



Mechanical energy harvesting and self-powered electronic applications of textile-based piezoelectric nanogenerators: A systematic review

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ABSTRACT

Environmental pollution resulting from fossil fuel consumption and the limited lifespan of batteries has shifted the focus of energy research towards the adoption of green renewable technologies. On the other hand, there is a growing potential for small, wearable, portable electronic devices. Therefore, considering the pollution caused by fossil fuels, the drawbacks of chemical batteries, and the potential applications of small-scale wearables and portable electronic devices, the development of a more effective lightweight power source is essential. In this context, piezoelectric energy harvesting technology has attracted keen attention. Piezoelectric energy harvesting technology is a process that converts mechanical energy into electrical energy and vice-versa. Piezoelectric energy harvesters can be fabricated in various ways, including through solution casting, electrospinning, melt spinning, and solution spinning techniques. Solution and melt-spun filaments can be used to develop woven, knitted, and braided textile-based piezoelectric energy harvesters. The integration of textile-based piezoelectric energy harvesters with conventional textile clothing will be a key enabling technology in realising the next generation smart wearable electronics. This review focuses on the current achievements on textile based piezoelectric nanogenerators (T-PENGs), basic knowledge about piezoelectric materials and the piezoelectric mechanism. Additionally, the basic understanding of textiles, different fabrication methods of T-PENGs, and the strategies to improve the performance of piezoelectric nanogenerators are discussed in the subsequent sections. Finally, the challenges faced in harvesting energy using textile based piezoelectric nanogenerators (T-PENGs) are identified, and a perspective to inspire researchers working in this area is presented.

1. Introduction

With the advancement of technology in electronic markets, the demand for energy harvesting has received significant attention in recent years. The demand for energy has increased due to the expansion in population worldwide [1–3]. In the past, fossil fuels were utilized mostly to accommodate the energy demand; however, fossil fuel consumption has pushed the world into a climate ‘danger zone’. Moreover, the greenhouse effect and global warming are the direct results of higher fossil fuel consumption. To work towards a solution, the entire energy market is now shifting towards renewable energy. Renewable energy is an eco-friendly energy source which does not have any harmful

environmental effects [4,5]. Interestingly, different renewable energy sources such as solar, thermal, and mechanical energy are available in our surroundings. Among them, mechanical energy is one of the most promising renewable energies owing to its abundant availability. Mechanical energy is available in different forms like wind, the flow of water, human body movement, walking, and so forth. Over the past two decades, different technologies have been utilized for harvesting mechanical energy [6–8]. These technologies include piezoelectric, triboelectric, electromagnetic, and electrostatic which are the common mechanical energy harvesting technologies. In this regard, the piezoelectric and triboelectric energy harvesting technologies are attracting more interest for harvesting the surrounding renewable energies owing

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to their several advantages like cost effectiveness, easy fabrication method, lightweight, flexibility, better energy harvesting capability, and so on [9]. Particularly, among the various energy harvesting technologies, piezoelectric energy harvesting is one of the most advantageous options [10]. The prime attractive features of piezoelectric energy harvesting technology are its reliability, easy processability, higher energy density, and cleanliness [11,12]. Piezoelectricity is a characteristic of certain materials which can generate an electric charge when the materials are subjected to mechanical strain and vice-versa.

Piezoelectric materials can be classified into two broad groups: natural and synthetic piezoelectric materials. Further, synthetic piezoelectric materials can be classified into three subgroups, such as ceramics, polymers, and composites [13]. Rochelle salt is the first naturally occurring material to exhibit piezoelectric properties [14]. Other natural piezoelectric materials include quartz, sugar, bone, and so on. Likewise, various ceramics (barium titanate), [15,16], lead zirconate titanate [17], potassium sodium niobate [18–20], bismuth and so on) and polymers (including poly(vinylidene difluoride), poly(vinylidene fluoride-trifluoroethylene), nylon-11, polypropylene, polylactic acid. etc.) also show the characteristics of piezoelectricity [21,22]. The composite or hybrid piezoelectric materials have come into the picture to compensate the individual advantages and disadvantages of ceramics and polymers, respectively [23]. To date, various composite-based piezoelectric materials have been developed by employing different fabrication methods. Moreover, among them, solution casting, melt spinning, solution spinning, and electrospinning are the most used fabrication methods.

Since wearable electronics are currently of high demand, researcher community are engaged to develop wearable power sources to supply power to various small-scale wearable and portable electronic gadgets. To date, rechargeable rigid batteries and electrochemical capacitors are generally used as energy storage devices for wearable power sources. However, due to their inherent drawbacks such as limited lifetime, frequent charging, higher disposal costs, safety risks, lower capacity, and more, they may not be appropriate for the future of wearable and portable electronics. Therefore, power harvesting directly from environmental energy sources is a perfect solution for the future of small-scale electronics [24]. Moreover, this process is economical, environmentally friendly, and long-term service oriented compared to its other counterparts [25].

To supply power to various small-scale wearable and portable electronics, textile based piezoelectric nanogenerators have drawn tremendous interest from research community due to their unique features, including softness, structural flexibility, light weight, low cost, and abundant availability. Although solid film based piezoelectric nanogenerators may generate higher piezoelectric responses, their rigid or brittle structure compared to textiles, limits their utility [24,25]. The textile based piezoelectric nanogenerator is more suitable for the future of wearable electronics, as the piezoelectric device made of textile materials can be attached to wearable clothing as a patch or alternatively, textile materials (fibres/filaments) can be directly incorporated into the wearable textiles/clothes. As a result, wearable smart textiles can be developed in both ways.

This review systematically explains the different possible piezoelectric materials, including ceramics and polymers, as well as providing a fundamental understanding of textiles and the basic principles of textile based piezoelectric devices. We subsequently discuss the recent progress on the textile-based piezoelectric nanogenerators (PENG) in the fields of both electromechanical energy harvesting and self-powered electronic devices. Moreover, we highlight existing challenges and prospects in textile-based piezoelectric nanogenerators. This review not only provides insights into the most recent technological advances, but also directs scientists towards the development of materials with improved performance for next-generation piezoelectric nanogenerators. It also provides ideas on the various avenues for future work in this direction.

2. Need for energy harvesting

Global energy demand has dramatically increased as a result of technological advancements in the manufacturing of various electronic goods and the rapid evolution of populations [26]. As per the U.S. Energy Information Administration (EIA) forecast, the total energy demand (region-wise) is projected to increase significantly from the year 2015–2035 (see Table 1) [27]. To date, the battery is used as a power source in many electronic products. However, batteries have a limited life span which restricts their applications in the electronic market [6,7]. As a result, the battery and renewable energy harvesting systems can work together to provide continuous power to small portable or wearable electronics. As the energy demand is higher, research has shifted towards harvesting energy in-line with sustainable development goals (SDGs). Thus, the global energy demand can be fulfilled by harvesting energy from different renewable energy sources. The total energy sources have been classified into non-renewable and renewable energy, respectively [28–30]. Non-renewable energy sources are categorized into two subgroups such as fossil fuels (oil, natural gas, and coal) and nuclear energy.

Use of fossil fuels causes air pollution and global warming through the emission of carbon dioxide (CO₂), methane (CH₄), and heat generation as well. Similarly, renewable energy sources can also be classified into different subcategories like solar energy, thermal energy, hydro, wind, biomass, tidal, and ocean energy [10]. Recently, attention has been paid to harvesting energy from renewable sources rather than non-renewable sources to solve the total world energy demand by reducing air pollution and the risk of global warming [6]. Renewable energy can be extracted from different natural sources, as mentioned above by using various technologies. In the year 2018, renewable energy consumption was very low as per the survey done by the European Union (EU) (Fig. 1a). At that time, renewable energy was shared from various sources, as shown in Fig. 1b [31]. Among different renewable energy sources, mechanical energy (vibration, wind, water flow, and human body movement) can be easily harnessed from our environment [11]. Specifically, the harvesting of mechanical energy from human body movement and energy from walking is a promising route towards the realisation of self-powered wearable electronic devices and sensors.

Table 1
Projected total world energy consumption [27].

Region/Country	2015	2020	2025	2030	2035
OECD*					
OECD Americas	126.1	131	135.9	141.6	147.7
US	102	104.9	108	111	114.2
Canada	14.6	15.7	16.4	17.6	18.8
Mexico/Chile	9.5	10.4	11.5	13	14.7
OECD Europe	83.6	86.9	89.7	91.8	93.8
OECD Asia	40.7	42.7	44.2	45.4	46.7
Japan	22.2	23.2	23.7	23.7	23.8
South Korea	11.1	11.6	12.4	13.1	13.9
Australia/New Zealand	7.4	7.8	8.1	8.5	8.9
Total OECD	250.4	260.6	269.8	278.7	288.2
Non-OECD					
Non-OECD Europe and Eurasia	51.4	52.3	54	56	58.4
Russia	31.1	31.3	32.3	33.7	35.5
Other	20.4	21	21.7	22.3	22.9
Non-OECD Asia	188.1	215	246.4	274.3	298.8
China	124.2	140.6	160.9	177.9	191.4
India	27.8	33.1	38.9	44.3	49.2
Other	36.2	41.3	46.7	52.1	58.2
Middle East	31	33.9	37.3	41.3	45.3
Africa	21.5	23.6	25.9	28.5	31.4
Central and South America	31	34.2	38	42.6	47.8
Brazil	15.5	17.3	19.9	23.2	26.9
Other	15.6	16.9	18.1	19.5	20.8
Total non-OECD	323.1	358.9	401.7	442.8	481.6
Total World	573.5	619.5	671.5	721.5	769.8

OECD* : Organisation for Economic Co-operation and Development

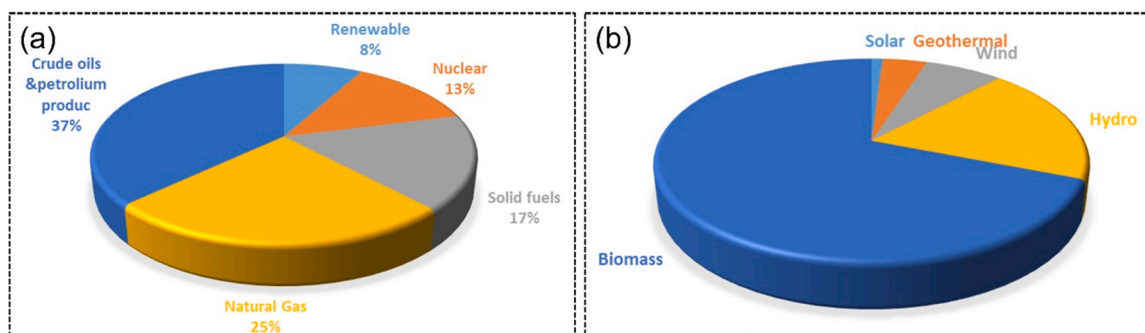


Fig. 1. (a) Energy consumption from the different sources, and (b) energy consumption from the renewable sources [31].

Further, the technique of harvesting energy from mechanical strain and converting this energy into electrical energy is called piezoelectric energy harvesting. Piezoelectric energy harvesting techniques have shown great promise in fulfilling the demand for energy in different portable and electronic goods where the demand for power is low [11].

3. Basic understanding of the textiles and electronic textiles

Textiles can be classified according to the raw material types, dimensions, and manufacturing methods. The applications of textiles mainly depend on their dimensions and properties. Dimensionally, textiles can be categorised into 0D, 1D, 2D, and 3D whereas, in terms of manufacturing methods, textiles can be classified as woven, knitted, braided, and nonwoven.

The basic unit of textile manufacturing is the fibre (0D) (Fig. 2). There are certain attributes that are essential in order to turn a fibre into a textile product. These are often referred to as fibre forming characteristics. Aspect ratio (length to breadth ratio more than 1000:1), molecular weight, linearity, configuration, orientation, and crystallinity are

few of the many fibres forming characteristics. Fibre can be broadly classified into two groups: natural and manmade fibres. Natural fibre again can be classified into the various groups as per their sources such as seed fibre (cotton), leaf fibre (sisal), bast fibre (jute), fruit fibre (coir), and so on. Similarly, manmade fibre can also be classified into the two groups organic and inorganic fibre. Organic fibres are mainly two types, natural polymer and synthetic polymer base. On the other hand, inorganic fibre are glass fibre, metallic fibre, and specialty fibres. This fibre is converted into yarn (1D) by the spinning process (Fig. 2).

Fibres are formed into yarns by the process of spinning. Based on fibre length, fibres may be spun into spun yarn or filament yarn. The spun yarn is made of fibres with a defined length, while filament yarns have a continuous fibres running along the full length of the yarn. Generally, natural fibres develop into spun yarn, while synthetic fibres form filament yarns. In the spinning of natural fibre-based yarn, a twisting operation is carried out. This helps to improve the fibre coherence during the yarn formation process. While synthetic yarn can be developed by various spinning methods like melt spinning, wet spinning, and dry spinning. Synthetic polymer chips are melted by an



Fig. 2. Textile fibres, yarns, and various textile fabrics.

extruder in melt spinning followed by melt spun filament formation. In wet spinning, a polymer solution is coagulated by using a nonsolvent so that wet spun filament can be formed. Then these yarns are used to make the textile fabric having different structures like woven, knitted, and braided (Fig. 2).

Textile fabrics (2D and 3D) can be manufactured by four different methods such as weaving, nonwoven, knitting, and braiding. When two types of yarn (warp and weft) are interlaced with each other, it is called a woven structure, and the process is referred to as weaving. The manufacturing of woven fabric depends on the textile design. As per this design, woven fabric can be called plain, twill, satin and many more. In the same way, when two yarns (wale and course) are interlaced each other, the structure is called a knitted structure, while the process is called knitting. The knitting process also has various designs like plain, rib, purl, interlock, etc. A nonwoven fabric structure is developed by the interlocking of the fibres (Fig. 2). Needle punching is one of the most promising nonwoven fabric manufacturing methods. Finally, in the braiding fabric manufacturing method, two or more yarns are braided together by using a specific machine [32–35]. Further, a classification of the different textiles in viewpoints of their dimensions and characteristics is highlighted in a tabular form (Table 2).

Nowadays, wearable electronic textiles (smart textiles) have drawn great interest owing to their multifunctional applications such as heat monitoring, health monitoring, touch, sensing, and many more. Since the present review focuses on the textile based piezoelectric nanogenerators intended for environmental mechanical energy (including biomechanical) harvesting and self-powered applications, an understanding of the fabrication of the wearable electronic textiles is an essential task. There are different manufacturing techniques such as embroidery, sewing, weaving, knitting, nonwoven, braiding, spinning, coating, plating, and printing [36–38]. All the manufacturing techniques have some merits and demerits, as presented in Table 3.

4. Piezoelectric materials

A piezoelectric material characteristically generates electric polarization when an external pressure is applied to it, and vice-versa [39–42]. Piezoelectric materials are broadly classified into two groups such as natural and manmade. Natural piezoelectric materials include quartz (SiO_2), topaz, tourmaline group minerals, Rochelle salt, and some natural organic materials such as silk, wood, rubber, dentin, bone, hair, and enamel. Rochelle salt was the first material to exhibit piezoelectricity, discovered by the Curie brothers in 1880 [43]. Manmade piezoelectric materials are classified into three types like polymers, ceramics, and composite-based materials. Table 4 shows different piezoelectric materials including natural and manmade piezoelectric materials. All piezoelectric materials have advantages and disadvantages. For instance, natural crystal based piezoelectric materials have a high mechanical quality factor (Qm), but they are expensive and difficult to

Table 2
Classification of the different textiles in terms of their dimensions, structural characteristics, merits and demerits.

Type of textiles	Dimension	Characteristics	Merits	Demerits
Staple fibre and filament	0D	L/D ratio: 1000:1, building entity of textiles	Flexible, micro or nanoscale size	Low strength
Yarn / thread (-staple spun yarn, monofilament yarn, and multifilament yarn)	1D	Continuous length of the interlocked fibres made by twisting and winding, and utilize to make textile fabrics	High strength, flexible, and elasticity	Low dimensional stability
Woven fabric (plain, twill, satin, and sateen)	2D and 3D	Made of yarns / threads through interlacement method	High dimensional stability, higher strength than fibre and yarn, high durability	Woven fabric is stiffer in nature
Knitted fabric (plain, rib, purl, and interlock)	2D and 3D	Made of yarns/threads through interlocking method	High stretchability, high strength, enough flexibility	Lower durability compared to woven fabric
Braiding (bi-axial and tri-axial)	2D and 3D	Made of yarns/threads through intertwining method	Higher strength than fibres or yarns, high stretchability	It can be stretched only in one direction
Nonwoven (needle punch, spun bond, melt bond, and electrospinning)	2D and 3D	Made directly from fibres by entanglement method	More flexible, higher specific surface to volume ratio (electrospun fibres)	Lower dimensional stability

Table 3
Summary of the different manufacturing techniques of the wearable electronic textiles.

Techniques	Merits	Demerits
Embroidery	Material cost is low	High machinery cost, complex processability, difficult to wear
Sewing	Low material cost, low machinery cost, lower complexity in processing	Difficult to wear
Weaving	Lower machinery cost	Higher material cost, higher process complexity, higher resistance to wear
Knitting	Lower machinery cost, easy to wear	Material cost is high, higher process complexity
Nonwoven	Material cost is low, lower process complexity, easy to wear	Machinery cost is high
Braiding	Low machinery cost, low material cost, and easy to process	Difficult to wear
Spinning	Low machinery cost, low material cost, easy to wear and process	Some of the spinning machineries is expensive and electrically or thermally hazardous like electrospinning (high electric field is required), melt spinning (high temperature is required)
Coating	Low material cost, easy to wear and process	Machinery cost is high, low speed (doctor blade coating), no controlled in thickness (solution cast)
Plating	Higher conductivity, higher speed, etc.	Lower stability, higher cost
Printing	Process complexity is lower and easy to wear	High material and machinery cost

process. On the other hand, ceramic based piezoelectric materials show a higher piezoelectric constant, but their brittle characteristics limit their usability. Similarly, polymer based piezoelectric materials show higher flexibility, but they have lower piezoelectric constant as compared to ceramic based piezoelectric materials. Furthermore, composite based piezoelectric materials are suitable for piezoelectric applications because they can show a synergistic effect of polymer and filler; however, there is a chance of neutralization effect due to inconsistent polarization direction [25].

Another important aspect is the toxicity of piezoelectric materials. The use of toxic materials is now discouraged and avoided, considering their detrimental effect on the surrounding environment. Therefore, the researchers working in the concerned area are advised to keep in mind the toxicity effect of their respective materials. One such piezoelectric material is PZT. It is well known that the PZT is one of the most promising piezoelectric materials to date because it exhibits higher piezoelectric properties. However, it contains more than 60% lead which is toxic in nature; hence, it is not an eco-friendly material [49,50].

Table 4
Different piezoelectric materials.

Piezoelectric Materials				
Natural	Manmade			
Topaz	Inorganic (ceramics)		Organic (polymers)	Polymer composites
	Lead based	Lead free [44]		
Quartz Crystals	Lead titanate (PT) [45,46]	Barium titanate (BT)	Poly(vinylidene difluoride) (PVDF)	PVDF/PZT
Rochelle Salt	Lead zirconate titanate (PZT)	Cadmium sulphide (CdS)	Poly(vinylidene fluoride-trifluoroethylene) (P(VDF-TrFE))	PVDF/ZnO
Topaz [47]				
Tourmaline group minerals	Lead lanthanum zirconate titanate (PLZT)	Potassium niobate (KNbO ₃)	Poly(vinylidene fluoride-tetrafluoroethylene) (P(VDF-TeFE))	PVDF/KNN
Wood	Lead magnesium niobate (MgNb ₂ O ₉ Pb ₃)	Lithium niobate (LiNbO ₃)	Nylon 11	PVDF/AgNWs
Silk		Lithium tantalate (LiTaO ₃)	Urea	PVDF-TrFE/MWCNTs
Rubber		Passium sodium niobate (KNN)	Poly(lactic acid) (PLA)	PVDF/BT NPs
Dentin		Molybdenum disulphide (MoS ₂)	Polypropylene (PP)	PVDF-TrFE/PZT
Bone		Zinc stannate (ZnSnO ₃)	Cellulose acetate (CA)	PVDF/MoS ₂
Hair		Iron oxide (Fe ₃ O ₂)	Cellulose	PVDF/ZnSnO ₃
Enamel		Zinc ferrite (ZnFe ₂ O ₄)	Polyacrylonitrile (PAN)	PVDF/Fe ₃ O ₂
		Zinc oxide (ZnO) [48]		

Therefore, to compete with PZT, KNN has been used tremendously by the different researchers. KNN exhibits appreciable piezoelectric properties, however, it is inferior in comparison to PZT. Moreover, to avoid the toxicity problem, researchers have also tried to develop other suitable piezoelectric materials based mainly on piezoelectric polymers (PVDF, its co-polymers). Therefore, researchers should take care of these important issues in their future research works.

5. Working principle of piezoelectric nanogenerators (PENGs)

The process by which electrical energy is generated from ambient mechanical energy (such as stress, strain, and vibration) is known as piezoelectric energy harvesting. Before discussing the working principle of the piezoelectric nanogenerator, it is essential to understand the mechanism of piezoelectricity. A systematic explanation of the theory of the piezoelectricity is provided to highlight the same [51–53]. Piezoelectricity is the combined effect of the linear behaviour of the piezoelectric material and Hooke's law for linear elastic materials. The respective mathematical equations for these two characteristics are as follow:

$$D = \epsilon E \Rightarrow D_i = \sum_j \epsilon_{ij} E_j$$

$$S = sT \Rightarrow S_{ij} = \sum_{k,l} s_{ijkl} T_{kl}$$

where, D is the electric flux density [54] (electrical displacement), ϵ is the permittivity (free body dielectric constant) and E is the electric field strength. Also, S and s is the linearized strain and compliance under short-circuit conditions and T is the stress applied.

These two equations may be combined into a coupled equations, of which strain-charge form is:

$$S = sT + \partial^t E \Rightarrow S_{ij} = \sum_{k,l} s_{ijkl} T_{kl} + \sum_k \partial_{ijk}^t E_k$$

$$D = \partial T + \epsilon E \Rightarrow D_i = \sum_{j,k} \partial_{ijk} T_{jk} + \sum_j \epsilon_{ij} E_j$$

where, ∂ is the piezoelectric tensor and the superscript t stands for its transpose. Due to the symmetry of ∂ , $\partial_{ijk}^t = \partial_{kji} = \partial_{kij}$.

In matrix form,

$$\{S\} = [s^E] \{T\} + [d^t] \{E\}$$

$$\{D\} = [d] \{T\} + [\epsilon^T] \{E\}$$

where, $[d]$ is the matrix for the direct piezoelectric effect and $[d^t]$ is the matrix for the converse piezoelectric effect. The superscript E indicates a zero, or constant, electric field; the superscript T indicates a zero, or constant, stress field; and the superscript t stands for transposition of a matrix [55,56].

All the ferroelectric materials belong to the class of piezoelectric materials. Ferroelectric materials have an oriented molecular structure so that the material can exhibit a local charge separation, known as an electric dipole. The electric dipole density is denoted by P , which can be calculated by summing up the dipole moments per unit volume. Under equilibrium conditions, the electric dipoles in the material remain arranged in a haphazard condition. Upon heating the material at a critical temperature, under the application of an external electric field, the electric dipoles in the material, tend to arrange in the direction of the electric field applied. This process is termed “poling” and poled materials maintain dipole orientation even after cooling. The materials will have an inherent piezoelectric effect once the poling process is completed.

Moreover, the piezoelectricity of materials depends upon the change of dipole density when they are subjected to mechanical stress. Dipole density changes because of the reconfiguration or rearrangement of the dipoles inside the material in a particular direction. The performance of piezoelectricity can be varied due to the alternation of physical factors like strength of polarization, dipole direction, and applied stress.

“Therefore, it is very crucial to know the working principle of the piezoelectric nanogenerator for performing research in this field. Generally, in the case of a piezoelectric nanogenerator, piezoelectric material is sandwiched between two electrodes, namely the bottom and top electrodes. Fig. 3 explains how a textile based piezoelectric nanogenerator works (Fig. 3). At the initial stage, in other way before applying any impact on the nanogenerator, there is no polarization effect in piezoelectric textiles (Fig. 3i). This is due to the cancelation and neutralization of the cations and anions presence in the piezoelectric materials. When an external pressure is applied to the piezoelectric device, the piezoelectric textile gets compressed and therefore, the centres of charges in the piezoelectric fibrous material are separated. This is because the net dipole moment gets changed, and an electrical potential difference takes place across the two electrodes. At this moment, if electrodes (for example conductive fabric) are connected with the external conductive wires, then charge gets flow from one electrode to other and after some time, an equilibrium state is achieved (Fig. 3ii). In this way, mechanical energy converts into electrical energy.

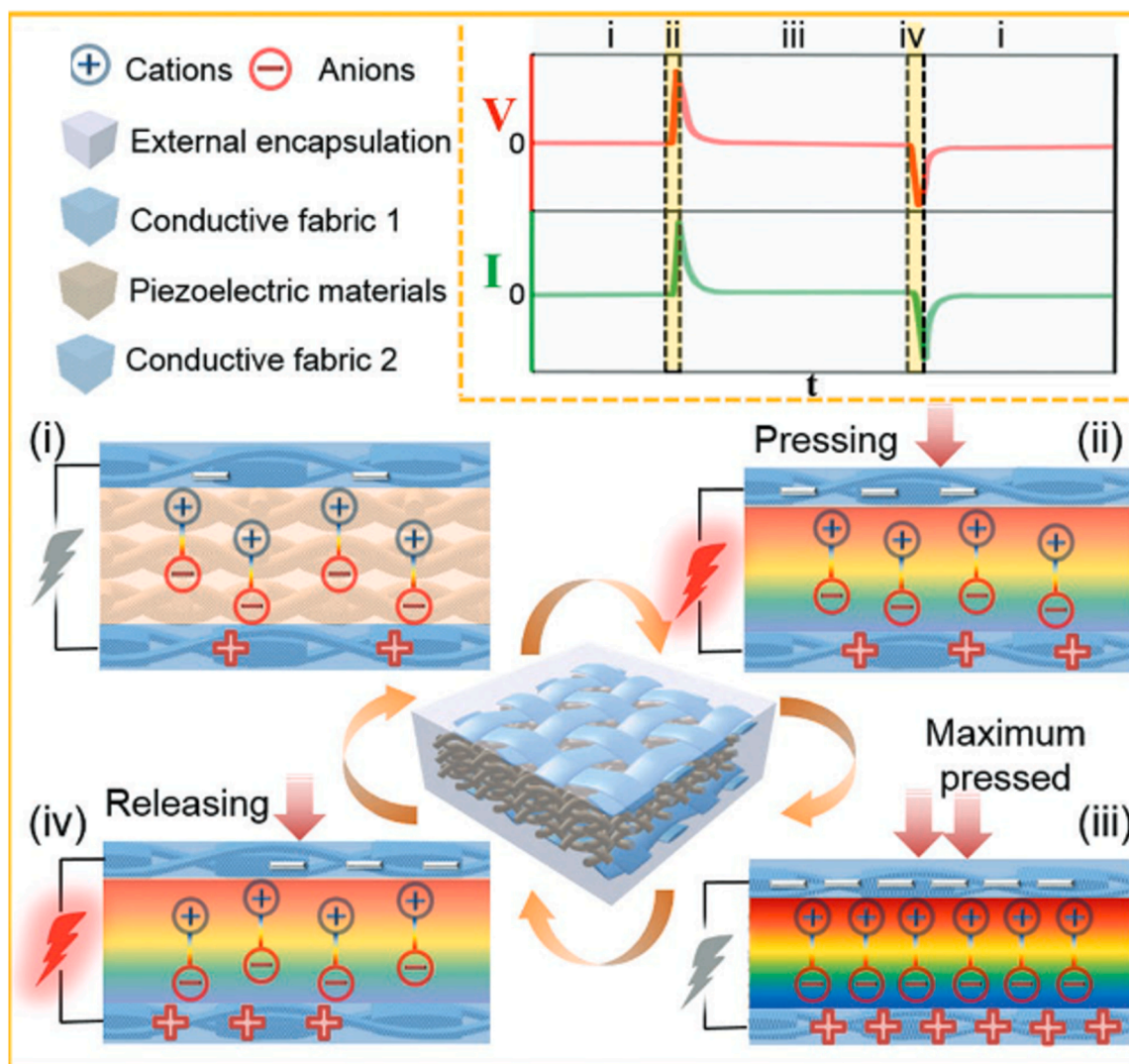


Fig. 3. Schematic diagram of the working mechanism of a textile fabric-based piezoelectric nanogenerator. Reprinted with permission from ref. [57]. Copyright 2020 John Wiley and Sons.

If pressure is applied continuously to the piezoelectric device, then a maximum stressed condition will occur, and therefore the dipole density will be higher at that stage (Fig. 3iii). Finally, when the applied pressure is released from the textiles, electrons will flow back to rebalance the charge induced by the strain released at the short-circuit condition (Fig. 3iv). Therefore, a steady state sinusoidal current will flow through an external circuit when a continuous reciprocating force (applying and releasing) is applied to the T-PENG. This mechanism is well described by Dong et al. as also represented in Fig. 3 [57].

6. Textile-based piezoelectric nanogenerator (PENGs)

The piezoelectric nanogenerator (PENG) is a special type of energy harvesting device that has the capability to convert nanoscale mechanical energy into electrical energy with the help of the piezoelectric effect. Interestingly, this converted electrical energy can be stored in storage devices like batteries and capacitors, and can be utilized as power sources in different portable, wireless, and wearable electronic devices (which need a very small amount of power to work smoothly) [58]. Since the human body is one of the most promising sources of mechanical energy, a lot of recent research has focused on wearable electronics. In this context, textiles could be a great interaction medium between electronic devices and the human body. Either an electronic

device can be integrated into the textile, or a direct textile based electronic devices can be developed for wearable electronic applications. Textiles are one of the most preferable candidates for wearable electronics because they are more comfortable, air permeable, and possess the required flexibility, etc.

The PENG could be developed using different forms of piezoelectric materials such as pure ceramic-based materials, polymer based, and composite materials as mentioned in the introduction part of this review. Since wearable electronic devices are rapidly becoming more prevalent, a suitable material selection for these devices is an important factor. Therefore, textile-based substrates could be a good candidate for making wearable electronic devices. Polymer based piezoelectric materials are comparatively better than ceramic materials, but polymer materials also have intrinsic drawbacks. In order to avoid drawbacks with the polymer materials, composite materials are used to provide a desired combination of piezoelectric properties and flexibility. A good piezoelectric performance and flexibility of the composite materials provide a path in the field of wearable electronics. However, the flexibility of composite materials-based nanogenerators is insufficient for use in remote areas [59]. To overcome this problem, the textile-based piezoelectric nanogenerator has drawn significant recent interest (because textile-based nanogenerators are flexible, lightweight, low cost, and stretchable). Due to the significant characteristics of the

textile-based piezoelectric nanogenerator, it can be easily integrated into clothing, shoes, and fashion accessories [60]. Furthermore, the use of wearable electronic devices has also become an increasing trend recently. For instance, data analysis and communication via wearable electronic goods have important real-world applications when used with the human body (e.g. health monitoring, etc.). In addition, smart textiles, especially in the field of piezoelectric energy harvesting and self-powered electronic devices, are also an increasing trend due to the development of technology in wearable electronic goods. In comparison to the development of technology, human consciousness about smart textiles is minimal. Therefore, the development of portable and wearable electronic goods is important to cover the power demand in different smart electronic products [61–64]. However, the development history, working principle, and critical analysis of the textile-based nanogenerator are still not clearly explored and reviewed. Moreover, selection of proper materials and manufacturing methods, for preparing textile-based nanogenerators also need to be looked into. In this section, the progress and challenges of fibres, yarns, and textile based piezoelectric nanogenerators are discussed systematically.

6.1. Fibre-based piezoelectric nanogenerator (PENGs)

6.1.1. Electrospun nanofibre-based PENGs

The electrospun nanofibre is produced by the electrospinning method. In the electrospinning method, the solution of polymer is poured into a plastic syringe. Thereafter, this plastic syringe is placed in the motor driven pump. Here, an external voltage is applied between the syringe needle tip and collector (where electrospun nanofibres are deposited). Due to this external electric field, a nanofibre jet is generated from the needle tip and deposits on the collector. In electrospinning process, different type of collectors such as flat plate, drum, disc type, etc., can be used. Electrospun nanofibres have a higher specific surface area to volume ratio compared to conventional macro or micro fibres or filaments. The higher specific surface to volume ratio of electrospun nanofibres results in numerous applications, such as in electronics, biomedical engineering and food packaging, etc. [65–69].

In recent years, piezoelectric nanogenerator based on electrospun nanofibres is one of the most promising research areas. Electrospun nanofibre-based piezoelectric nanogenerators have many advantages: they enable higher output signals and have no requirement for additional poling unlike solution cast and melt-spun materials due to the in-situ poling that occurs in the electrospinning process itself. Dipoles get oriented in a particular direction as per the applied external electric field in electrospinning and therefore do not need extra poling equipment [70–74]. Furthermore, electrospun nanofibre-based piezoelectric nanogenerators are lighter, more flexible, and less expensive than solution/melt-cast films or melt-spun filaments. Due to these advantages of electrospun nanofibres, many researchers are using electrospun nanofibres as piezoelectric materials throughout the world. There are two electrospinning methods, Far Field Electrospinning (FFES) and Near Field Electrospinning (NFES). In FFES, the distance between the needle tip and collector is longer as compared to NFES. In both cases, there are some advantages and disadvantages due to their different process lines [75–78]. Although a surge of research has already been done using both FFES and NFES based piezoelectric materials, it is not yet clear which method has a greater piezoelectric response. To evaluate the piezoelectric performance of these piezoelectric materials (FFES and NFES), a systematic review of these materials is summarized here. This section explains NFES-based piezoelectric materials first, then FFES.

In 2019, Yu et al. [79] studied the concept of far-field electrospinning (FFES) and near-field electrospinning (NFES) of the PVDF polymer. Furthermore, the authors also investigated the piezoelectric responses of FFES and NFES piezoelectric materials. In case of the FFES method, the gap between the tip of the needle and substrate was 10 cm. Whereas, the distance between needle tip and collector was shortened to 2 mm in the NFES method. Since the distance is lower in the case of the near electric

field electrospinning method, a strong polymeric jet whip can reach the collector at an earlier time. Therefore, the structure containing the deposited nanofibres is arranged randomly with the help of a circulating drum. Fig. 4a(i) shows PVDF nanofibres with the help of an FFES rotating drum. The IR analysis was carried out to evaluate the β -crystalline phase of the developed electrospun webs. It was found that the β fraction value ($\sim 83.5\%$) was higher in the case of the FFES method as compared to the NFES method ($\sim 64.79\%$). This is because of lower mechanical stretching in NFES than FFES based electrospun webs. Moreover, the researchers checked the similarity with the piezoelectric characteristics of such samples by applying force at the right angle direction to the layers with contained electrodes. At the same time, they fabricated a nanogenerator incorporating attached FFES PVDF nanofibres. The attached PVDF nanofibres manufactured with FFES offer good electrical based pressure sensing properties. As seen in Fig. 4a(ii), a process to prepare a nanofibre containing PVDF containing a layer having a thickness of $110\ \mu\text{m}$ which were shaped in the form of a shoe pad containing the size of US8.5 (or EU42). As the given force was applied perpendicular to the layer; therefore, at the time of wearing, an aluminium foil containing electrodes was accompanied to extract the electrons. Fig. 4a(iii) demonstrates that, while a person walks wearing such a shoe pad, the open circuit voltage is lesser than when running. This is likely due to the higher impact force and frequency when the person is running as compared to walking. The nanogenerator made by the FFES based electrospun web showed an optimum power of $\sim 6.45\ \mu\text{W}$ having a load of $5.5\ \text{M}\Omega$ [79]. In another study, Lee and co-workers [80] reported a self-powered aquatic sensor based on the PVDF NFES piezoelectric fibres as depicted in Fig. 4b. To harvest sound energy the developed nanogenerator was attached to the speaker and interestingly it was found that the nanogenerator had an output voltage of $0.13\text{--}0.23\ \text{V}$ with corresponding to the sounds level of $60\text{--}120\ \text{dB}$. Also, the developed nanogenerator was used to detect different voices by attaching on the throat. A significant response was found with different kind of hums as shown in Fig. 4b (below image).

In NFES method, one promising problem is the deposition of fibres at the subsequent sites, since charges of the deposited fibres made them ground very quickly. To solve this problem, a third medium mainly a porous paper is required on the conductive collector. Fuh et al., in 2016 [81] studied the piezoelectric performance of a 3D PVDF NFES piezoelectric fibres (Fig. 4c). A printing paper was pasted at the center of the conductive collector (Fig. 4c(i)). When electrospun fibres are deposited on the paper, the residual solvents of the electrospun fibres can infiltrate the paper substrate, which results improvement of the charge transfer between the deposited fibres and the collector. In this arrangement, it was found that the developed piezoelectric nanogenerator can generate an output voltage of $4\ \text{V}$ and the current of $100\ \text{nA}$. The authors also demonstrated the application of the developed piezoelectric nanogenerator as a human motion sensor. To detect human motion, nanogenerator was attached on the underarm of clothing and an arm was moved at the different angles. It was found that the motion sensor can generate different levels of output signals with the different angles of the arm (as depicted in Fig. 4c(ii)).

To further improve the piezoelectric response of NFES electrospun fibres, stacking of the fibres during electrospinning, was carried out. One such example, Lo et al., in 2021, [82] developed a 3D stacked P(VDF-TrFE) NFES nanofibrous porous web based self-powered smart sensor (Fig. 4d(i)). In their study, electrical response was evaluated of the different materials such as pure P(VDF-TrFE), graphene modified P(VDF-TrFE), and oil modified P(VDF-TrFE), respectively. And it was found that $3\ \text{wt}\%$ graphene modified P(VDF-TrFE) based sensor showed 2.7 times better output (voltage $3.8\ \text{V}$ and current $243.6\ \text{nA}$) compared to pure P(VDF-TrFE) based piezoelectric sensor (voltage $1.4\ \text{V}$ and current $125.9\ \text{nA}$). Finally, it was explained that the developed sensor can monitor the foot pressure very accurately (as shown in Fig. 4d(ii) & (iii)).

In NFES, there are many challenges, such as less time for evaporating

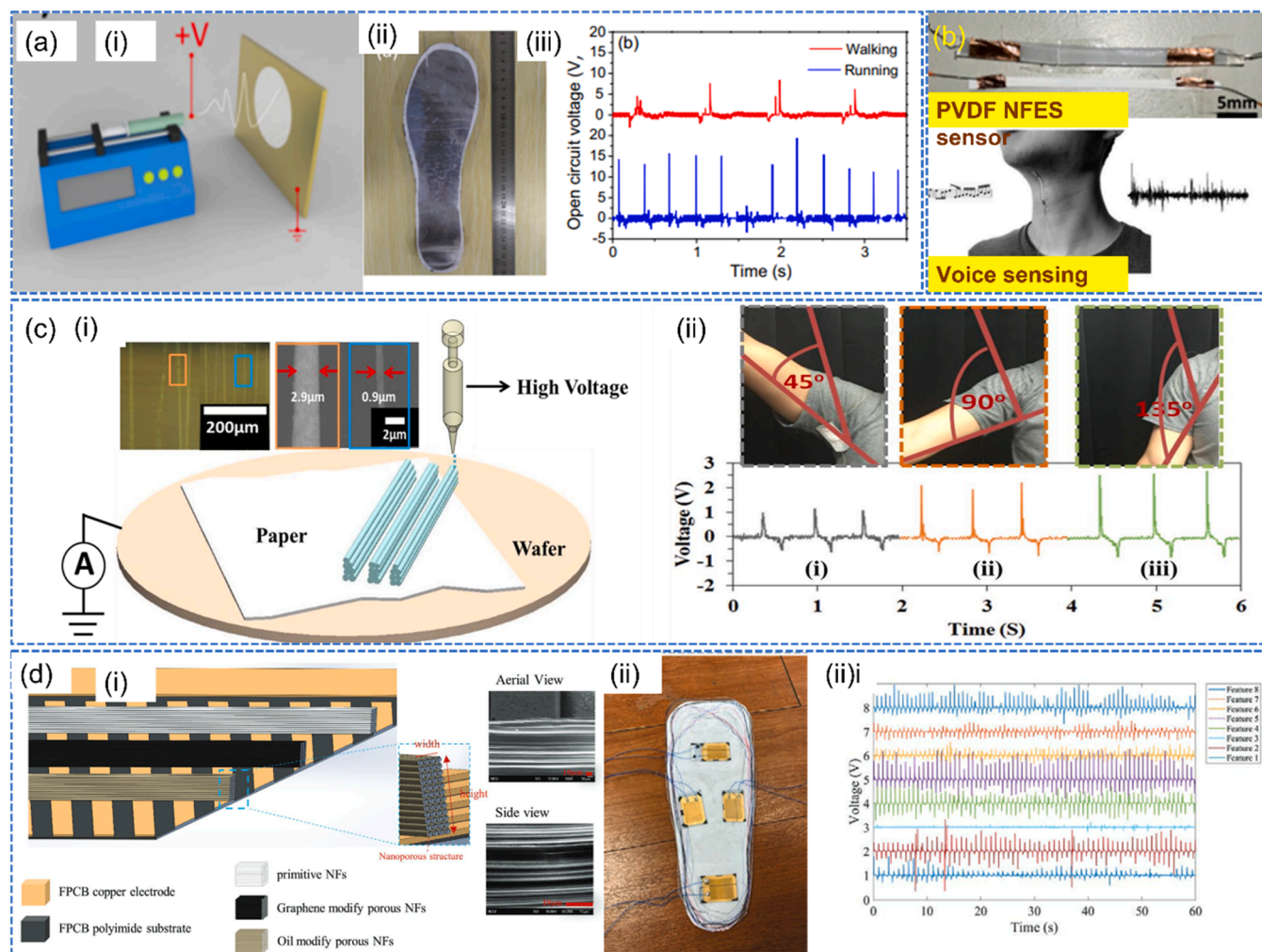


Fig. 4. a.(i) FFES setup, (ii) shoe pad fabricated by using FFES fibres, (iii) output voltage generated by FFES fibres-based PENG during walking and running. b. PVDF-TrFE fibres-based sensor and its application to detect voice of a human being. Open Access [80]. c. (i) The deposition of PVDF-TrFE NFES fibres on a printing paper, (ii) the output signals corresponding to the movement of the human arm. d. (i) The piezoelectric sensor based on the layer-by layer stacking of the PVDF-TrFE electrospun fibres, (ii) a digital photo of the smart gait sensing shoe, (iii) The signal corresponding to the pressure generated inside the shoe.

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residual solvents, lower amount of stretching on the electrospun fibre as the flying time is shorter, and so on. These challenges result in lower piezoelectric properties for the electrospun nanofibres. Therefore, researchers have tried to develop electrospun nanofibres by the FFES method. For instance, Persano et al. [83] reported a flexible piezoelectric nanogenerator based on PVDF-TrFE electrospun nanofibres (Fig. 5a (i)). High density nanofibres having a particular orientation have been developed and this high density nanofibre web was used to develop a flexible piezoelectric nanogenerator. The developed piezoelectric nanogenerator showed excellent mechanical properties (bending and twisting). This nanogenerator can generate an output voltage of 1.5 V and a current of 40 nA under the bending conditions. In addition, the developed nanogenerator was used in different wearable sensors applications (Fig. 5a (ii) & (iii)) for examples (i) pressure sensor: the as prepared piezoelectric device was utilized as a pressure sensor which showed better sensitivity even at very low pressure (i.e., 0.1 Pa), (ii) vibration/ acceleration sensor: in this case, the nanogenerator was used as a diaphragm across a hole opened in an underlying plastic film, sealed over a closed transparent box. Then a vibration is generated in the electrospun piezoelectric material by supplying of sound pressure having a capacity of 60–80 dB. As a result, the piezoelectric nanogenerator has shown an output voltage corresponding to input sound intensity,

and (iii) an orientation sensor: the electrospun nanogenerator was also used as an orientation sensor. And it was found that this sensor can sense even a very small inclination. Recent developments in pressure sensors with very small dimensions (μm to nm) have drawn a lot of interest and increased the potential applications of piezoelectricity [84,85]. Particularly, methods including nanostructured devices like different nano-related products comprising wires [86,87], rods [88,89], fibers [90–95] and sheets [96] having 1D or 2D structural forms. Moreover, nanofibres can be electrospun easily, which produces a phase containing liquid substances that can be stretched into a fiber by applying a continuous and strong electric field. Poly(vinylidene fluoride) (PVDF) and its copolymer, poly(vinylidene fluoridetrifluoroethylene) (PVDF-TrFE) exhibit semi crystallinity, demonstrating a wide range of uses based on nanofibrous related pressure electric sensors [97–100]. Likewise, Kim et al. [101] also reported the effective and non-complex processes (multiple layers of the electrospun mat and integration of the microbead electrodes with the electrospun mat) to improve the resultant function of the piezoelectric nanogenerator composed of PVDF-TrFE electrospun mat, as shown in Fig. 5b. Such multilayered piled piezoelectric devices can enhance the resultant output voltage and current since in that nanogenerator multiple number of nanofiber mats are placed together. The engineered piezoelectric nanogenerator offers an

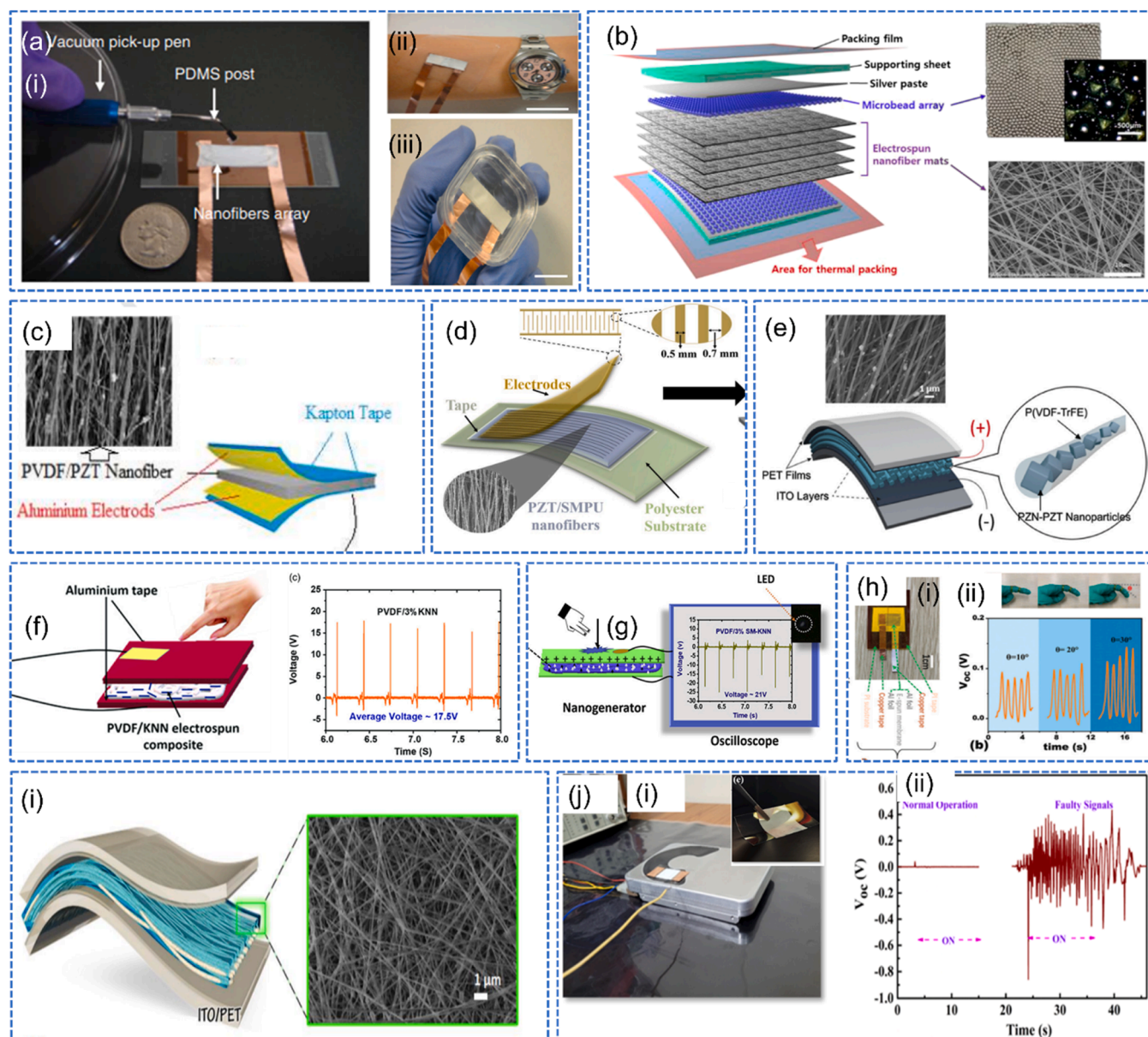


Fig. 5. a. (i) P(VDF-TrFE) electrospun nanofibres based PENG, (ii) the digital photograph of a P(VDF-TrFE) electrospun nanofibres based accelerometer. Open Access [83]. b. The schematic diagram of the electrospun PVDF-TrFE fibres mat and microbeads electrodes based piezoelectric nanogenerator. c. PVDF/PZT electrospun nanocomposite fibres-based PENG. d. SMPU/PZT electrospun nanocomposite-based PENG. e. PZN-PZT/P(VDF-TrFE) electrospun nanocomposite fibres-based PENG. f. PVDF/KNN electrospun nanofibres based PENG. g. PVDF/SM-KNN electrospun nanocomposite fibres based PENG. h. (i) AIN/PVDF-TrFE electrospun membrane based PENG, (ii) the application of the AIN/PVDF-TrFE based PENG. i. BT NPs/PVDF-TrFE electrospun membrane based PENG (inset shows a SEM image of the developed electrospun fibre). j. (i) The application of PVDF-BT electrospun nanocomposite fibre based PENG as a vibration sensor (inset shows the digital photo of developed PENG), (ii) The response signal of the developed PENG against the harddisk vibration. (b) Reprinted with permission from ref. [101]. Copyright 2018 American Chemical Society. (c) Reprinted with permission from ref. [102]. Copyright 2020 Elsevier. (d) Reprinted with permission from ref. [103]. Copyright 2020 Elsevier. (e) Reprinted with permission from ref. [104]. Copyright 2020 Elsevier. (f) Reprinted with permission from ref. [105]. Copyright 2019 John Wiley and Sons. (g) Reprinted with permission from ref. [106]. Copyright 2020 Elsevier. (h) Reprinted with permission from ref. [107]. Copyright 2021 Royal Society of Chemistry. (i) Reprinted with permission from ref. [108]. Copyright 2016 Elsevier. (j) Reprinted with permission from ref. [109]. Copyright 2022 American Chemical Society.

output voltage and current of 10.4 V and 2.3 μ A, respectively. To prove its practical ability, the authors have also explored the application of the developed piezoelectric nanogenerator as an effective pressure sensor. Interestingly, the nanogenerator was found to have good pressure sensitivity even at the very low pressure of 100 Pa. All these three above discussed works only focused on the fabrication of the flexible piezoelectric nanogenerator based on a pure polymer electrospun web. There are also many studies where researchers have reported the electrospun nanocomposite fibrous webs for engineering some of the more effective

piezoelectric nanogenerators as discussed below.

Research finds that the electrospun nanocomposite based piezoelectric nanogenerator can generate more electrical outputs as compared to pure polymer based electrospun fibres. Electrospun nanocomposite is nothing but to use filler components with the polymer materials during an electrospinning method. Furthermore, in nanocomposite concept, it is worth mentioning that the organic polymers (PVDF, PVDF-TrFE, PLA, PP, etc.) [110–114] and inorganic piezoelectric materials (piezoceramics-PZT, KNN, BT, etc, and semiconductors-ZnO, ZnSnO₂, MoS₂, etc)

[15,17,115–118] are commonly used as a matrix and filler component, respectively. To enhance electrical outputs of the electrospun nanofibre based piezoelectric nanogenerator, already a huge number of works have been carried out by the different research groups on the electrospun nanocomposites. One such example, Koc and colleagues [102] fabricated a flexible piezoelectric nanogenerator based on PVDF/PZT electrospun nanocomposite fibrous web (Fig. 5c). The electrospun nanocomposite fibrous web was developed by PVDF polymers along with different concentrations of PZT (10%, 20% and 30% on weight of PVDF polymers). A rotating drum-based collector has been used during the electrospinning method for getting aligned nanofibers. It was found that the capacitance value is increased by increasing the PZT concentration up to 20% concentration. This is due to the higher number of interfacial interactions between the PVDF polymer chains and PZT filler. Whereas the capacitance value is decreased with the PZT concentration of 30%, due to the agglomeration of PZT fillers at higher concentration. The result is lower interaction between polymer chains and filler. Moreover, the PVDF/10% PZT based piezoelectric nanogenerator showed 6.35 μ W power which is 85% higher than power generated by a pure PVDF based nanogenerator (3.44 μ W). Similarly, in another study, a PVDF/PZT nanocomposite nanofibres based piezoelectric nanogenerator was also reported by Chamankar and their co-workers [119] (Fig. 3j). In their study, electrospun nanocomposite fibres were prepared by the combination of PVDF and various percentages of PZT nanoparticles. It was observed that output voltage is high in the case of PVDF/0.37% PZT nanoparticle based nanogenerators i.e. 184 mV under a applied force of 2.1 N, compared to 0.011% PZT incorporated PVDF polymer based nanogenerator (22 mV). This is due to the higher interfacial polarization at higher numbers of PZT nanoparticles in PVDF polymers. Later, in the year of 2020, Guan et al. [103] reported the flexible piezoelectric nanogenerator based on shape memory polyurethane (SMPU)/PZT electrospun nanocomposite fibres (Fig. 5d). The authors have observed that all aligned nanofibres showed better energy harvesting efficacy compared to randomly oriented nanofibres. The piezoelectric nanogenerator based on an oriented nanofibrous web had an output voltage of 421 mV. This is 5.4 times higher than the equivalent randomly oriented fibres-based piezoelectric nanogenerator (65.6 mV). The reason is that aligned nanofibres have lower surface area and higher density than the randomly oriented fibres. In addition, aligned fibre results in compactness and lower porosity which also influences the output voltage of the nanogenerator. Using PZT (Pb (Zn_{1/3}Nb_{2/3})O₃-Pb (Zr_{0.5}Ti_{0.5})O₃) fillers in the P(VDF-TrFE) polymer during electrospinning, Liu et al. [104] also fabricated a piezoelectric nanogenerator (Fig. 5e). It was found that the developed energy harvester can generate an output voltage and an output current of 3.4 V and 240 nA with 20% nanoparticle concentration.

Other than PZT based fillers, researchers have also tried to develop lead-free, environment friendly electrospun nanocomposite fibres based piezoelectric materials by using other lead-free fillers. For example, a study [120] revealed the development of a non-complex and exclusive lightweight energy harvesting device comprising of poly (vinylidene fluoride) (PVDF)/potassium sodium niobate (KNN) electrospun nanocomposite. Different factors were established to fabricate and modify KNN as nanostructured substances comprising a high aspect ratio to enhance an exclusive link with potassium sodium niobate. These structures containing potassium sodium niobate function as a β -nucleating agent in the matrix, which improves the beta crystal phase resulting in a web of fibre. This web like morphology increases the ability to extract piezoelectricity compared with a non-treated PVDF web. Such in-situ oriented nanostructures of KNN offering a load of 5% inside the fibrous nanocomposite successfully provides high piezoelectric output. X-ray diffraction and Fourier transform infrared studies confirmed the mixture of the α - and β -crystal phase of pure poly (vinylidene fluoride) that can be converted in the beta state as soon as KNN nanostructures are placed in a web of fibre. The PVDF/KNN supported device showed a peak voltage of 1.9 V whereas an output voltage range

from 50 mV to 100 mV was obtained from the pure PVDF electrospun web. The drastic enhancement in the output voltage of the PVDF/KNN nanofibrous web was related to the β -nucleation with KNN, in-situ poled β -crystals of PVDF arranged in KNN along with the axis of the fibre. So, the existence of KNN in the PVDF matrix provides the advantage to nucleate and the rise of crystal spots into the linear matrix of polymer which improved nucleation to give rise to the beta phase. The tests related to energy harvesting revealed that the composite nanogenerators offer a voltage of 1.9 V that was comparatively forty times higher than raw PVDF-based devices. Thus, light weight (PVDF)/potassium sodium niobate (KNN)-supported nanogenerators can be effectively used in wearables devices [120]. The same group also fabricated a PVDF/KNN nanorod electrospun nanofibre-based piezoelectric energy harvester (Fig. 5f) [105]. Here, they used PVDF as a polymer component and KNN as a filler wherein the KNN nanorod was synthesized by the hydrothermal synthesis method having an aspect ratio of \sim 8.5. Different contents of KNN nanorod with various percentages were incorporated in PVDF polymer by the electrospinning method. The developed PVDF/KNN electrospun nanocomposites were characterized by FTIR, XRD (for structural analysis) and SEM, TEM (for morphology analysis). Finally, a piezoelectric nanogenerator was fabricated by using PVDF/KNN electrospun nanocomposite followed by piezoelectric characteristics which were evaluated in the form of output voltage, current, and power density. Interestingly, it was found that the β -fraction value (calculated by FTIR spectra) was gradually decreased (97–78%) with an increase in the percentage of KNN in PVDF polymer. It might be due to the higher number of β -spherulite getting merged and shifted to α -crystalline phase. However, PVDF/KNN based piezoelectric nanogenerator showed higher piezoelectric output signals than the pure PVDF-based nanogenerator. Optimized electrospun nanocomposite i.e. PVDF/3% KNN showed maximum output voltage and current of \sim 17.5 V and \sim 0.522 μ A, respectively, against repetitive finger tapping. Whereas, the pure PVDF-based nanogenerator showed only output voltage of \sim 0.5 V and current of \sim 0.089 μ A, respectively. The reason behind this could be attributed to a β -nucleating effect of KNN in PVDF polymer and in-situ poling of the KNN and PVDF polymer during electrospinning. In another study, Bairagi et al. [106] studied piezoelectric properties of the silane-modified KNN (SM-KNN) incorporated PVDF polymer based nanocomposite, which was also developed by electrospinning (Fig. 5g). Surprisingly, all the properties of PVDF/SM-KNN electrospun nanocomposite followed the same trend as PVDF/KNN based electrospun nanocomposite. Even more, piezoelectric output signals also showed similar behaviour unlike untreated KNN incorporated electrospun nanocomposite web based piezoelectric nanogenerator. However, the output signals were improved with SM-KNN incorporated PVDF based nanogenerator. The PVDF/3% SM-KNN based piezoelectric nanogenerator showed output voltage of \sim 21 V and output current of \sim 22 μ A, respectively. This improvement in output signals of PVDF/SM-KNN based nanogenerator was due to the better compatibility between the SM-KNN and PVDF polymer. This results in a higher β -nucleating effect in PVDF due to the presence of a higher number of KNN in PVDF polymer. In all three of the above studies, authors have also utilised their piezoelectric nanogenerator for glowing light-emitting diodes (LEDs), which provides evidence of self-powered applications of the electrospun nanofibre-based piezoelectric nanogenerators.

Yang et al. [107] have reported a flexible piezoelectric nanogenerator based on aluminium nitride (AlN)/PVDF-TrFE electrospun nanocomposite (Fig. 5h(i)). They observed a significant improvement in the piezoelectric performance of PVDF-TrFE polymer having very tiny AlN doping. The electrospun nanocomposite is composed of 15% PVDF-TrFE polymer with 0–0.2 wt% of AlN. After developing all the electrospun nanocomposites, the piezoelectric characteristics of the device made by as-prepared PVDF-TrFE/AlN nanocomposite were evaluated against applied force in the perpendicular and transverse directions. In both cases, the PVDF-TrFE/0.1% AlN nanocomposite-based piezoelectric device exhibited optimum results. When the pressure was

applied in the perpendicular direction, the piezoelectric device exhibited an output voltage of ~ 106 mV and an output current of ~ 1.1 nA, respectively, at 5 Hz frequency and 4 N force. On the other hand, a piezoelectric device having the composition of PVDF-TrFE/0.1% AlN-based piezoelectric device had an output voltage of ~ 82.1 V when the pressure was applied in the transverse direction. This is due to the higher deformation of the PVDF/AlN nanocomposite structure in the transverse direction as compared to the perpendicular direction. The authors also studied some wearable applications of their electrospun nanocomposite based piezoelectric nanogenerator. For instance, first, the developed nanogenerator was used to perceive human finger bending. In this application, the nanogenerator was attached to the joint of an index finger and the output signals were recorded during bending of the finger. It was found that the attached nanogenerator has an output signal of 0.9–1.3 V with a finger bending angle in the range of 10° to 20° (Fig. 5h(ii)). In a second application, the developed nanogenerator was used as a pulse sensor. For pulse sensing, the nanogenerator was attached to the wrist artery by an elastic band, and it was found that a signal in terms of an output voltage is generated from the nanogenerator, which is then can be captured and analysed. From this analysis, it was reported that the beat per minute (bpm) value is 73. 2.

So far, enhancing the output of piezoelectric nanogenerators by means of different techniques has continued to be the principal inspiration for effective self-powered devices. However, stability and durability of piezoelectric nanogenerators also need to be considered important factors in order to maintain long-lasting self-powered devices [121–125]. In 2016, Siddiqui and colleagues [108] prepared a stable and durable piezoelectric nanogenerator based on the electrospun nanocomposite. Herein, barium titanate nanoparticles (BT NPs) were incorporated into poly(vinylidene fluoride-trifluoroethylene) (P(VDF-TrFE)) polymer by an electrospinning method. A nanofiber PENG (nf-PENG) was fabricated by encompassing nanofibers in an elastomeric PDMS film (Fig. 5i). The fabricated nanocomposite fibres-based PENG was placed inside the shoe for harvesting the biomechanical energy during human walking. And interestingly, it was found that the as fabricated PENG having a BT NP concentration of 15% showed an output voltage of 25 V during walking at a walking frequency of 0.6 Hz and a higher load of 600 N. Finally, the authors have explored a wearable application of their developed piezoelectric nanogenerator. In a wearable application, they highlighted the power supply to a strain sensor using their developed nanogenerator and a capacitor. A strain sensor (based on carbon nanotubes) was interfaced with the fabricated piezoelectric nanogenerator and a capacitor. By walking, energy is generated from the piezoelectric nanogenerator, and it is stored in the capacitor, and that stored energy is used to power up a strain sensor without using any external power sources. The whole fabricated strain sensing system was also attached to the wrist, and the response was recorded during hand movement. Similarly, in the year 2022, Athira and colleagues [109] investigated a flexible BaTiO₃/PVDF electrospun nanocomposite fibres based piezoelectric nanogenerator (Fig. 5j(i)). It was found that the electroactive β crystalline phase in PVDF polymer was improved with the incorporation of BT nanoparticles. The possible reason is the interfacial interaction between BT nanoparticles and PVDF polymer matrix during electrospinning process. The fabricated piezoelectric nanogenerator had an average open-circuit output voltage and short-circuit current density of ~ 50 V and 0.312 mA/m², respectively. The nanogenerator also showed an output power density of ~ 4.07 mW/m², which is 10 times larger than the pristine PVDF polymer based piezoelectric nanogenerator. The authors also have demonstrated the self-powered vibration sensing applications of the developed nanogenerator. Where, the piezoelectric nanogenerator was used for mapping of the real time mechanical vibrations of an electric sewing machine, a faulty CPU fan and a hard disk drive (HDD) (Fig. 5j(ii)). It was found that the piezoelectric nanogenerator had an output signal of 400 mV for a faulty CPU fan which is enough to distinguish it from the normal CPU fan. Similarly, Zhao et al. fabricated carbon-based

polyacrylonitrile/barium titanate (PAN-C/BTO) nanofibrous webs (using electrospinning) that can sense various types of functions. These sensors exhibit piezoresistive, piezoelectric, and triboelectric effects, which play a key role in sensing. The flexibility of the polyacrylonitrile/barium titanate nanofibrous web during bending has been measured and this type of nanofibres can sense up to 1.12 deg⁻¹ from 58.9° to 120.2° and at a working range of 28° – 150° C. A factor obtained by measuring the gauge lies in the range of 0.15–25 N. The resulting voltage and short circuit current of nanofibre-containing devices at different pressures can then be obtained. It offered an output voltage and short circuit current of 12 V/0.08 μ A, 15 V/0.11 μ A, 32 V/0.32 μ A, and 49 V/0.48 μ A under 0.15, 1.5, 15, and 25 N impulse force, respectively. In comparison with the polyacrylonitrile/barium titanate device, it showed that polyacrylonitrile-based devices offered less electrical output at similar pressure conditions. The output voltage was found in the range of 1, 2, 12, and 15 V, whereas the short circuit current was 6, 13, 120, and 205 nA at 0.15, 1.5, 15, and 25 N impulse forces, respectively. It was found to be stable for a longer period even at 60000 cycles. The incorporation of barium titanate nanoparticles (BTO NPs) in the nanofibrous web caused 2.4 times improvement in its pressure sensing ability by the effect of both piezo and triboelectric effect. A wide range of applications of such devices includes human sensations during the swallow, walking, flexibility, and tapping of fingers in wearable sensing devices [126].

Likewise, Garain and co-workers [127] reported an electrospun nanocomposite fibrous web based piezoelectric nanogenerator. The nanogenerator mainly consists of Ce³⁺ doped PVDF/graphene electrospun nanofibers as shown in Fig. 6a(i). It was found that the Ce³⁺ doped PVDF/graphene based nanogenerator had an average output voltage of 11 V and with a current density of ~ 6 nA/cm² (Fig. 6a(ii)) at a constant load of 8 N and at 8 Hz frequency, respectively. Whereas only Ce³⁺ doped PVDF based nanogenerator had an electrical output voltage of 4.5 V with the same applied load and frequency. This is due to the inherent crystal structure and delocalized π -electrons of the graphene which enhances its electrical characteristics. The developed nanogenerator can glow ten blue light-emitting diodes (LEDs) upon spontaneous application of mechanical stress. Additionally, such nanogenerators were integrated with musical vibration to prove the random vibration energy harvesting capability of this nanogenerator. For instance, in the case of the sound-driven power generation, when an applied sound of ~ 88 dB (intensity) was employed to the ANG, it generated an AC voltage of approximately 3 V (Fig. 6a(iii)). This generated sound wave was applied to cause a vibration of the lightweight ANG which offers an electric potential from the PVDF-Ce-G nanofibres (NFs). Such sensors obtained from nano-pressure allow exceptional piezoelectric features to carry out ultrahigh sensitivity for measuring pressure (at very low values like 2 pascals). Therefore, Garain et al. demonstrated a nano-pressure sensor capable of detecting small outer impact/pressure inclination due to lighter weight objects (such as thermocol and the flow of wind). So, it can be concluded that ANGs can be utilised over a wide range of devices and for harvesting energy.

Some of the researchers have also explored different semiconducting fillers (ZnS, ZnO, etc.) rather other inorganic piezoelectric fillers for fabricating the electrospun nanocomposite based piezoelectric nanogenerators. For example, Sultan and co-workers [129] reported a piezoelectric nanogenerator composed of zinc sulphide (ZnS) nanorods/PVDF electrospun nanocomposite fibres. Such nanogenerators offer a huge amount of sound perception with a detectable range of $\sim 86 \pm 3$ Hz. The open-circuit voltage as a function of sound frequency under uniform sound pressure level (SPL) was observed along with its presentation in the fast Fourier Transform (FFT) process. These devices offer a good sensitivity to sound pressure levels ranging from 50 to 100 decibels and they can also be utilised for the detection of noise. This is essentially a powerful electro-mechanical coupling which allows them to automatically start a microphone and begin recording sound. The energy conversion with the wind provides efficiency of about 58%

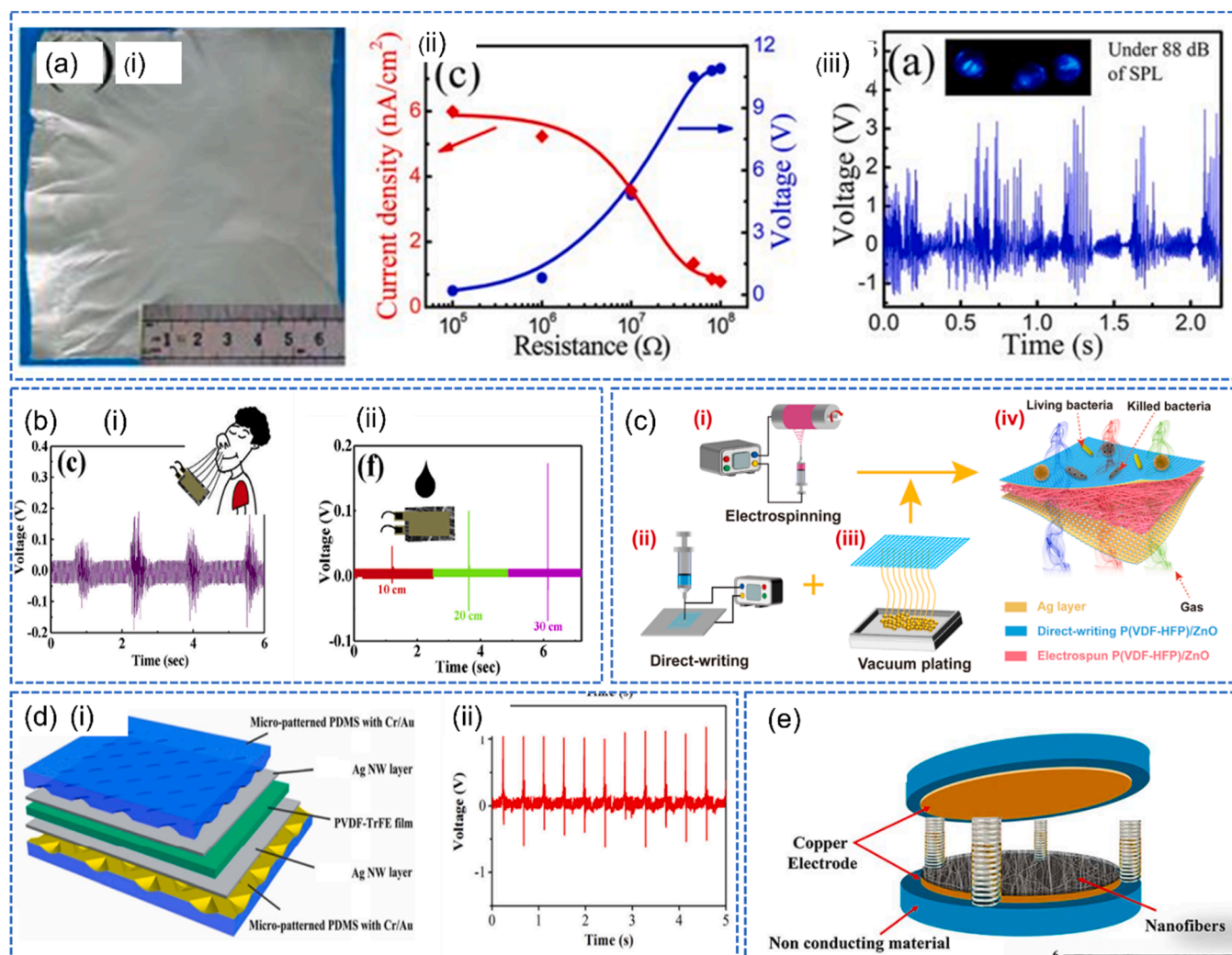


Fig. 6. a. (i) Ce^{3+} doped PVDF/graphene electrospun fibres, (ii) output voltages and current density of the Ce^{3+} doped PVDF/graphene electrospun fibres based PENG, (iii) output voltage generated by Ce^{3+} doped PVDF/graphene electrospun fibres based PENG under sound vibrations. b. (i) output voltage generated by PVDF/zinc sulphide nanorod electrospun nanofibres based PENG against human respiration, (ii) an output voltage generated by PVDF/zinc sulphide nanorod electrospun nanofibre-based PENG against water drop. c. The schematic diagram of the preparation of P(VDF-HFP)/ZnO electrospun nanocomposite fibre based PENG. d. (i) Polydimethylsiloxane (PDMS)/silver nanowire (Ag NW)/poly(vinylidene fluoride-trifluoroethylene) [P(VDF-TrFE)] based PENG [128], and (ii) an output voltage generated by polydimethylsiloxane (PDMS)/silver nanowire (Ag NW)/poly(vinylidene fluoride-trifluoroethylene) [P(VDF-TrFE)] based PENG. e. Schematic diagram of a setup for testing piezoelectric performance of the PANI/HNT/PVDF electrospun nanofibres based PENG.

(a) Reprinted with permission from ref. [127]. Copyright 2016 American Chemical Society. (b) Reprinted with permission from ref. [129]. Copyright 2019 Elsevier. (c) Reprinted with permission from ref. [130]. Copyright 2023 Springer Nature. (d) Reprinted with permission from ref. [128]. Copyright 2015 AIP Publishing LLC. (e) Reprinted with permission from ref. [131]. Copyright 2018 John Wiley and Sons.

which can detect respiration in humans. Fig. 6b(i) demonstrates the nanogenerators which are actuated by exhalation of humans when kept at a distance of 10 cm from the nose. Furthermore, to extract energy from raindrops, such nanogenerators can also be used. Pressurised water droplets can generate output voltage accompanying the nanogenerator. It can also react to one drop of water struck at different altitudes (Fig. 6b(ii)), establishing its capability to segregate far output pressure effects. Therefore, this study displays a multifunctional inorganic-organic mixed nanogenerator towards mechanical energy harvesting from different vibrations from the environment and self-powered microphone application. In particular, due to the flexibility, ease of preparation, such multi-functional sensors can be used in many fields such as in the detection of different forms of energy including sound, rain, wind and biomechanical signals. The authors have also explored some applications of the developed piezoelectric nanogenerator. They reported that the nanogenerator can be used as a self-powered microphone. The commercially available microphone is nothing but a device that can

convert sound energy into electrical signal; hence an advanced human machine interfacing system can also be developed using the flexible piezoelectric nanogenerator. It was found that the flexible piezoelectric nanogenerator based microphone can generate an electrical signal with different output profiles when different words (alpha, beta, gamma, one, two, three, four five, zinc sulfide, and microphone) are pronounced in front of that nanogenerator. Therefore, the flexible piezoelectric nanogenerator can be used as a self-powered microphone for sound recording. Likewise, in 2023, Fan et al. [130] developed an ultra-thin piezoelectric nanogenerator having breathability, superhydrophobicity and antibacterial properties. The piezoelectric nanogenerator is composed of a P(VDF-HFP)/ZnO electrospun nanocomposite membrane (Fig. 6c). The piezoelectric output voltage was found to be 9.22 V for the ZnO (15 wt%)/P(VDF-HFP) based nanogenerator, whereas the pristine P(VDF-HFP) nanogenerator showed an output voltage of 5.41 V, under a constant load of 20 N and at 2 Hz frequency. This is because of the inherent piezoelectric characteristic of the ZnO fillers. The developed

piezoelectric nanogenerator was used as a pressure sensor to monitor motion and health in the human body. It was found that the nanogenerator can detect significant motion of the arm, throat, and wrist. It can also sense the pulse beat from muscle movement in terms of an output voltage i.e 13 mV. Similarly, in a study, Zhang et al. [132] reported an effective PVDF/BiCl₃/ZnO electrospun nanocomposite fibres based piezoelectric nanogenerator. And they found that the nanogenerator based on PVDF/BiCl₃/ZnO showed higher output voltage and current (12 V and 80 nA) as compared to its counter parts (pure PVDF, PVDF/ BiCl₃, PVDF/ZnO). This is due to the p-n junction of BiCl₃ and ZnO. Finally, the developed nanogenerator was attached to an elbow or knee to detect movement of the human arm and leg. Interestingly, it was found that the piezoelectric nanogenerator has an output voltage of 1.2–3 V with arm bending from 40° to 80°. In another study, Bairagi et al. [133] also reported the effect of additional semiconductive (ZnO) fillers along with KNN in PVDF polymer. In this context, ZnO nanorods were synthesized by the hydrothermal synthesis method and incorporated in PVDF polymer along with KNN nanorods. Since it is well known that ZnO is piezoelectric, it is expected that ZnO/KNN incorporated PVDF electrospun nanocomposite webs would exhibit higher piezoelectric signals as compared to KNN-incorporated PVDF polymer-based nanocomposite. Remarkably, it was found that the ZnO/KNN-incorporated PVDF polymer-based piezoelectric nanogenerator showed the maximum output signals (output voltage ~ 25 V, and output current ~1.81 μA) compared to the KNN/PVDF based nanogenerator (as mentioned above). ZnO nanorods are semiconductive having a piezoelectric property which results in higher energy harvesting efficiency. This is due to interfacial polarization between filler and PVDF polymer matrix in the presence of ZnO filler. Even though the third component, specifically ZnO incorporated nanocomposite nanofibre based piezoelectric energy harvester has shown significantly higher piezoelectric properties, there are still some drawbacks in terms of the energy scavenging ability of the device. When pressure is applied, charge separation takes place inside the materials, followed by charges coming to the electrode materials. Therefore, it may happen that all the generated charges are not able to come into the electrode materials.

In this context, researchers have developed electrospun nanofibre-based piezoelectric materials with at least one conductive filler layer composed of piezoelectric polymers. The conductive fillers will provide a conductive path in the piezoelectric material, and therefore, when pressure is applied to the piezoelectric material, charges inside materials will try to flow through that path. Owing to this phenomenon, the resultant piezoelectric outputs will be enhanced for the piezoelectric nanogenerators. For example, in a study, Liu et al. [128] reported a piezoelectric nanogenerator composed of the conductive fillers (Ag NWs) incorporated polydimethylsiloxane (PDMS) polymer as depicted in Fig. 6d(i). Such nanogenerator was prepared by spin-coating a AgNW mixture on the top of a P(VDF-TrFE) piezoelectric textile. AgNW coated P(VDF-TrFE) piezoelectric textile was kept between two Cr/Au layer-sputtered micro-patterned PDMS films, which act as electrodes. The AgNWs sandwiched in the P(VDF-TrFE) piezoelectric textile and PDMS electrodes, offer a powerful transmitting layer that carries out the deposition along with the flow of the charge. The applications of the Ag NWs and micro-patterned PDMS film electrodes offer a significant output power of the resultant nanogenerators. The fabricated nanogenerator offers an open voltage and peak short current of 1.2 V and 82 nA, respectively (Fig. 6d(i)). In addition, to check the effect of AgNW fillers on the piezoelectricity, a controlled device was prepared by using a P(VDF-TrFE) nanofiber mat and flat PDMS layers along with Cr/Au layers as electrodes. And it was found that the piezoelectric device containing AgNWs showed an output short circuit current of 68 nA, while the controlled device generates an output current of 33 nA under the same force and frequency. From these results, it can be concluded that AgNW filler has an immense effect in piezoelectricity of the resultant piezoelectric materials. This is due to a conductive path in the PDMS polymer, provided by the Ag NWs fillers. These results support the developed

piezoelectric nanogenerators can be integrated with small scale electronic devices to make them self-powered electronic devices. These generators quantify the externally compressed signals at fewer frequencies as in the case of motion caused by humans (walking). Such demonstrations radiate the energy that can potentially be harvested from these nanogenerators [128]. The same concept was also explored in another study by Bairagi et al. [70] where CNTs were used as an additional filler with KNN nanorods in PVDF polymer. A KNN/CNT incorporated PVDF polymer-based nanocomposite material was developed by the electrospinning method. This piezoelectric material-based nanogenerator showed better piezoelectric properties (output voltage of ~23.24 V and current of ~ 52.29 μA, respectively) as compared to only KNN-incorporated PVDF polymer electrospun based nanogenerator. This may be due to the interfacial polarization and conductive path in the presence of CNTs in the matrix of PVDF. An approach to fabricating a nanogenerator containing PANI (polyaniline)/HNT (halloysite nanotube)/PVDF (poly(vinylidene fluoride)) using electrospinning has also been developed (Fig. 6e) [131]. The materials (PANI and HNT) act as an agent for nucleation and conduction in PVDF polymer and therefore, the overall piezoelectric property of the PVDF polymer is enhanced. Under different outer stresses caused mechanically (like by pressure, pattering, and impact) the device showed a voltage of 7.2 V. The nanogenerator adhered to the arms of humans by providing brilliant pressure electric output at the time of arm movement as well as high sensitivity, flexibility, and finally durability. Thus, PANI-supported devices could potentially cover a wide range of applications in the areas of piezoelectricity to develop self-powered electronic devices [131].

Piezoelectric performances along with applications of the different electrospun nanofibres based piezoelectric nanogenerators are summarized in Table 5.

Despite all this, piezoelectric energy harvesters based on electrospun nanofibres or nanocomposite fibres are still not optimized properly. More research is required to optimize this piezoelectric energy harvester. Moreover, electrospun nanofibre-based PENGs have some drawbacks too like lower durability and poor washing ability specifically in the field of wearable electronic applications. In addition, the electrospinning method is a costly and complex method compared to other methods.

6.1.2. Melt spun fibre-based PENGs

Melt spinning is a process of manufacturing manmade fibre/filament using synthetic polymeric materials. This process is mainly used for different synthetic polymers, such as PVDF, polyester, nylon, etc. In a melt spinning line, there are five main sections, namely feeding, melting and mixing, spinning, cooling and winding zone as depicted in Fig. 7a [151,152]. Melt spinning is one of the most promising methods to prepare piezoelectric melt spun filaments with a high production rate and low cost [25]. However, so far, a very limited number of works have been reported on the melt spun filament/fibre based piezoelectric nanogenerators by the different research groups. Moreover, mainly PVDF polymer has been utilized to prepare the piezoelectric nanogenerator through melt spinning [153]. One such example, Hadimani et al., in 2013 [154] reported the fabrication of continuous PVDF melt spun piezoelectric fibres, for the first time. The piezoelectric polyvinylidene fluoride (PVDF) fibres were manufactured by continuous melt extrusion and inline poling (Fig. 7b(i)). The extent of polarisation is related to the pressure electric performance. For causing polarisation, polyvinylidene fluoride dipoles need to be oriented in another form by applying the powerful field of electricity accompanying higher temperature of 80 °C. The domains are locked in the polarized state by lowering the temperature in the presence of the electric field. The poling of PVDF fibres was carried out at an extension ratio of 4:1, at 80 °C and a high voltage of the order of 13000 V on a 0.5 mm diameter fibre in a melt extruder. The entire process of making PVDF fibres from granules and poling to make piezoelectric fibres was carried out in a continuous process using a melt extruder. Fibres formed from pressure electricity

Table 5

Piezoelectric data of different reported piezoelectric nanogenerators composed of electrospun nanofibrous webs.

Materials	Dimens. (cm×cm)	F (N)	Fre. (Hz)	Voc (V)	Isc (μA)	J (μW/cm ²)	Applications	Ref.
PVDF NFES	-	-	7	0.076	0.039	-	Energy harvesting	[134]
P(VDF-TrFE) NFES	5 × 2	-	-	2.2	-	-	Wind energy harvesting	[135]
PVDF-Ce-G	-	8	4	11	0.072	0.56	Pressure sensing and energy harvesting	[127]
PVDF-Er ³⁺ /Fe ³⁺	2.4 × 2	Hand tapping	-	115	32	76.8	LEDs glowing and capacitor charging	[136]
PVDF-TrFE-BT NPs	0.5 × 0.5	-	-	12.46	3.65	5.05	Biomechanical energy harvesting and capacitor charging	[137]
PDMS/Ag NW/PVDF-TrFE	-	2000	2	1.2	0.082	0.0082	Biomechanical energy harvesting	[128]
PAN-C/BTO	2 × 2	25	-	12	0.08	0.24	Pressure sensing, motion sensing, and energy harvesting	[138]
PVDF-TrFE	2.5 × 2.5	100	-	10.4	2.3	5.98	Energy harvesting	[101]
PVDF/ZnS	-	-	-	6	9.6	0.15	Energy harvesting and self-powered microphone	[129]
PANI/HNT/PVDF	2 × 2	300,000	-	4.5	0.75	0.25	Energy harvesting	[139]
PVDF/KNN NRs	3 × 3	20	7	17.5	0.522	2.3	Energy harvesting and LED glowing	[140]
PVDF/SM-KNN NRs	2 × 2	20	7	20	37	185	Energy harvesting and LEDs glowing	[141]
PVDF/ZnO/KNN NRs	3 × 3	40	10	3.4	0.32	0.58	Energy harvesting and motion sensing	[142]
PVDF/CNT/KNN NRs	3 × 3	20	7	23.24	9	52.29	Energy harvesting and LEDs glowing	[70]
PU/nanohydroxyapatite (nHA)/MWCNTs	2 × 2	1.73	-	1.95	1.95	0.9506	Biomechanical energy harvesting	[143]
PVDF/BT NPs	6 × 6	-	15.7	1.56	0.156	0.0202	Energy harvesting and capacitor charging	[144]
PVDF/PZT	2 × 4	5	3	62	2.28	17.1125	Biomechanical energy harvesting, LEDs glowing and capacitor charging	[145]
PVDF/cellulose	4 × 4	2	1	7.5	2.1	2.26	Energy harvesting, LEDs glowing and capacitor charging	[146]
PAN/CuO	3 × 3	-	-	5	0.172	0.215	Energy harvesting	[147]
PVDF/ZnO	2 × 2	40	-	15.3	2.1	3.6	Energy harvesting	[148]
PVDF/CsPbBr ₃	1 × 1	1.7	-	33	-	2.2	Energy harvesting, LEDs glowing and wrist watch glowing	[149]
PVDF/KNN-ZS	3 × 3	20	7	1.68	0.216	0.09075	Energy harvesting and LEDs glowing	[150]

were analysed at an impact force (1.02 kg) released from an altitude of 5 cm which gives rise to 2.2 V (Fig. 7b(ii)) and compared with the industrial pressure electric PVDF devices. The difference in mechanical features can be judged by observing scanning electron microscopy (SEM) images which display non-uniform structures of the poled and unpoled PVDF fibres. The greater and lower amounts of beta and alpha phases of such poling-induced fibres can be analysed by FTIR microscopy. The reverse happens in the case of non-poled fibres. The output voltage generated confirmed the existence of the beta phase in the poling-induced polyvinylidene fluoride fibre. It can effectively be used to harvest energy from wind, rain waves, and tide. Although the preparation of pristine PVDF polymer based piezoelectric filaments is comparatively easy, they will have very low piezoelectric responses. To enhance the piezoelectric performance of the melt spun PVDF filaments, nano filler components are mainly mixed with the PVDF polymers. For example, in 2019, Bairagi et al. [19] prepared a melt spun nanocomposite piezoelectric filament based on PVDF/KNN as depicted in Fig. 7c. The PVDF/KNN composite filaments have been prepared by twin-screw melt mixing and subsequently melt-spinning process with various percentages of KNN nanorods (0%, 2%, 4%, and 6% on the weight of polymer). Here, KNN is used as filler material in PVDF polymer matrix since KNN has higher piezoelectric properties than other lead-free filler materials. The resultant voltage of such a device is measured by finger patterning using a digital oscilloscope. This result in the establishment of a potential gradient throughout the prepared nanogenerators which can measure output voltage. The nanogenerator containing PVDF-KNN at a proportion of 4% nanorods resulted in a signal of 3.7 V and 0.326 μA current; whereas the other proportions as mentioned above nanorods produced voltages of 1.9, 2.7, 2.6 V. Bairagi et al. therefore, established that the nanogenerator made of PVDF/4% KNN NRs based filament exhibited the highest F (β) value (i.e., 26% polar β-phase). The dielectric constant for the PVDF/4%KNN NRs filament-based nanogenerator was 175, which covered the highest value as compared to other nanogenerators (73 for P-PVDF, 134 for PVDF/2% KNN NRs, and 131 for PVDF/6%KNN NRs filament-based nanogenerators). In this study it was found that PVDF-KNN nanorods having

a KNN proportion of 4% are the most favourable combination to prepare nanogenerators by the melt spinning process. This nanocomposite filament can be used in portable and wearable electronics containing textiles that require a minimum quantity of power to operate the device [19]. Although ceramic filler incorporated PVDF polymer nanocomposite filament-based PENG has shown better piezoelectric performance compared to only PVDF polymer filament-based PENG, there is still not sufficient output to supply power to different small scale electronic devices. Lately, in 2020, Chowdhury and co-workers [155] have also investigated the piezoelectric properties of PVDF/PU melt extruded piezoelectric materials (Fig. 7d). To prepare PVDF/PU-rGO melt extruded filaments, first PU, PVDF, and r-GO were mixed in acetone followed by vacuum drying was carried out to evaporate acetone and therefore, r-GO coated PU and PVDF were collected for the next melt mixing process in twin-screw extruder. In their study, r-GO was used as a compatibilizer to make compatibility between the PVDF and PU polymers. Pristine PVDF, shape memory PU, PVDF/PU-0.1%r-GO, PVDF/PU-0.3%r-GO, and PVDF/PU-0.5%rGO samples were prepared by the twin-screw melt mixing equipment. Thereafter, as produced filaments were cut into several small pieces and were sandwiched between the top and bottom aluminium electrodes, to evaluate piezoelectric performance. It was found that, at 0.3% r-GO concentration, the PVDF/PU melt extruded filament based piezoelectric nanogenerator showed better piezoelectric performance compared to its counter parts. PVDF/PU/0.3% r-GO based piezoelectric nanogenerator showed an output voltage and current of 349 mV and 0.294 μA, respectively, under a constant load of 3.82 N and at a 2.5 Hz frequency. This is because, 0.3%r-GO is the threshold concentration where a preferable conformation takes place for the superior piezoelectric performance. On the other hand, it was found that the piezoelectric performance is decreased with increase the r-GO concentrations because an agglomeration takes place at higher concentration.

Generally, to perform piezoelectric testing of the melt spun fibres, the fibres need to be cut into small pieces and placed side by side on the bottom electrode and then a top electrode is placed on the fibres. Then copper wires are connected with both electrodes to perform

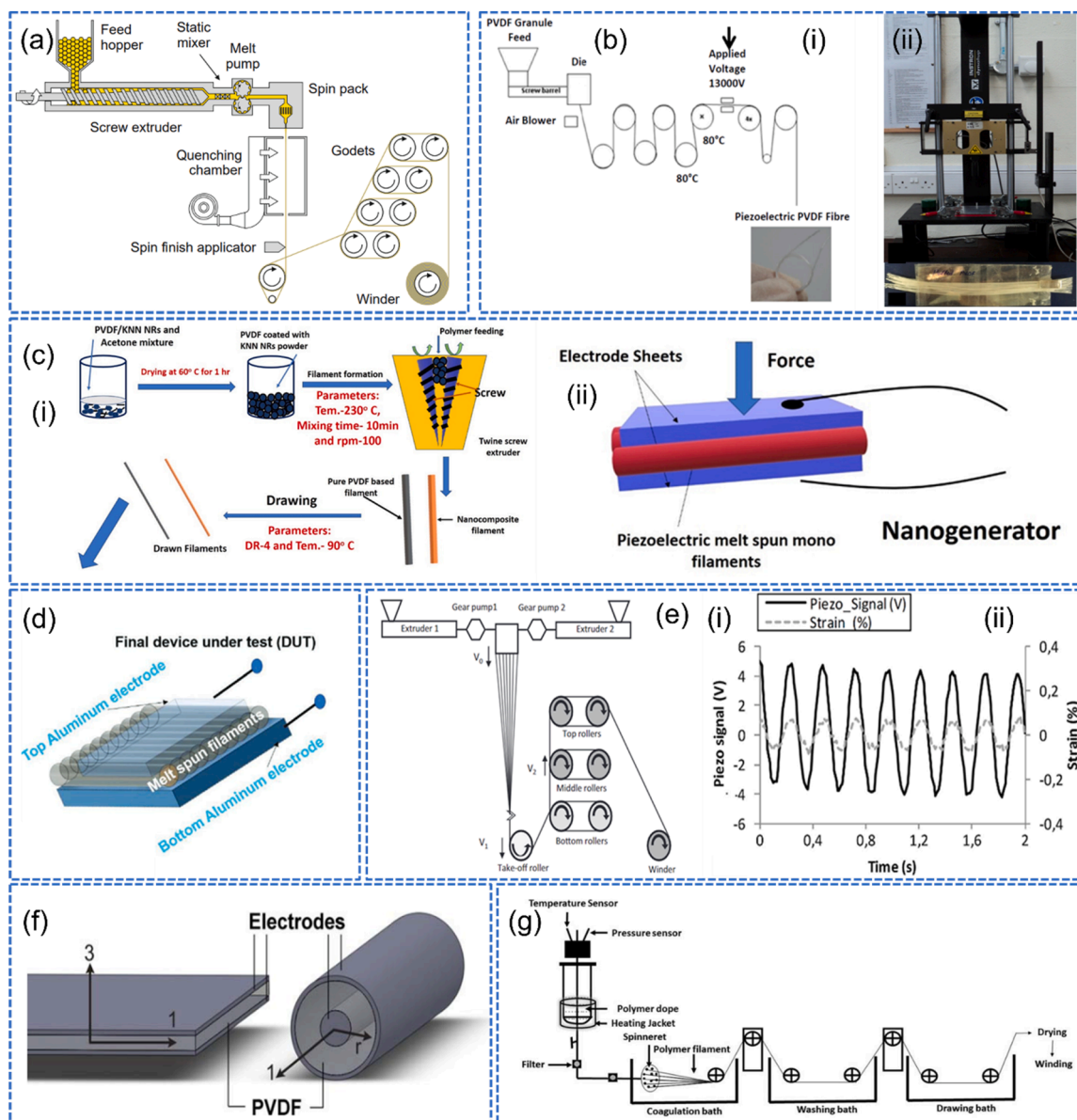


Fig. 7. a. Schematic diagram of the melt spinning method [152]. Open Access; b. (i) PVDF melt spun filament fabrication, (ii) an arrangement for applying load on the PVDF melt spun filament based PENG. c. (i) PVDF/KNN melt spun filament fabrication, (ii) fabrication of PVDF/KNN melt spun filament-based PENG. d. The piezoelectric nanogenerator based on the PVDF/PU/rGO melt extruded filaments. e. (i) PVDF/HDPE-CB core sheath melt spun filament development, (ii) an output voltage generated by PVDF/HDPE-CB core sheath melt spun filament-based PENG. f. The parallel and coaxial PVDF filament produced by a coextrusion method. g. Schematic diagram of the solution spinning method [152]. Open access. (b) Reprinted with permission from ref. [156]. Copyright 2013 Institute of Physics Science. (c) Reprinted with permission from ref. [19] Copyright 2019 Elsevier. (d) Reprinted with permission from ref. [155]. Copyright 2020 John Wiley and Sons. (e) Reprinted with permission from ref. [157]. Copyright 2013 Elsevier. (f) Reprinted with permission from ref. [158]. Copyright 2020 John Wiley and Sons.

piezoelectric test. But this is one of the most difficult methods. Moreover, nano filler loading in a polymer matrix through melt spinning is also a challenging task as in most cases, agglomeration tendency of high surface energy nano fillers inside the fibrous matrix is unavoidable.

To resolve the problems associated with placement of electrodes on filament array, researchers have tried to prepare bi-component melt spun fibre based piezoelectric nanogenerator where core component should be conductive polymer, which will act as an inner electrode, and sheath component is piezoelectric polymer [159]. One such example, Nilsson et al. [157] developed a piezoelectric nanogenerator using polyvinylidene fluoride (PVDF) and high-density polyethylene (HDPE)/carbon black (CB) based conductive filament prepared by the melt spinning process (Fig. 7e(i)). The developed nanogenerator was used to monitor human health by attaching it to the human chest [160]. The

developed nanogenerator was integrated into a belt [161] and also attached to the finger by a velcro [162] for health monitoring. The fibres were prepared by melt spinning from Extrusion Systems Limited (ESL, Leeds, England) contained with 2 screw extruders (the first for the outer part and the other for the inner part, respectively). Their temperature setups were as follows: 190 °C, 220 °C, and 230 °C for the first, second and third parts, respectively. The temperature of gear pumps and spinneret was set as 230 °C together with the spinneret containing 24 pores of 0.6 mm diameter and a length of 1.2 mm. Polyvinylidene fluoride fibres were prepared by melt spinning using two processes named contact and non-contact poling. Contact poling offers the best pressure electric output. For avoiding failure, the field generated by such fibres must be incorporated at a temperature greater than 60 °C. Permanent polarization was obtained at a duration of two seconds by

non-contact-based corona poling. The average power of about 15 nW was obtained from 25-millimetre fibre length (Fig. 7e(ii)). Therefore, to establish sensing characteristics the prepared yarns were converted into fabric by weaving which had the potential of human heartbeat detection [157]. Similarly, in another study, Martins and colleagues [158] reported a coaxial melt extruded filament based piezoelectric nanogenerator (Fig. 7f). A three-layers piezoelectric monofilament composed of a PVDF layer surrounded by two conductive layers in a coaxial arrangement.

Although melt spinning method is the most cost-effective way to prepare synthetic fibre but it was noticed that the melt spinning method for developing piezoelectric fibre is not an attractive solution (if we compare with especially electrospinning method) because, for melt filaments, extra poling arrangements must be made, which reduces cost effectiveness and the entire procedures become more electrically hazardous. Thus, the manufacturing unit requires additional expertise to develop a melt-spinning system integrated with high voltage poling accessories. Else, the already produced filaments require additional poling with high temperature separately. But poling the filament strand along with drawing before its solidification (during spinning itself) should bestow the best results.

6.1.3. Solution spun fibre based PENGs

The solution spinning method (Fig. 7g) is better than the melt spinning method, for developing piezoelectric fibres [151]. This is because, in solution spinning there is a chance of crystal phase transformation of the polymeric material, if a polar solvent is used to prepare the polymer solution. Moreover, in solution spinning there is no limitation on the processing temperature, unlike the melt spinning method. However, there is no single research work reported, where solution spun fibres were explored for developing piezoelectric devices. Only a few research works were carried out on the development of solution spun PVDF fibres but those were not explored for piezoelectric studies. One such example, Tascan and colleagues [163] studied the effects of process parameters on the properties of PVDF based solution spun fibres. It was found that the drawing temperature and drawing ration have immense effect on the tensile properties (strength and elongation) of the PVDF fibres. It was also observed that the slope of tensile strength vs drawing ratio and elongation vs drawing ratio were increased when drawing is carried out in the second drawing zone. This increasing trend was not found when drawing was performed in the first drawing zone. Hence, it can be summarized that the process parameters of solution spinning have a significant effect on the PVDF fibre properties. Later, Tascan [164] has also studied the optimization of wet spun polyvinylidene fluoride fibre through a statistical analysis using Taguchi method. In Taguchi method, breaking force, tensile strength, and breaking elongation were considered as different factors to optimize the fibres structure.

From these studies, it can be summarized that the piezoelectric fibre prepared by the solution spinning method can find promising applications in near future as the structure of the developed fibre can easily be tuned as per the needs by varying the process parameters in a systematic way.

Although electrospun nanofibre-based piezoelectric nanogenerators have shown significant piezoelectric performance, this nanogenerator is not suitable as a wearable device due to its lower durability and impaired mechanical properties compared to melt spun, and solution spun fibres. As melt spun, and solution spun fibres possess better mechanical properties which are the prime requirements for long term use as wearable energy harvesters, are mostly desirable options. However, being a 1D materials, their piezoelectric performances are not sufficient for real time applications. Therefore, for wearable energy harvesters, textile structures (woven, knitted, and braided) should be a good alternative. Textile structure based piezoelectric nanogenerators are discussed in detail in the next section.

6.2. Fabric based piezoelectric nanogenerators (PENGs)

Textile materials which can sense and return to their original phase in response to varying environmental changes are called smart textiles [165,166]. Smart textiles could potentially take on a significant function regarding waste energy harvesting by using different environmental and biomechanical energy sources. The harvesting of waste energy and its utilization in the electronics sector could potentially solve the problem of reliance on batteries (used in the various portable and wearable electronics as a powered source) [167–170]. Among the different forms of energy, wearable-based energy harvesters may be the most promising. It is the most achievable and harmonious form of energy to supply power in the different portable electronic goods, biomedical applications, and so forth. In the last couple of years, research on pyroelectric [171], photovoltaic [172–175], thermoelectric [176], triboelectric [177], and piezoelectric [154,178,179] materials and their incorporation into the textile structures has proceeded at pace. In this context, the main challenge is maintaining the fabric properties like flexibility and wearability while also integrating the device functionality. To solve these problems, several different textile-based nanogenerators have been developed. There are two textile forms: fibre based which is already discussed in Section 2 and the fabric-based textiles. Among the fabric-based textiles, different textile structures such as woven fabric, knitted fabric, and braided fabric are studied. The detail research works on piezoelectric nanogenerators using fabrics are therefore discussed in the subsequent sub sections.

6.2.1. Woven fabric based piezoelectric nanogenerator (PENG)

The piezoelectric nanogenerator made from single fibre can generate very low piezoelectric outputs since it has a very limited active area, and its utility would be limited because of its 1D structure. To resolve these real problems, fabric based piezoelectric nanogenerators have been introduced and being researched extensively from the recent past. Fabric based piezoelectric nanogenerator is nothing but the combination of the multiple single piezoelectric elements combined by various fabric manufacturing techniques. One of the most promising techniques is weaving. In this technique, woven fabric is developed simple by interlacing the warp and weft yarns. Since, wearable electronics is one of the fastest growing sectors, woven fabric-based piezoelectric energy harvesters have piqued the interest of both the research community and the industry. Several research investigations have already been carried in the recent past and many more are active in this area by several research groups at the moment. For example, a woven fabric based piezoelectric nanogenerator has been reported by Lund et al. [180], where PVDF was melt-spun with conductive core filaments in warp and other conducting yarn in the weft direction of the fabric have been inserted by using a simple band weaving machine. To prepare piezoelectric fabric, a bicomponent melt-spun fibre was developed using a twin-screw extruder at a collection speed (V_c) of 5.3 m per minute and take-up speed (V_t) of 116 m per minute. The developed melt-spun yarn was characterized by using different techniques such as XRD (for structural conformation) and tensile testing. Thereafter, the piezoelectric characteristic of the developed filament was characterised by a dynamic mechanical analyser (DMA). Silver paste was used on the surface along a length of 10 and 20 mm of the filament which acted as an external electrode and another electrode was the conductive core yarn of the melt-spun bi-component fibre. Lund et al. found that the piezoelectric filament could generate an output voltage of 1 V with the minimum strain (0.5%). Finally, two types of woven fabrics (plain and twill weave) were developed using this PVDF piezoelectric melt spun bi-component filaments (Fig. 8a(i) & (ii)), where conducting yarns were used as weft and piezoelectric filaments as warp. It was found that the twill fabric based nanogenerator showed better piezoelectric outputs compared to plain fabric based nanogenerator. Finally, the piezoelectric performance of this fabric was tested under both dry and wet conditions, and it can be seen from Fig. 8a (iii) that, even after immersion of the

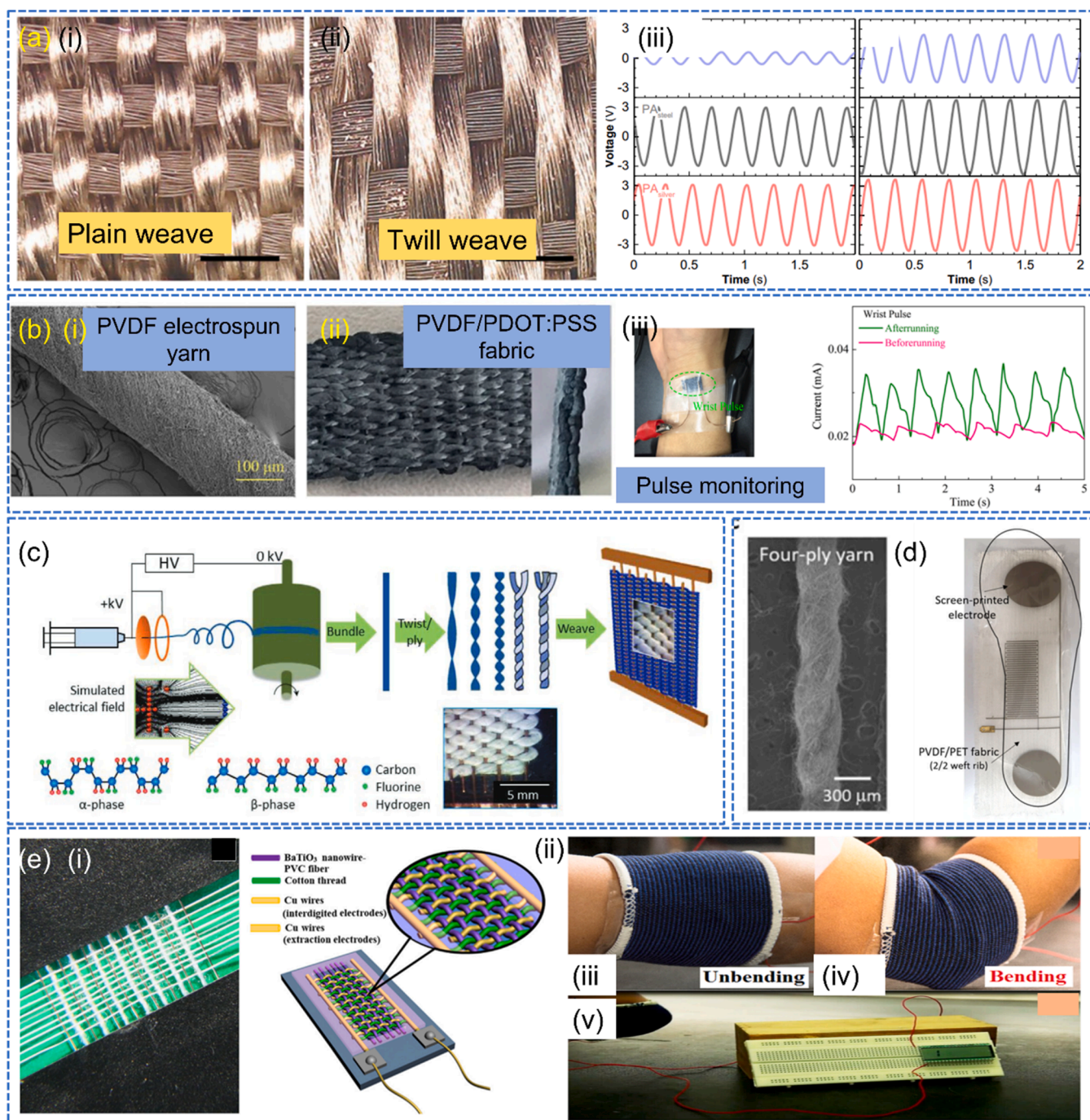


Fig. 8. a. (i) & (ii) Plain and twill weave based piezoelectric fabric made from PVDF core-sheath yarns, (iii) output voltage of the PVDF woven fabric based PENG for both plain and twill fabric based PENGs [180]. Open Access; b. (i) SEM image of a PVDF electrospun single yarn, (ii) a woven fabric based on PVDF electrospun yarns, (iii) a PEDOT-based sensor as an artificial human skin has the potential to control human motion and heart pulse [182]. Open Access; c. Schematic diagram of fabrication of a PVDF electrospun yarns based PENG. d. SEM image of a PVDF four ply electrospun yarn, (ii) image of the shoe insole pressure sensor made from the PVDF four ply yarns [183]. Open Access; e. (i) PVC-BT nanocomposite filament made piezoelectric energy harvester, (i) & (ii) energy harvesting from the developed PVC-BT based PENG by the movement of elbow, (iii) a LED screen glowing by using the developed PVC-BT based PENG. (c) Reprinted with permission from ref. [184]. Copyright 2020 John Wiley and Sons. (e) Reprinted with permission from ref. [185]. Copyright 2015 Elsevier.

fabric in the water, output voltage still existed. Finally, authors could able to lighten LED using the developed piezoelectric nanogenerator, to demonstrate a wearable application. Similarly, in another study, Rundqvist et al. [181] have also demonstrated a woven fabric-based piezoelectric sensor where PVDF melt spun bi-component piezoelectric fibre was used in the warp direction and three different conductive yarns were used in the weft direction of the woven fabric. The influence of different weaving construction (plain, twill, and weft rib) on the

piezoelectric performance of the sensors was analysed. The fabric with twill construction showed maximum output voltage (13.0 V/strain %) than plain (2.1 V/strain %). The piezoelectric performance of different sensors was examined by using the tensile testing instrument.

In recent years, a plenty of research work is in progress on the electrospun yarns based woven piezoelectric nanogenerators. This is an easy process to prepare textile yarns, by directly depositing the electrospun nanofibres on a core material. As shown in Fig. 8b, the PVDF

electrospun fibres were converted into a single yarn by applying twist and it was used to fabricate a woven fabric by a simple weaving technique. The developed fabric was used to develop a fabric-based pressure sensor for sensing heart pulse, human motion [182]. Electrospun nanofiber bundles are transferred into a single yarn (Fig. 8b(i)) followed by a coating of PEDOT on the surface and finally, a double-layer woven fabric was developed (Fig. 8b(ii)). After fabrication of this piezoelectric fabric, the output voltage and current were checked, and it was observed that the voltage-current characteristics of this fabric-based sensor varied by the variation of voltage in the range of 0–1 V. Finally, the sensitivity of the fabric-based pressure sensor was checked (18.376 kPa–1) at ~100 Pa. It was demonstrated that the use of a PEDOT-based sensor as an artificial human skin has the potential to control human motion and heart pulse (Fig. 8b(iii)). Thus, PEDOT fabric-based sensors can be utilized in wearable electronic goods. Likewise, Forouzan et al. [184] developed a piezoelectric nanogenerator based on PVDF and P(VDF-TrFE) electrospun yarns having different fineness (Fig. 8c). The as prepared woven fabric based piezoelectric nanogenerator has shown an output voltage of 2.5 V and it was observed that the piezoelectric output is improved with decreasing the fineness of the yarns and increasing of the twist, ply, and fabric density. Later, in the year 2022, Kim and co-workers [183] also reported a piezoelectric pressure sensor composed of woven fabric based piezoelectric nanogenerator (Fig. 8d). The woven fabric based piezoelectric nanogenerator was fabricated by using PVDF yarns as the weft yarns and PET yarns as the warp yarns. Two types of weave patterns namely plain and twill, were explored during the weaving process. The piezoelectric outputs of the as developed fabric based piezoelectric nanogenerator, were increased with increase the applied pressure which is quite obvious. The plain-woven fabric based nanogenerator showed an output voltage and current of 0.09 V and 7 nA with an applied load of 4 N, whereas nanogenerator had an output voltage and current of 0.62 V and 63 nA, with an applied load of 24 N, correspondingly. On the other hand, twill woven fabric based nanogenerator showed an output voltage and current of 2 V, and 109 nA respectively. Therefore, it can be seen that the twill woven fabric based nanogenerator has shown better piezoelectric output as compared to the plain fabric based nanogenerator. Finally, the authors evaluated the sensing characteristics of the developed piezoelectric sensor, and it was found that the sensitivity of the twill fabric-based sensor was 83 mV/N. They have also used the piezoelectric pressure sensor to measure pressure distribution on the human foot, in addition, a touch pad was also fabricated using the pressure sensor (Fig. 8d). Lately, Wu and co-workers [186] have also investigated the piezoelectric performance of the woven fabric based piezoelectric nanogenerator made of Cesium Lead Halide g (CsPbBr₃) perovskite decorated PVDF electrospun yarns. To prepare electrospun yarns, CsPbBr₃/PVDF nanofibres were applied directly on to the stainless-steel yarn, and thereafter the woven fabric was made by using piezoelectric yarns at the weft and bare stainless-steel at the warp direction. The developed piezoelectric fabric had an output voltage of 45 V and a current of 470 nA, with interlacing points (warp and weft) of 5 × 5, which is comparatively higher than the 1 × 1 interlacing points of the warp and weft (voltage 21.7 V and current 339 nA). Although, the fabric-based PENG made of one kind of piezoelectric fibre is relatively simple and easy to prepare, the piezoelectric performance is not at all satisfactory.

Therefore, woven fabric based PENGs made of composite or blended piezoelectric fibres is the main concern. Many studies have been conducted in this area, where composite or blended piezoelectric filaments have been used to fabricate woven fabric based piezoelectric energy harvesters. For example, Zhang et al. [185] developed a hybrid yarn-based wearable piezoelectric nanogenerator for energy harvesting using human body moment (Fig. 8e (i)). The nanogenerator was fabricated by using PVC-BaTiO₃ nanorod hybrid yarns in the warp direction, metal copper in the weft direction and cotton threads as spacer yarns in the weft direction of the woven fabric. When the nanogenerator is in bending condition, the direction of the dipoles and the applied force is

found in the same direction, which results in d_{33} (direction of applied pressure and induced response are the same) based piezoelectric constant. At that stage, a potential difference takes place between the two surfaces of the nanogenerators, and a forward signal gets generated and vice-versa. In the study, this woven nanogenerator was attached to a hand-knitted fabric to be used as an energy harvester at the elbow of a human. This nanogenerator showed the capability to generate an output voltage of 1.9 V and an output current of 24 nA. And it was found that the piezoelectric output is enough to light an LCD screen, which substantiates the ability of the developed piezoelectric nanogenerator for wearable application (Fig. 8e(ii), (iii) & (iv)). The piezoelectric device consists of composite filament only shows 1.9 V which is lower than even the pure PVDF fibre-based PENG (2.3 V). This is because PVC polymer has no piezoelectric effect like PVDF polymers. Similarly, in 2022, Kim et al. [187] have also reported a woven fabric based piezoelectric nanogenerator. The developed nanogenerator consists of melt spun PVDF/BaTiO₃ fibres having different biomimetic cross-sections (Fig. 9a). The piezoelectric outputs were found to be higher (an output voltage and current of 36.05 V and 3.126 μA) in case of daffodil flower-shaped PVDF fibre based piezoelectric nanogenerator compared to other counterparts (a radish flower, a papyrus stem, and a stalk grain stem based on PVDF fibres). The reason is obvious: the daffodil flower shaped PVDF fibre has a higher surface area compared to others. The higher surface area exhibits higher mechanical deformation during piezoelectric performance. Furthermore, the effect of BaTiO₃ morphology on the piezoelectric properties of a PVDF fibre based piezoelectric nanogenerator was also investigated. It was found that BaTiO₃ nanorods/ daffodil flower shaped PVDF fibre based nanogenerator had higher piezoelectric outputs (voltage of 62 V and a power of 91 μW) compared to BaTiO₃ nanoparticles incorporated nanogenerator. The reason is higher surface area of the BaTiO₃ nanorods compared to nanoparticles which exhibits a higher beta nucleating effect in PVDF polymer.

In other studies, researchers have also investigated the piezoelectric properties of PVDF nanocomposite fibre based piezoelectric nanogenerator composed of ZnO fillers. For instance, DEMİR et al. [194] studied the effect of ZnO nanowire concentrations on piezoelectric properties of ZnO nanowire-coated cotton fabrics. It has been found that the piezoelectric properties in terms of output voltage and current is increased with increasing concentration of ZnO nanowires. The optimized piezoelectric energy harvester has shown an output voltage of 1.12 V and an output current of 52 nA, respectively. p-n junction based ZnO nanorods with different aspect ratio, have also been used to fabricate textile based piezoelectric energy harvesters (Fig. 9b (i) & (ii)) [188]. In this work, a Cu/Ni coated polyester woven fabric has been covered by ZnO nanorods through an in-situ chemical synthesis method and thereafter, the polyester fabric covered with ZnO was coated with PDOT: PSS and cuprous thiocyanate (CuSCN). Finally, it was found that longer p-n junction based ZnO nanorods coated piezoelectric energy harvester showed higher output voltage (1.32 V) and current (0.56 μA) compared to shorter ZnO nanorod-based piezoelectric energy harvesters (0.43 V and 0.16 μA). This is due to the greater freedom of movement in longer ZnO nanorods compared to shorter ZnO nanorods at same vibration amplitude, which results in a greater strain and therefore potential difference is improved. Likewise, in another study, Zhang et al. [189] have also reported a piezoelectric nanogenerator based on PVDF/ZnO woven fabrics (Fig. 9c(i)). Here, the nanogenerator was fabricated by sandwiching ZnO nanorod arrays using two Ag coated textile fabrics (Fig. 9c(ii)). Interestingly, it was found that the developed nanogenerator had an output voltage of 4 V and an output current of 20 nA, under palm clapping, whereas values were 0.8 V and 5 nA, respectively under finger bending. In addition, the as fabricated nanogenerator was utilized to power up different microelectronics, such as glowing of LEDs, and a miniature display screen was lit using energy generated by palm clapping (Fig. 9c(iii)). Although the above three studies have shown somewhat better piezoelectric properties in the

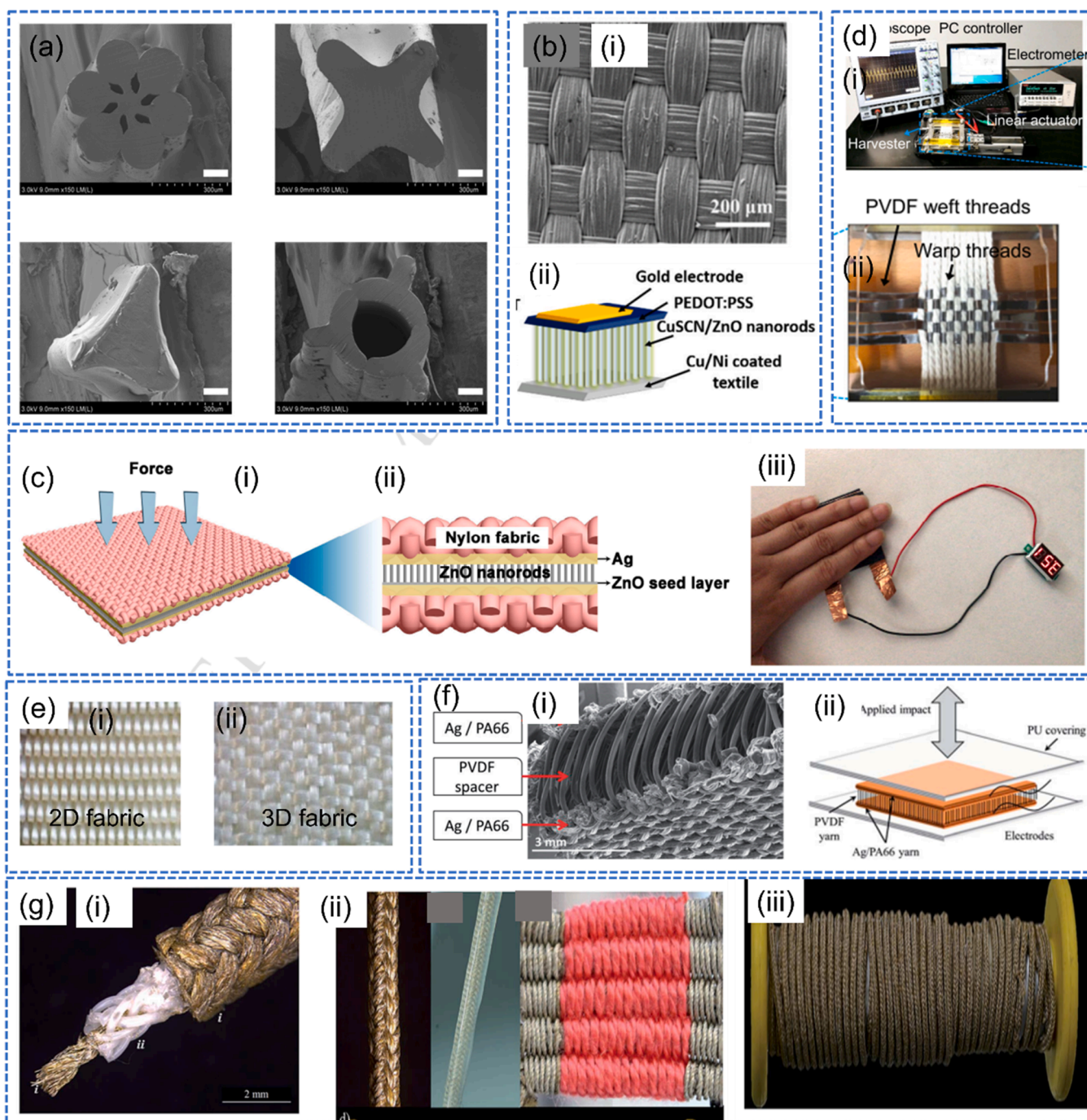


Fig. 9. a. Melt spun PVDF/BaTiO₃ fibres having different biomimetic cross-sections. b. (i) SEM image of the ZnO nanorods on Cu/Ni coated textile, (ii) the schematic diagram of a PENG based on CuSCN/ZnO coated Cu/Ni conductive textile fabrics. c. (i) The piezoelectric nanogenerator based on PVDF/ZnO woven fabrics, (ii) A schematic diagram the developed PENG, (iii) glowing of a LED screen by the developed PENG. d. (i) The setup for testing of PVDF fabric-based PENG, (ii) A digital photo of the developed woven fabric. e. (i) & (ii) 2D plain weave canvas and 3D diagonal interlock fabrics using PVDF melt-spun filament yarns. f. (i) 3D knitted spacer fabric based PENG, (ii) application of force on the piezoelectric energy harvester made of 3D knitted spacer fabric. g. (i) (ii) & (iii) braided structure-based PENG.

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presence of piezoelectric nanofillers, still output signals are not very satisfactory.

Hence, continued efforts have been put forward to work on developing wearable woven fabric based piezoelectric energy harvesters having higher piezoelectric responses. In a study, Song and co-workers

[190] reported the woven fabric based piezoelectric nanogenerator which was developed using PVDF thin film-straps. To prepare PVDF thin film-straps, first PET film with one side adhesive was pasted on the PVDF thin film sheet. Then the PET film was cut using a cutting plotter and removed the extra PET film from the PVDF sheet. After that, the PVDF

sheet was cut into multiple small straps having the width of 3 and 5 mm (two different widths). Finally, the PVDF thin film-straps were inserted manually as warp and weft threads, to form a textile structure followed by testing of the piezoelectric performance was performed (Fig. 9d(i) & (ii)). It was observed that this piezoelectric fabric can generate a maximum power density of 81 and 125 $\mu\text{W}/\text{cm}^2$ with a fibre diameter of 5 mm and 3 mm, respectively. In addition, the piezoelectric performance of this fabric could be improved by using a lower diameter thread with their higher packing (more no. of threads in an unit area) in the fabric structure. Later, the textile-based wearable piezoelectric sensor used in the form of an energy harvester has been developed by the same research group [178]. In this case PVDF thin film-straps were also prepared by the same way as mentioned in the above research work. Then, the fabric was developed using the elastic hollow tubes and the as prepared PVDF thin film-straps. At the beginning, the elastic hollow tubes were woven into the mesh structure by intersecting one another in orthogonal direction. Thereafter, the PVDF straps were inserted through each of the column and rows to make a resultant fabric with the dimensions of $9 \times 9 \text{ cm}^2$. Finally, the piezoelectric performance of the developed textile based piezoelectric nanogenerator was examined. And it was found that the developed nanogenerator showed an output voltage of 51 V and a power of 850 μW under lateral stretching of the device. In this way, woven fabric-based piezoelectric nanogenerator and sensors have been developed which could perform very effectively. Thus, the woven fabric-based piezoelectric devices could be a big solution in the global electronic markets for altering electronic goods. At the same time, this piezoelectric device could be used directly as a sensor or actuator in various biomedical applications.

All the above-mentioned studies mainly focused on the 2D woven fabric based piezoelectric nanogenerators. In 2D woven fabric, the name itself implies that the fabric should have a specific length and width. Owing to their inherent dimensions, they can only show very limited piezoelectric output since they lower the deformation possibility during piezoelectric applications. On the other hand, 3D fabrics, which have a specific length, width and thickness can result in higher piezoelectric output due to the higher dimensional deformation possibility during piezoelectric applications. To take advantage of 3D fabric, researchers have tried to develop 3D fabric based piezoelectric nanogenerators. One such example, Talbourdet et al. [191] developed a 2D plain weave canvas and 3D diagonal interlock fabrics using PVDF melt-spun filament yarns (Fig. 9e (i) & (ii)). The fabrics were made-up on a hand loom using 100% PVDF multifilament having fineness of 246 tex. Two different weaves namely 2D plain canvas and 3D diagonal interlock were chosen for this study, since they have different characteristics. To experience the effect of the fabric weave, the piezoelectric performance was carried out for both 2D and 3D fabric. The piezoelectric performance of the plain weave canvas fabric in terms of output voltage was 0.14 V for the polarized fabric and 0.08 V for the non-polarized fabric. The piezoelectric performance of the 3D interlock fabric in terms of output voltage, it was 2.3 V for the polarized and 1.29 for the RMS fabric. Therefore, from this observation, it was concluded that the 3D diagonal fabric is a far better option for piezoelectric applications compared to the 2D plain-woven fabric. This is because 3D fabric deforms more compared to 2D fabric during the piezoelectric performance.

Although the electrical output of 2D fabric-based PENGs has been greatly improved compared with single fibre-based PENGs, further enhancement to the energy harvesting capability of fabric-based PENGs are still required. Moreover, woven fabric is not that flexible and therefore it is difficult to use it in remote locations. To solve this problem, 3D woven fabric and knitted fabrics come into the picture. The next section is focused on knitted fabric based piezoelectric energy harvesters.

6.2.2. Knitted structure-based piezoelectric nanogenerators (PENGs)

Knitted structure-based textile materials are another important route to develop lightweight, flexible and wearable piezoelectric energy

harvesters. Knitted-based piezoelectric devices can be integrated easily in different remote places where flexibility is the utmost requirement. For instance, a 3-D knitted spacer-based piezoelectric nanogenerator has been reported by Anand et al. [192] as shown in Fig. 9f(i) & (ii). The 3-D spacer fabric was developed by using a circular weft knitting instrument. Three types of threads were used for making this 3-D knitted piezoelectric fabric. i.e. conductive yarns, insulating yarns, and piezoelectric yarns. The conductive yarn was Ag coated PA66 yarn denoted by 'A', the insulating yarn was PET yarn denoted by 'B' and the piezoelectric yarn was PVDF monofilament denoted by 'C'. The PVDF monofilament was used as a spacer yarn in this knitted structure. The piezoelectric performance of this 3-D knitted spacer fabric was evaluated by the application of 0.106 MPa pressure (6 s). The output voltage and current generated by the fabric-based piezoelectric nanogenerator were 14 V and 29.8 μA , respectively. The power density of the nanogenerator was evaluated in such a way that, 1.10 $\mu\text{W}/\text{cm}^2$ to 5.10 $\mu\text{W}/\text{cm}^2$ power density was generated with the applied pressure in the range of 0.02–0.10 MPa. Therefore, the textile structure-based piezoelectric nanogenerator could be used as an energy harvester in various wearable electronics applications for supplying power. In another study, a piezoelectric strap was integrated into the textile structure by knitting method. In this regard, Atalay et al. [195] have reported a piezo strap integrated knitted fabric for health care monitoring. In their study, a piezoelectric PVDF strap was used to control the overall respiration and heartbeat. The piezoelectric nanogenerator worked based on the sensing vibration from the cardio respiration, heartbeat, and responding as an output signal. However, some unavoidable interferences were noticed in the output signal due to human body movement and surrounding causes. The Fast Fourier Transform (FFT) technique was used to evaluate average respiration and heartbeat frequency. Moreover, a digital filtering method (IIR) was used to filter the signals of breathing and heartbeat from the raw signal data. The piezoelectric fabric sensor recorded the frequencies 1.47 Hz and 0.2 Hz for the heartbeat and respiration rate, respectively. Based on the significant results, the prepared fabric sensor could be used in sports, the detection of sleep apnea, and the treatment of heart disease. Atalay et al. [196] have also reported a weft knitted fabric sensor for monitoring human respiration. The PVDF piezoelectric yarn was used in combination with conductive yarn. The respiration frequency of a human was evaluated similarly, as mentioned earlier.

6.2.3. Braided structure based piezoelectric nanogenerators (PENGs)

Recently, smart textile-based piezoelectric sensors become very popular due to their enough flexibility, lightweight, and the reasonable piezoelectric performances. However, these are associated with some disadvantages like lower power output, durability, and sensitivity, etc. It results in a lower interfacial area and poor electrical connection between the piezoelectric component and the electrode components. This problem could be solved by employing the new strategy of the textile structure, for example braiding technology [193]. In this regard, few study-based on tri-axial braided piezoelectric nanogenerators have been reported with multiple steps of braiding technology. The silver-coated nylon yarn has been used in the core of the whole birded structure which works as an internal electrode material. Thereafter, piezoelectric PVDF fibre has been braided on the silver-coated nylon core yarn which acts as a piezoelectric component. Finally, another silver-coated nylon yarn was braided on the piezoelectric fibre to provide the outer electrode material. The braiding process and whole structure of the tri-axial braided piezoelectric yarn were taken by the optical microscope are shown in Fig. 9g. The piezoelectric performance of the developed tri-axial braided piezoelectric device has been examined by dropping a ball from two different heights on the surface of the piezoelectric yarn-based device. After dropping, the ball bounce height was observed to be decreased. Each bounce of the ball acts as an impact on the surface of the device which generated an output signal. The output voltage values for the 0.17 mPa and 0.23 MPa applied pressure were found to be

230 mV and 380 mV, respectively. Another important observation was that the strength of the output signal gradually decreased with time in line with the decreasing bouncing height of the ball. Furthermore, it can be seen from the output signals of the piezoelectric device, that unequal output signals and higher negative voltage are obtained. It may be due to the unique tri-axial braided structure which enhanced the contact area between the PVDF piezoelectric part and the electrode part under the impact of the device. It can also be found that the output current and power from the tri-axial braided yarn-based piezoelectric nanogenerator have been varied by applying different pressures. The output voltage and power concerning the different capacities of resistance can also be observed. The maximum power of the piezoelectric device was found as 0.16 μW with the corresponding output voltage of 380 mV at the pressure of 0.23 MPa. The maximum power density of the braided yarn-based piezoelectric nanogenerator was 29.62 $\mu\text{W}/\text{cm}^3$. The value of power density was higher than the previously reported piezoelectric nanogenerator based on the PVDF polymer [193]. Thus, a piezoelectric nanogenerator based on braiding technology could be a great solution in the textile-based energy harvesting area in terms of its higher compatibility between the piezo material and electrode materials to render higher power density.

The piezoelectric performances and applications of the fabric based piezoelectric nanogenerators are shown in Table 6.

Finally, the different textile methods for preparation of the piezoelectric nanogenerators along with their advantages and disadvantages have been tabulated in Table 7. In addition, different textile structures based piezoelectric nanogenerators and their merits and demerits have also been summarized in Table 8.

7. Some strategies to improve the performance of PENG devices

Over the past few decades, different strategies have been tried to enhance the performance and applicability of piezoelectric nanogenerators. There are various strategies to improve the piezoelectric performance of piezoelectric nanogenerators, such as the selection of piezoelectric materials, structural modifications of piezoelectric materials, hybridization of piezoelectric materials, the selection of proper substrates, and the selection of preparation methods for piezoelectric materials. This section focuses on the brief discussion on various strategies to improve the performance of piezoelectric nanogenerators [197].

7.1. Selection of piezoelectric materials

There is a range of piezoelectric materials such as inorganic (ZnO,

Table 6
Output signals of various fabric based piezoelectric nanogenerators.

Materials	Methods	Voc (V)	Isc (μA)	Power (μW)	Applications	Ref.
PVDF-PET strap based woven fabric (array of device)	Weaving	51	16.67	850	Energy harvesting and sensing	[178]
PVDF-filament based woven fabric (twill)		3.5	0.29	0.13	Energy harvesting	[180]
PVDF-HDPE/CB yarn based woven fabric		13	-	-	Energy harvesting	[181]
PVDF electrospun yarn based woven fabric		1	-	-	Electronic skin for the health monitoring and human motion detection	[182]
PVDF-TrFE electrospun yarn based woven fabric		2.5	-	-	Energy harvesting	[184]
PVDF electrospun yarn/PET		2	109	-	Energy harvesting and pressure sensing	[183]
PVDF nanofibres based yarn/ CsPbI ₂ Br		45	470	-	Energy harvesting	[186]
PVDF-filament based fabric (3D interlock)		2.3	-	-	Energy harvesting	[191]
PVC-BaTiO ₃ NRs-hybrid yarn based woven fabric		1.9	0.024	0.046	Energy harvesting and a LCD glowing	[185]
PVDF/BaTiO ₃ hybrid yarns		36.05	3.126	-	Energy harvesting	[187]
PVDF/ZnO hybrid yarns		1.12	52	-	Energy harvesting	[194]
Cu/Ni coated PET/ZnO		1.32	0.56	-	Energy harvesting	[188]
PVDF-PET cast film 7 warp and 5 weft)		38	10	380	Energy harvesting	[190]
PVDF/AgNylon/PET	Knitting	14	29.8	-	Energy harvesting	[192]
PVDF/AgNylon/PTFE		-	-	-	Health monitoring	[195]
PVDF/Ag coated Nylon	Braiding	0.230	-	0.16	Energy harvesting	[193]

Table 7

Different textile methods for preparation of the piezoelectric nanogenerators along with their advantages and disadvantages.

Textile PENG fabrication methods	Advantages	Disadvantages
Electrospinning	In-situ poling, flexible materials, high aspect ratio of the fibres	High voltage risk, high production cost, low yields of fibres
Melt spinning	High production, low cost	High operation temperature, only meltable polymers can be processed
Solution spinning	In-situ phase change of polymer can take place	Poor conductivity, chemical safety required
Weaving (woven fabric)	High structural stability, easy to wear, high strength	Low dimensional deformations
Knitting (knitted fabric)	Flexible, high compressibility, high porosity	Low structural stability
Braiding (braided fabric)	High dimensional stability	Cost effective method

Table 8

Different textile structures based piezoelectric nanogenerators and their merits and demerits.

Methods	Merits	Demerits
Textile fibre based PENGs (staple fibres and filaments)	Easy to incorporate into the cloth, flexible	Low piezoelectric outputs
Electrospun fibre based PENGs	High piezoelectric outputs	Low durability and stability
Textile fabric based PENGs	High dimensional stability, high piezoelectric outputs than staple fibre or filament based PENGs	High complexity in the preparation method

BaTiO₃, KNN, PZT, ZnSnO₃, etc.) and organic (PVDF, PVDF-TrFE, PAN, Nylon 11, etc.) materials. Inorganic piezoelectric materials can show better piezoelectric performance since they have better piezoelectric characteristics (piezoelectric constant, Curie temperature, etc.) compared to organic based piezoelectric materials. However, inorganic materials are brittle by nature which limits their applications in different remote areas, while polymeric piezoelectric materials are flexible and has ability to take shape in a complex form. Therefore, selecting appropriate piezoelectric materials is an important criterion for any targeted applications.

7.2. Structural modifications of piezoelectric materials

As mentioned above, both organic and organic based materials have their own pros and cons for piezoelectric applications. Thus, combination of both (composite structure) can be a strategic approach to obtain best performance. Even, the structure of the inorganic materials within a composite plays a vital role to excel their properties. These materials can be modified into different forms, such as nanoparticles, nanorods, nanowires, etc. The nanostructures based piezoelectric materials will have better piezoelectric performance compared to bulk piezoelectric materials with their lower incorporation inside the matrix. Hence, structural modification is also an essential factor for improving performance of piezoelectric nanogenerators.

7.3. Hybridization of piezoelectric materials

The piezoelectric materials, especially the inorganic materials, have different piezoelectric characteristics. For instance, ZnO is the most usable semi conductive piezoelectric material, but it has lower piezoelectric performance compared to PZT, KNN, etc. Therefore, hybridization or doping is required to enhance the charge separation ability (more asymmetric) of the fine structure of crystalline zone and to improve the piezoelectric performance.

7.4. Selection of suitable substrates

Another important aspect is the selection of suitable substrates to improve piezoelectric performance. A flexible substrate like cellulose paper based piezoelectric materials can show better piezoelectric performance, owing to the higher dimensional deformability of piezoelectric nanogenerators composed of the same.

7.5. Selection of suitable preparation methods

Last but not the least, the selection of a proper preparation method to develop piezoelectric materials is also an important factor to obtain best piezoelectric performance. For example, the solution cast method for preparation of the PVDF polymer based piezoelectric material is advantageous. Because, in the solution cast method formation of β -crystalline phase takes place during its preparation itself if we use polar solvent like DMF and so on. But the developed β -crystals may not be oriented in a particular fashion. Thus, an extra arrangement for the poling of the solution cast film is essential, which may increase the additional cost. To resolve this problem of poling, the electrospinning method has been extensively explored to prepare in-situ poled piezoelectric materials. Moreover, electrospun piezoelectric materials are more flexible and they have higher a surface-to-volume ratio, which results in higher piezoelectric performance compared to other counter parts. However, electrospun piezoelectric materials have durability and long-term stability issues. Therefore, a suitable materials preparation method is crucial for improved piezoelectric performance.

8. Conclusions and future perspectives

The demand for energy harvesting techniques continues to grow worldwide. The reason behind this is the energy crisis that the world faces – something which has become the dominating issue of our generation. Generally, devices like batteries contain electrochemical energy, which can limit their applications to some extent as they need to be recharged periodically and they are also not environmentally friendly. This problem can be mitigated by including setups that can harvest ‘free’ energy sustainably. Harvesting energy mechanically includes methods like induction by the process of electromagnetism, static and pressure electricity. Such pressure electric devices offer the most favourable outputs as they are cheaper, easier to prepare, and finally can enable the production of efficient energy. Pressure-based electric materials include

semiconductor (ZnO, ZnS, GaN, etc.), ceramics (PZT, BaTiO₃, NaNbO₃, KNbO₃, KNN, ZnSnO₃, etc.), polymers (PVDF and its copolymers, polyamides, parylene-C, etc.), and naturally produced piezoelectric materials such as bone, hair, collagen fibrils, peptide, cellulose and sugar cane, etc. PZT is a widely applicable ceramic material; however, it hampers the ecosystem as it includes 60% lead. Such unfavourable conditions can be minimised by incorporating lead-free substances (such as the use of ceramic materials). Much research has been carried out on solution cast film based piezoelectric devices. Either pure piezoelectric polymer or filler incorporated in the polymer (composite) based materials have been utilized to develop piezoelectric devices. But in the case of solution cast piezoelectric materials, an extra electrical poling step is compulsory for getting better piezoelectric properties. Another problem associated with the solution cast method is the greater likelihood that fillers may reduce the flexibility of the polymer which can limit its ability to operate in situations requiring flexibility (such as at various locations on the human body). Therefore, to solve such problems the various textile-based piezoelectric devices have been studied and reviewed in detail. For instance, various fibre-based piezoelectric nanogenerators by including Ce³⁺ doped PVDF/graphene composite nanofibrous webs have been developed by the process of electrospinning. Such processes have enabled significant results along with strong piezoelectric beta fraction obtained without using any latter applied poling method. As analysed, in-situ fabrication produces a voltage of as high as ~11 V together with a current density of about ~6 nA/cm² by the spontaneous action of stress applied mechanically which can glow ten LEDs. Also, its vibrations are caused by the ecosystems like the flow of wind, water, and transport of automobiles and such nanogenerators can be capable of powering 3 blue LEDs which can offer the preparation of highly sensitive nanogenerators. However, the electrospinning method is not an easy process to develop nanofibrous web based piezoelectric materials, since there are numerous parameters in the electrospinning method which must be tuned perfectly. Nowadays the demand for wearable and portable electronic devices is growing continuously. Based on the experience of several studies, it has been observed that piezoelectric devices can be fabricated having lightness, flexibility, and compatibility which are the main requirements for wearable and portable electronic devices. In this context, textile based piezoelectric devices can be a good candidate to supply power in wearable devices and electronic textiles. By these kinds of textile based piezoelectric devices, various waste mechanical energy generated during walking and human body movement can be harvested as usable electrical energy which can be directly used in various wearable and portable electronic devices. For this reason, many works on woven, knitted, and braided-based textiles have already been carried out effectively in piezoelectric energy harvesting applications. And based on significant results, it can be concluded that the development of textile-based piezoelectric flexible devices may open an innovative area of research to harvest energy from environmental resources in the coming years. Therefore, in the wearable and portable electronic markets, textile-based flexible piezoelectric devices can be utilized as a good candidate to harvest energy and supply this energy to self-powering electronic devices. Even though significant progress has been made in the field of textile-based piezoelectric nanogenerators, there is still a vast gap between the research and the commercial applications of these devices. Therefore, in this present review, the present challenges and possible perspectives of the textile-based piezoelectric nanogenerators have been explained in Fig. 10.

8.1. Electrical output

Textile based piezoelectric devices can only produce a very low amount of voltage and current, which results in lower power density, i. e., in the range of a few nW/cm² to a few μ W/cm². Hence, still, it is a challenging task to enhance the power density of the piezoelectric device. Lots of research has already been carried out to improve

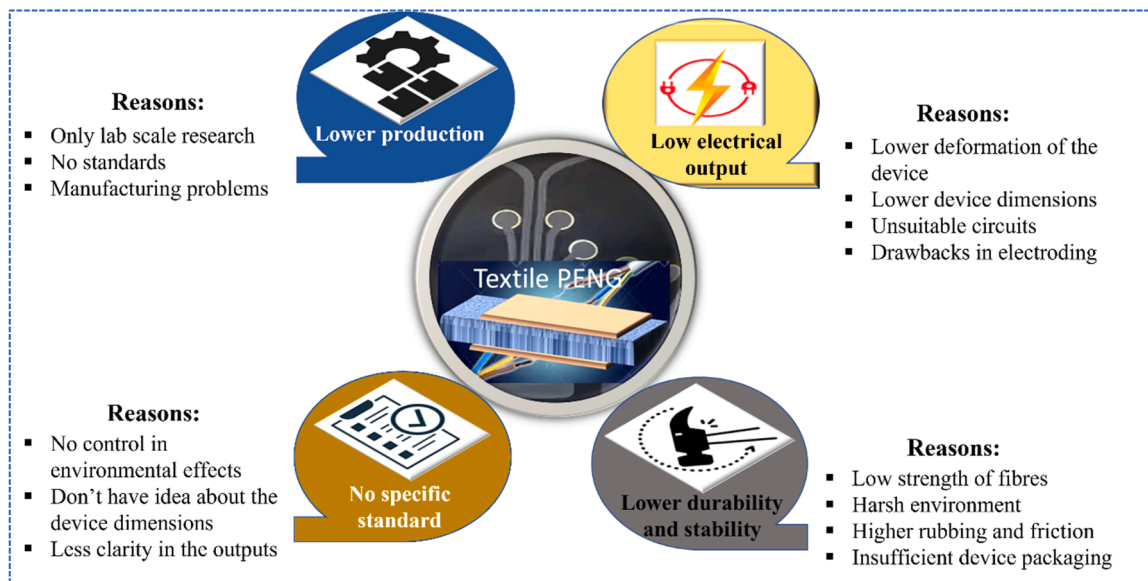


Fig. 10. Summary of research challenges facing textile-based piezoelectric energy harvesters.

piezoelectric power density by modifying materials, the structure of the device, and so forth. However, to enhance the power density of piezoelectric devices, more research needs to be done. We recommend various ways in which the power density of the textile based piezoelectric device might be enhanced. For instance, (i) **fabrication of 3D fabric based piezoelectric device**: it is expected that the 3D textile fabric based piezoelectric nanogenerators will have more piezoelectric properties as compared to single fibre, and 2D fabric based piezoelectric nanogenerators, since more dimensional deformation will be taking place during the piezoelectric characterizations. In this prospect, although few works have already been carried out on the 3D textile fabric based piezoelectric nanogenerators [191,192], still research needs to be done to improve performance of the textile based piezoelectric nanogenerators, (ii) **increase the area of textile based nanogenerators**: at the current scenario the dimension of the textile based piezo device is in the range of 1–25 cm², hence, increasing the dimension of these soft devices might be another vital approach to enhance the performance of the textile based piezoelectric devices. However, the proposed dimension should be realistic in terms of wearability, since it is an important factor for the wearable electronics, (iii) **critical analysis of the interface between piezoelectric material and electrode**: the interface between piezoelectric material and electrode has a tremendous influence on the output electrical properties of the piezoelectric device. The interface should have enough capability to transfer all surface charges to the electrode, in some cases, specifically for porous piezoelectric materials (electrospun web, other textile structures, and so on), conductive (Al, Cu, and Ag) tape has been used as an electrode material, and hence, there is the likelihood of (nano or micron-sized) gaps between the piezoelectric material and an electrode. These gaps at the interface region of the piezoelectric material and the electrode must be studied to explore their effect on output. Additionally, direct deposition of conductive material onto the piezoelectric device has been explored as a solution for the gap issue between the electrode and piezoelectric materials. However, a drawback still exists in terms of the charge transfer capability of the electrode, as a surface-deposited electrode has a higher capacitance value. To address this problem, researchers have attempted to fabricate interdigitated electrodes (IDE), and it has been found that IDE have a lower capacitance value, enabling more effective charge transfer to circuits adjacent to the piezoelectric device [198]. Therefore, in the future, researchers may explore the use of IDE-type electrode materials to improve the performance of textile-based piezoelectric devices, (iv) **suitable circuit design**: a suitable circuit design is

necessary to capture harvested energy in a piezoelectric device. There are many factors, such as the resistance of wires, type of resistance, type of capacitance, and more, that can significantly impact the output electrical properties of piezoelectric devices. Specifically, to supply power from textile-based piezoelectric devices, a rectifying circuit is necessary to convert AC signals to DC signals. However, rectifying circuits have two drawbacks: diode forward voltage decreases the maximum voltage across the storage device (capacitor), and diode reverse leakage current decreases the output power of the piezoelectric device [199,200]. Therefore, a suitable circuit design for extracting power from textile-based piezoelectric devices needs to be developed in future research, (v) **design of the suitable power management circuits**: from the very beginning, in the year 2001, power management circuits for piezoelectric devices have been disclosed in the literature, but a complete overview is still lacking. However, in 2018, Dell'Anna et al. [201] provided a comprehensive review of power management circuits to improve the performance of piezoelectric devices. Therefore, future studies should address the use of suitable power management circuits to scavenge power from textile-based piezoelectric devices, (vi) **use of the multiple piezoelectric device**: piezoelectric device based on the multi-layers stacking have already been reported by various research groups to improve the performance of the textile based piezoelectric nanogenerators [74,189,196,202,203]. However, more emphasis needs to be placed on future research to enhance the performance of textile based piezoelectric nanogenerators.

8.2. Durability and stability of the device

The durability of the piezoelectric device is one of the most essential properties in the case of wearable electronics. In this regard, the durability of fibres and textile based piezoelectric energy harvesters is still not adequate for use as energy harvesters for long term use. Because if we look into fibre-based piezoelectric energy harvesters, electrospun nanofibres have shown good piezoelectric properties compared to melt spun and solution spun fibres, but their mechanical strength is very low [198,199]. Moreover, electrospun nanofibres can only be utilized as patches by integrating them in the textile fabrics. In wearable electronics, repeated friction and rubbing during human activities may result in lower lifetime of the fibre-based piezoelectric energy harvesters. In addition, the performance of textile based piezoelectric energy harvesters gets declined gradually due to the influences surrounding conditions like temperature, humidity, etc. Therefore, more

research needs to be carried out to solve all these practical issues with the textiles-based energy harvesters. The piezoelectric properties get reduced with increasing working temperature, since the materials lose their piezoelectric characteristics above their Curie temperature [200, 201]. Likewise, the humid environment also affects the piezoelectric performance; mainly piezoelectric effect deteriorates in the presence of water [204]. Furthermore, the long-term exposure of the piezoelectric device may create problems with the accumulation of contaminants, which results in lower piezoelectric outputs. Therefore, increasing the service life of the textile based piezoelectric devices needs to be taken care of in future research. The durability of the textile based piezoelectric device might be improved by proper flexible encapsulation of the device.

8.3. Nullifying the triboelectric contribution from piezoelectric output

Numerous research studies have investigated piezoelectric nanogenerators made from solid films or flexible textiles based on piezoelectric materials. These investigations have demonstrated exceedingly high piezoelectric output, sometimes even exceeding those of triboelectric nanogenerators. However, it is important to consider the appropriate range of output for both piezoelectric and triboelectric nanogenerators. Piezoelectricity is a bulk property of a material, whereas triboelectricity is a surface property of materials. Therefore, researchers should take the triboelectric effect into account when measuring piezoelectric outputs. However, it may be challenging task to separate these two characteristics during measurement of piezoelectric performance, leading to a surge of research reports on combined results for a single piezoelectric device [147,150,205–208]. Wearable textile-based piezoelectric nanogenerators pose a particular risk of the triboelectric effect due to the friction and rubbing necessary to apply pressure to these devices. While extensive research has been conducted on textile-based piezoelectric nanogenerators, there remains a significant gap in reducing the triboelectric effect from these devices. Nullifying the triboelectric effect from piezoelectric textile-based nanogenerators is a complex problem and requires careful consideration of material selection, surface modification, electrode and mechanical design, and measurement techniques. This area in textile-based piezoelectric nanogenerators presents a significant opportunity for future research in this field.

8.4. Standardization

To date, there is no specific standard for developing and characterizing the textile-based piezoelectric energy harvesters and therefore, it is very difficult to be conclusive about the optimum piezoelectric performance of a device. Various parameters are reported by different labs (output voltage, current, power, dielectric constant, dielectric loss, capacitance, sensitivity, and so on) but very often there are differences in device construction and testing that make any meaningful comparison between results difficult. Examples of parameters that are not well controlled in tests include the applied normal contact pressure, the later dimensions of the device, device thicknesses, electrode material characteristics and attachment to the PENG device, insulation, wire connection, circuit, etc. Therefore, much more advanced standardisation is required.

8.5. Large-scale production

One of the most advantageous tasks for their commercial applications is the industrial-level production of textile-based piezoelectric energy harvesters. To date, textile-based energy harvesters have only been fabricated on a lab scale, but researchers now need to develop approaches that can facilitate large scale production. Nevertheless, many attempts have been made in this direction, but there are many challenges in terms of proper material selection, suitable fabricating

method, and liability cost, etc., that need to be overcome. With respect to materials, single fibres or yarns have a curvilinear surface, which results in a lower real contact area among the fibres or yarns, although they have a higher specific surface area. On the other hand, 3D fabric or multilayer fabric stacking-based piezoelectric nanogenerators can show better piezoelectric performance, but they have wearability challenges. Despite those shortcomings, fabrication of a textile-based piezoelectric nanogenerator using piezoelectric fibres or yarns is an important step. In textile manufacturing, there are many factors (such as rubbing and friction of the piezoelectric materials with the different machine parts, etc.) that may cause the inherent piezoelectric characteristics of the piezoelectric fibres or yarns to be reduced and need to be considered during the fabrication of a device. Moreover, a proper design of the device is also an essential task if someone intends to commercialize a textile-based piezoelectric nanogenerators.

CRediT authorship contribution statement

Satyanarjan Bairagi: Conceptualization, Investigation, Validation, Formal analysis, Writing – original draft, Writing – review & editing. **Shahid-ul-Islam:** Supervision, Investigation, Project administration, Software, Validation, Writing – review & editing. **Mohammad Shahadat:** Investigation, Methodology, Software, Validation, Writing – review & editing. **Daniel M. Mulvihill:** Supervision, Investigation, Project administration, Software, Validation, Writing – review & editing. **Wazed Ali:** Supervision, Conceptualization, Project administration, Resources, Writing – original draft, Writing – review & editing, Funding acquisition.

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

No data was used for the research described in the article.

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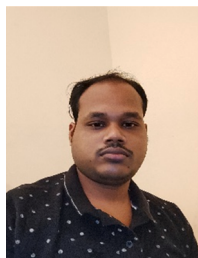
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