aspects of self-powered PENGs are their packaging and biocompatibility. As a result of the physical activity of the human body and flexible packaging, the device should be able to withstand all types of deformation without being damaged. To prevent body fluids from leaking into the device's internal structure, tight packaging is required. Next, device packaging, such as PDMS, is in close contact with skin or tissues; therefore, non-bio-toxicity that avoids a healthy immune system is essential. Finally, the most critical aspect is the control of packaging thickness to maintain the sensitivity of PENGs.

5.2. Long-term stability

Long-term stability is one of the most essential characteristics for implantable nanodevices. Surgery is required to place the device in the target place and requires anesthesia, suturing, and a thoracotomy. As a result, in vivo energy harvesters will spare patients from needless suffering and financial burden following surgery.

5.3. Power management

Power management can significantly improve the energy-harvesting efficiency of PENGs. Generally, implantable PENGs should be implanted or fixed in close contact with the human body or organs. It is possible to reduce the mechanical disparity between a PENG device and the organism, hence boosting biological safety during the prolonged implantation process, and the selection of electrical connections is vital in IMEs. Copper, gold, carbon paste, and PTFE are commonly used electrical wires, which help to improve output efficiency. Furthermore, material selection is the critical factor in enhancing the performance of PENGs. The choice of appropriate materials significantly impacts the device sensitivity, mechanical robustness, and overall electrical performance, all of which are essential for effective power management and optimizing the output power of the PENG.

5.4. Assistive technology

Recently, researchers have been focusing on how to use integrated sensors with AI applications like machine learning and remote control to do a lot of different things. It will be advantageous to increase the use of PENGs as active, self-powered sensors in sensor networks if they can integrate with other modules in sensor systems and operate on their own power in a complete wireless sensor mode, including the supporting components. An evolving future trend is that of deploying AI in PENGs to achieve the desired electrical performance.

5.5. Future solutions

In the future, addressing additional challenges will be essential to further advance the practicality and performance of PENGs:

Irregular Outputs: Researchers need to investigate and develop methods to address irregular outputs of PENGs to ensure consistent and reliable energy harvesting.

Nature of Tapping Elements: Understanding the behavior and performance of different tapping elements used in PENGs is crucial to optimize their efficiency and ensure long-term stability.

Types of Encapsulating Materials: Exploring various encapsulating materials and their effects on PENG performance can lead to improvements in biocompatibility and overall device functionality.

Energy Conversion Efficiency: Enhancing the energy conversion efficiency of PENGs is vital for maximizing energy harvesting from various sources. Researchers should focus on improving the materials, fabrication and structural designing processes to achieve higher efficiency in converting mechanical energy into electrical energy. This will result in more effective and sustainable power generation for self-powered health care electronics and medical devices.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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