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# Wearable nanocomposite textile-based piezoelectric and triboelectric nanogenerators: Progress and perspectives

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#### ABSTRACT

In recent years, the widespread adoption of next-generation wearable electronics has been facilitated by the integration of advanced nanogenerator technology with conventional textiles. This integration has led to the development of textile-based nanogenerators (t-NGs), which hold tremendous potential for harvesting mechanical energy from the surrounding environment and serving as power sources for self-powered electronics. Textile structures are inherently flexible, making them well-suited for wearable applications. However, their electrical performance as nanogenerators is significantly limited when used without any modifications. To address this limitation and enhance the electrical performance of textile-based nanogenerators, nanocomposite textiles have been extensively utilized for fabricating advanced nanogenerators. This critical review focuses on the recent progress in wearable nanocomposite textiles-based piezoelectric and triboelectric nanogenerators. The review covers the fundamentals of piezoelectricity and triboelectricity, the working principles of nanogenerators, and the selection of materials. Furthermore, it provides a detailed discussion of nanocomposite textiles in various forms, such as fibers or yarns, fabrics, and electrospun nanofibrous webs, which are employed in piezoelectric and triboelectric nanogenerators. The review also highlights the challenges associated with their implementation and outlines the prospects of textile-based nanogenerators. It can be concluded that nanocomposite textile based piezoelectric and triboelectric nanogenerators exhibit better electrical output and mechanical strength compared to conventional textile based nanogenerators. Nanocomposite electrospun web based piezoelectric nanogenerators exhibit higher piezoelectric output compared with nanocomposite fibre/yarn or fabric based piezoelectric nanogenerators. This is because an in-situ poling takes place in electrospinning unlike with fibre or fabric based piezoelectric nanogenerators. Nanocomposite electrospun web based triboelectric nanogenerators also show better triboelectric output compared to the fibre or fabric-based equivalents. This is due to the higher contact area developed with electrospun nanocomposite webs compared to the fibre or fabric cases. Overall, it can be concluded that while nanocomposite construction can boost output and durability of textile based nanogenerators, more research is required to bring output, stability and durability up to the levels achievable with non-textile based devices.

### 1. Introduction

In recent years, there has been a growing interest in small-scale

wearable and portable electronic devices, which have become an integral part of people's daily lives, enabling smart lifestyles [1–5]. This interest is primarily driven by their lightweight, small architecture, and

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low power consumption. These devices are typically powered by batteries, and as a result, ongoing research has focused on modifying and improving battery technology [6,7]. These modifications aim to increase the energy density and overall performance of batteries. However, batteries have inherent limitations, such as a limited lifespan, requiring periodic recharging, which can eventually impact the efficiency of electronic devices. To address this battery life issue, researchers are exploring alternative power sources for small-scale portable and wearable electronic devices [8]. In this context, piezoelectric [9,10], triboelectric [11-14], and thermoelectric [15,16] harvesting technologies have emerged as potential solutions for providing continuous power to various small electronic devices. These technologies have the potential to convert renewable environmental waste into usable energy. Renewable energy sources such as solar energy [17,18], thermal energy, and mechanical energy can be harnessed for this purpose. Among them, mechanical energy is particularly abundant in the surrounding environment [1,19]. Therefore, the ubiquitous utilization of mechanical energy in wearable and portable electronic devices through piezoelectric or triboelectric energy harvesting technologies holds great promise for wearable and portable electronic devices. Each of the mentioned energy harvesting technologies have its own advantages and drawbacks, but they can be employed based on the environmental energy sources including solar energy, thermal energy, electromagnetic energy, and mechanical energy as illustrated in Fig. 1. For example, piezoelectric and triboelectric technologies have been successfully employed to harvest electrical energy from mechanical energies present in our surroundings. On the other hand, thermoelectric technology is utilized to scavenge electrical energy from thermal energy in the environment. Among these, triboelectric nanogenerators exhibit higher electrical outputs compared to other counterparts [20].

Triboelectric energy harvesting is a technology that enables the scavenging of electrical energy through tribo-electrification and electrostatic induction when two dissimilar material surfaces come into contact [22–25]. A triboelectric nanogenerator (TENG) compromises two materials with different electron affinity (positive or negatively charged) and an electrode that produces an external current circuit [26]. The development of a triboelectric nanogenerator (TENG) involves employing various suitable manufacturing processes based on specific application requirements. [27-30]. To date, several methods have been employed for the development of triboelectric nanogenerators. These methods include as the solution cast film method, direct textile methods such as weaving, knitting, braiding, and nonwoven techniques, among others. Among these methods, textile-based triboelectric nanogenerators (t-TENGs) have garnered significant interest compared to other approaches [31]. This is primarily attributed to their light weight, good flexibility, easy processability, cost effectiveness as well as other advantages [32-35]. Numerous research groups have conducted studies to explore the fabrication of highly promising triboelectric nanogenerators by optimizing their structure and manufacturing methods. Despite these efforts, the development of triboelectric nanogenerator structures and manufacturing methods is an ongoing process, aimed at achieving large-scale production and high-performance products [36].

In recent years, hybrid energy harvesters (piezoelectric and triboelectric) have also attracted attention to supply power to the different wearable electronic devices, because hybrid energy harvesters can generate high output current and voltage. However, the suitable structural designs and polarization direction modulations are very important factors to get combined piezoelectric and triboelectric effects. Moreover, the piezoelectric and triboelectric effect can easily be combined since they have matching impedance, similar structural features, and more importantly, both of them possess the capability to convert mechanical energy to electrical energy [37,38].

Due to the increased interest in wearable electronics, researchers have been actively developing wearable nanogenerators to power various small-scale wearable and portable electronic devices. Piezoelectric and triboelectric nanogenerators, fabricated using films, can be attached as patches to wearable garments to harvest mechanical energy from the human body. Alternatively, direct textile structure-based piezoelectric and triboelectric nanogenerators offer an appealing approach for the development of wearable smart textiles.

	Photovoltaic	Thermoelectric	Electromagneti	e Piezoelectric	Triboelectric
	light	++++++ Heating	Electromagnetic field	Pressure	Friction ++++++
Voltage	0.5 - 0.9 V	0.1 - 1.0 V	0.1 - 10 V	1 -100 V	3 -1500 V
Current	100-500 μA	5 -30 μΑ	-	$0.01-10\;\mu A$	10 -2000 μA
Power density	$5-30 \text{ mW/cm}^2$	$0.01-3 \text{ mW/cm}^2$	-	$0.001 - 30 \text{ mW/cm}^2$	$0.1-100 \text{ mW/cm}^2$
Efficiency (%)	0.5 - 45	0.1 - 25	5 - 90	0.01 - 21	10 - 85
Merits	<ul> <li>High power output</li> <li>Continuous DC output</li> </ul>	<ul> <li>Easy scale down</li> <li>No moving component</li> </ul>	<ul> <li>High conversion efficiency</li> <li>Large current at low voltage</li> </ul>	<ul> <li>High sensitivity to stimuli</li> <li>Easy integration</li> <li>Easy miniaturization</li> </ul>	<ul> <li>High power output</li> <li>High conversion efficiency</li> <li>No material constrain</li> <li>Cost-efficient</li> <li>Structural flexibility</li> </ul>
Demerits	<ul> <li>Light present is necessary</li> <li>Low conversion efficiency for organic solar- cells</li> </ul>	<ul> <li>High temperature difference required</li> <li>Low conversion efficiency</li> <li>Low output</li> </ul>	<ul> <li>Heavy weight</li> <li>Short range power transmission</li> <li>Structural complexity</li> </ul>	<ul><li>High impedance</li><li>Low conversion efficiency</li><li>Low output</li><li>Pulse output</li></ul>	<ul> <li>High impedance</li> <li>High frictional damage</li> <li>Pulse output</li> </ul>

Fig. 1. Comparison of different energy harvesting technologies [21].

Considering the recent advancements in textile-based nanogenerators (t-NGs) for energy harvesting and self-powered electronics, this comprehensive review provides an overview of wearable nanocomposite textile-based nanogenerators (t-NGs) for energy harvesting and self-powered electronics (see Fig. 2). The review begins by focusing on the fundamental concepts of piezoelectricity and triboelectricity, explaining the working principles of piezoelectric and triboelectric nanogenerators, and discussing the selection of suitable materials. A subsequent section of the review delves into a detailed discussion of nanocomposite textiles based on piezoelectric and triboelectric nanogenerators. Additionally, the review explores the potential avenues for overcoming the challenges and highlights the future prospects of nanocomposite textile-based piezoelectric and triboelectric nanogenerators, emphasizing their role in advancing wearable and portable electronics, energy harvesting, and other related fields.

# 2. What is piezoelectricity and triboelectricity?

Piezoelectricity means an electricity generation due to pressure. Furthermore, piezoelectricity refers to the inherent property of certain materials to generate electricity when subjected to mechanical force, and conversely, to deform or vibrate when an electric field is applied to them. The process of capturing or harnessing the electric energy generated through piezoelectricity is commonly referred to as piezoelectric energy harvesting. There are two type of piezoelectric effects; namely, direct piezoelectric effect discovered by Pierre and Jacques Curie in 1880 [45] (mechanical energy is converted into the electrical energy) and inverse piezoelectric effect discovered by G Lippman in 1881 [46] (electrical energy is converted into the mechanical energy). Generally, sensors work on the direct piezoelectric effect while actuators work on the inverse piezoelectric effect. Materials having piezoelectric characteristics are known as the piezoelectric materials [47]. To understand piezoelectricity, one should consider a systematic theory of piezoelectricity [48-50]. The combined occurrence of the Hooke's law for liner elastic materials and the linear behaviour of piezoelectric materials is known as piezoelectricity. These two properties' respective mathematical formulae are as follows:

$$D = \varepsilon E \Rightarrow D_i = \Sigma_i \varepsilon_{ij} E_j \tag{i}$$

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

where, D,  $\varepsilon$  and E are the electric flux density [51], the permittivity, and the electric field strength. On the other hand, S and s are denoted as the linearized strain and compliance under short-circuit conditions and T is the applied stress.

Further, these two above mentioned formulas can be combined to a single formula:

$$S = sT + \partial^{t} E \Rightarrow S_{ij} = \Sigma_{k,l} s_{ijkl} T_{kl} + \Sigma_{k} d^{t}_{ijk} E_{k}$$
(iii)

$$D = \partial T + \varepsilon E \Rightarrow D_i = \Sigma_{i,k} d_{iik} T_{ik} + \Sigma_i \varepsilon_{ii} E_i$$
(iv)

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

In matrix form,

$$\{S\} = \begin{bmatrix} s^E \end{bmatrix} \quad \{T\} + \begin{bmatrix} d^t \end{bmatrix} \quad \{E\}$$
(v)

$$\{D\} = [d] \quad \{T\} + [\varepsilon^T] \quad \{E\}$$
(vi)

where, [d] and  $[d^t]$  are the matrices for the direct piezoelectric effect and the converse piezoelectric effect. Whereas, *E* and *T* denote the electric field and the stress field. The superscript *t* denotes transposition of a matrix [52,53].

On the other hand, triboelectricity refers to the generation of electric charge through repeated contact (e.g. sliding). This phenomenon, commonly known as static electricity, was first discovered by Thales of Miletus (Greek philosopher) about 2500 years ago. The triboelectric effect occurs when two dissimilar materials make repeated contact. This effect can be observed abundantly in our daily life. In past decades, different industries have mentioned that this electrostatic charge can lead to dust explosions, electronic damage, and ignition. However, from the energy point of view, the electrostatic charges involved can form the basis of an energy harvester. From this energy device concept, earlystage electrostatic generators such as the "friction machine" and "van de Graaff generator", were fabricated. The modern triboelectric



**Fig. 2.** Various nanocomposite textile based piezoelectric and triboelectric nanogenerators (a) PVDF (Poly (vinylidine fluoride)/ potassium sodium niobate (KNN) nanocomposite melt spun filament based piezoelectric nanogenerator [39], (b) BaTiO<sub>3</sub>/PVC nanocomposite and cotton yarns based woven fabric for the piezoelectric nanogenerator [40], (c) PVDF/KNN electrospun nanocomposite fibrous web based piezoelectric nanogenerator [41], (d) PVDF/ graphene electrospun nanocomposite web based triboelectric nanogenerator [42], (e) black phosphorus (BP) and hydrophobic cellulose oleoyl ester nanoparticles (HCOENPs)/PET fabric based triboelectric nanogenerator [43], and (f) PU/PTFE core-sheath fibre based triboelectric nanogenerator [44].

nanogenerator operates by the combined phenomenon of the triboelectric effect and electrostatic induction. Prof Z.L.Wang was the first to elucidate the concept of a triboelectric nanogenerator [54]. Triboelectric nanogenerators mainly harvest the waste mechanical energy which is available in different forms in our daily life [26]. The main physics behind this triboelectric effect for energy harvesting was first discussed by Wang et al. [55]. Wang et al. has discussed a relationship between the triboelectric output and the Maxwell's current displacement in this article. The electrical displacement vector as per Maxwell can be denoted as:

$$D = \varepsilon o E + P \tag{vii}$$

where, *E* and *P*, are the electric field and the polarization vector. Since displacement current has an immense role in the output of the triboelectric nanogenerator, the displacement vector and the displacement current density can be expressed as:

$$D = \varepsilon o E + P + P s \tag{viii}$$

$$JD = \frac{\partial D}{\partial t} + \frac{\partial P_s}{\partial t}$$
(ix)

In the case of triboelectric nanogenerators, the current density mainly depends on the surface charge density ( $\sigma_T$ ) and dielectric constant ( $\epsilon_1$ ,  $\epsilon_2$ ) of the triboelectric layers. Hence, the current density at short circuit condition can be denoted as:

$$J_D \approx \sigma T \frac{dH}{dt} \frac{\frac{d1\epsilon 0}{\epsilon 1} + \frac{d2\epsilon 0}{\epsilon 1}}{\left(\frac{d1\epsilon 0}{\epsilon 1} + \frac{d2\epsilon 0}{\epsilon 1} + z\right)^2}$$
(x)

The triboelectric nanogenerator having area of *A*, can generate output current of  $I = A \times J_D$ . So, an equation for this output current can be expressed as:

$$I \approx A\sigma_T \frac{dH}{dt} \frac{\frac{d1\epsilon 0}{\epsilon 1} + \frac{d2\epsilon 0}{\epsilon 1}}{\left(\frac{d1\epsilon 0}{\epsilon 1} + \frac{d2\epsilon 0}{\epsilon 1} + z\right)^2}$$
(xi)

Therefore, Eq. (xi) can be separated into three parts for a clearer understanding; namely, P1 is the surface tribo-charge density ( $\sigma_T$ ) which depends on properties of the triboelectric materials, P2 (dH/dt) is corresponding to the mode of operation of the triboelectric nanogenerator, and P3 ( $\frac{\frac{d1(t)}{r_1} + \frac{d2\rho}{r_1}}{\left(\frac{d1\epsilon_0}{\epsilon_1} + \frac{d2\epsilon_0}{\epsilon_1} + z\right)^2$ ) is attributed to the electrostatic induction

which depends on the permittivity and thickness of the triboelectric materials [56,57].

# 2.1. Piezoelectric vs triboelectric: working principles of nanogenerators (NGs)

The working mechanism of a piezoelectric device is illustrated in Fig. **3a**. Before applying any electrical field, the electric dipoles within

the composite-based material are randomly aligned between the two electrodes. When pressure is applied, the charge within the piezoelectric material separates, leading to a net polarization between the top and bottom of the surface, as shown in Fig. 3a. Consequently, positive, and negative charges accumulate at the bottom and top of the electrode, respectively, resulting in the generation of the positive voltage signals from the microdevice, corresponding to the first positive peak of voltage. Upon releasing the pressure from the device, a negative signal is generated. The piezoelectric potential arises from the movement and accumulation of free charges in piezoelectric materials. After returning to the original state, the accumulated electrons flow back in the opposite direction, resulting in a negative signal (Fig. 3a). The generated electrical energy is collected using an appropriate circuit directly connected to the electrodes [58,59].

As previously mentioned, the discovery of the triboelectric nanogenerator dates back to 2012. Since then, numerous researchers have been actively engaged in this field due to the increasing significance of TENGs. There are four different working modes of triboelectric nanogenerator, contact separation (CS), sliding mode (SM), single electrode (SE), and free standing (FS), modes (Fig. 3b). In the contact separation mode, as the name implies, two material surfaces undergo repeated cycles of normal contact (or tapping). In this mode, the two materials should have different electron affinity so that they can exchange charges between them. CS mode based TENGs generally have two components: contact material and electrode, respectively. When the two material surfaces come in contact by means of external force, one material surface loses charges (becomes positively charged) and the other surface gains the charges (and becomes negatively charged). On the other hand, when two material surfaces get detached from each other, a surface potential difference takes place on the material surface due to the unbalance of electric charges. Therefore, when two materials come in contact and get separated from each other, an electrical current flow through the wire connected with the materials until the TENG releases charges fully. In this process, when again force is applied, the potential difference will drop away due to the gap reduction between the two material surfaces and an electric current will flow in the opposite direction. Therefore, in this mode, the TENG will generate alternating current (AC) as shown in Fig. 3b. In contact separation mode, output current and voltage can be expressed as follows [57,60,61]:

$$I = c\frac{\delta V}{\delta t} + V\frac{\delta C}{\delta t} \tag{xii}$$

$$V = \frac{\sigma d}{\varepsilon_0} \tag{xiii}$$

where, *C* and *V* are the capacitance and voltage of the triboelectric nanogenerator (TENG) and  $\sigma$ ,  $\varepsilon_0$  are the charge density and vacuum permittivity, respectively and *d* is the distance between the two materials surfaces.

In sliding mode (SM), two dielectric materials are arranged in a similar manner to CS mode. However, the materials are sliding one over



Fig. 3. Working principle of the (a) piezoelectric and (b) triboelectric nanogenerator.

another tangentially rather than separating normally, which results in charge generation on the material surfaces. An AC output voltage is generated when an external load is connected to the TENG layers. The sliding mode operation of TENG is depicted in Fig. **3b**.

On the other hand, to reduce the manufacturing cost and energy harvesting from such applications where two electrodes are not compatible, a new modification in the TENG device has been carried out. Where a single electrode and one grounding arrangement based TENG is enough to harvest electrical energy from rotating tires, touched pads, and so on. The working mechanism of a single electrode TENG based on charge transfer between an electrode and the ground is shown in Fig. **3b(ii & iii)**.

A variation of the sliding mode where a single layer of dielectric material slides over and back on two laterally separated tribo-layers is known as the 'free-standing mode' - see Fig. 3b.

#### 2.2. Piezoelectric and triboelectric materials

Materials that can generate electric charge under external mechanical stress and vice versa are referred to as piezoelectric materials. The piezoelectric effect is observed in both natural and synthetic materials. Examples of natural piezoelectric materials include quartz (single crystal), SiO<sub>2</sub>, topaz, tourmaline group minerals, Rachelle salt (inorganic natural piezoelectric materials) and wood, silk, bone, rubber, dentin, hair, and enamel (organic natural piezoelectric materials). Synthetic piezoelectric materials can be classified into four groups namely piezoceramics (such as lead zirconate titanate (PZT), KNN, barium titanate (BT), etc.), piezo semiconductors (such as zinc oxide (ZnO), zinc stannate, etc.), piezopolymers (such as PVDF, poly (vinylidene fluoridetrifluoroethylene) (PVDF-TrFE), polypropylene (PP), nylon11 etc.), and piezocomposites (such as PVDF/KNN, PVDF-TrFE/BT, etc.). There are 32 crystal classes which are divided into seven groups such as triclinic, monoclinic, orthorhombic, tetragonal, trigonal, hexagonal, and cubic. Among these 32 classes, only 20 crystal classes have piezoelectric properties. Out of 20 crystals, 10 have polar characteristics, signifying spontaneous polarization is occurring without any external mechanical stress, and this is because of the non- cancelling electric dipole moment associated with their unit cell. The remaining 10 of the 20 crystal classes will show polarization only when they are subjected to an external mechanical load. Some of the piezoelectric materials along with their main piezoelectric properties are shown in Table 1 [62-64]. All the piezoelectric materials have some benefits and drawbacks; for example, single crystal piezoelectric materials have high mechanical quality factor, but they are very expensive. Similarly, ceramic piezoelectric materials have high piezoelectric constant and coupling factor, but they are brittle in nature (brittle piezoelectric materials are not suitable for using in certain applications). Polymer based piezoelectric materials have high flexibility (flexible piezoelectric materials can be used where flexibility is required unlike the ceramic based piezoelectric materials), but they have low piezoelectric constant, coupling factor, and dielectric constant. To solve individual drawbacks in ceramic and polymer based piezoelectric materials, composite based piezoelectric materials have become popular, but several material processing challenges remain in the case of composite piezoelectric materials [59].

Currently, there is a wide range of materials that exhibit triboelectric characteristics and can be employed in triboelectric nanogenerator applications. The selection of triboelectric materials plays a vital role in the performance of a triboelectric nanogenerator, as it relies on the utilization of two dissimilar layers with different polarities. The polarity basically depends on the capability of a material to gain electrons (an electro-negative material) or lose electrons (an electro-positive). The triboelectric materials can be arranged as per their polarity and charge density. The triboelectric series was first published by John Carl Wilcke in 1757 [78,79]. In reality, materials listed farther from the chart's centre tend to be more positively or negatively charged, and the distance between two materials in the series determines how much charge is

Table 1

Some popular piezoelectric materials and their main piezoelectric properties.

Piezoelectric M	aterials
-----------------	----------

Materials	Properties			
	Piezoelectric coefficient (d) (pC/N)	Relative permittivity (ε <sub>r</sub> )	Electromechanical coupling factor (k)	Ref.
Single crystals:				
AIN	$d_{15} = -4.07$ $d_{31} = -2$ $d_{33} = 5$	$\boldsymbol{\xi}_{33}=11.4$		[65]
BaTiO <sub>3</sub>	$d_{15} = 392$ $d_{31} = -34.5$ $d_{33} = 85.6$	$\begin{array}{l} \xi_{11} = 2920 \\ \xi_{33} = 168 \end{array}$	$\begin{array}{l} k_{15}=0.57\\ k_{31}=0.315\\ k_{33}=0.56 \end{array}$	[66]
LiNbO <sub>3</sub>	$d_{15} = 68$ $d_{31} = -1$ $d_{33} = 6$	$\begin{array}{l} \epsilon_{11}=84\\ \epsilon_{33}=30 \end{array}$	$k_{31} = 0.02$	[67]
LiTaO <sub>3</sub>	$d_{15} = 26$ $d_{31} = -3$ $d_{33} = 9.2$	$\begin{array}{l} {{\epsilon _{11}} = 53} \\ {{\epsilon _{33}} = 44} \end{array}$	-	[68]
Ceramics:				
BaTiO <sub>3</sub>	$d_{15} = 270$ $d_{31} = -79$ $d_{33} = 191$	$\begin{array}{l} {\xi _{11} = 1440} \\ {\xi _{33} = 1680} \end{array}$	$k_{15} = 0.57$ $k_{31} = 0.49$ $k_{33} = 0.47$	[69]
BiFeO <sub>3</sub>	$d_{33} = 37$	-	$K_{33} = 0.6$	[70]
PMN-PT	$d_{31} = -74$	$E_{33} = 1170$	$k_{31} = -0.312$	[71]
PZT-5A	$d_{31} = -171$ $d_{33} = 374$	$\boldsymbol{\xi}_{33}=1700$	$k_{31} = 0.34$ $k_{33} = 0.7$	[72]
KNN-BZ	$d_{33} = 400$	2	54.7	[73]
KNN-BT	$d_{33}=225$	1.06	36	[74]
Polymers:				
PVDF	$egin{array}{llllllllllllllllllllllllllllllllllll$	6–12	$k_{31} = 10.3$ $k_{33} = 12.6$	[75]
P(VDF-TrFE)	$d_{31} = 12 - 25$	18	$k_{31} = 0.16 \ k_{33} = 0.29$	[76]
P(VDF-HFP)	$d_{31} = 30-43$ $d_{33} = 24$	11	$k_{31}=0.187 \ k_{33}=0.36$	[77]
Nylon 11	$d_{31} = 14-100$ $d_{33} = 4$	5	$k_{31} = 0.049$	[77]
Polylactic acid (PLA)	$d_{31} = 1.58$	3–4	-	[77]
Cellulose	$d_{31} = 1.88 - 30.6$ $d_{33} = 5.7$	-	-	[77]
Polyurethane (PU)	$d_{31} = 27.2$	4.8–6.8	-	[77]

transmitted between them. The list of materials with triboelectric properties is shown in Fig. 4 below. The materials that are most compatible with one another and provide the most triboelectric interactions are those that have comparable opposing electric charges.

#### 3. Fundamentals of textiles and textile nanocomposites

Fibres serve as the primary raw material in textile manufacturing and can be broadly classified into natural or synthetic fibres. Regardless of their origin, fibers are characterized by certain parameters including a high length-to-width ratio, flexibility or softness, dyeability, good strength, and elongation. Examples of natural fibres include cotton (seed fibre), sisal (leaf fibre), jute (bust fibre), silk (protein fibre), wool (protein fibre), coir (fruit fibre), and many more. On the other hand, synthetic or manmade fibres are polyester, polypropylene, nylon, glass, metallic, specialty fibres and so on. Historical records indicate that fibers were initially sourced from natural materials (plants or animals), and it was only in the 1940 s and 1950 s that synthetic fibers were developed from chemical sources. In addition to natural and synthetic fibers, there is also a category of regenerated fibers (such as viscose, rayon, cellulose acetate, etc.), which are derived from natural cellulose sources, but undergo chemical treatment during the manufacturing process.[81,82].

A yarn refers to a strand of fibers that has undergone the process of

Triboelectric materials	Electron affinity (nC/J)	+	Triboelectric materials	Electron affinity (nC/J)
Polyurethane (PU) foam	+60	1	Sorbothane	+58
Hair, oily skin	+45		Solid PU	+40
Magnesium fluoride	+35		Nylon, dry skin	+30
Machine oil	+29		Nylatron	+28
Glass (soda)	+25		Paper	+10
Wood (pine0	+7		Cotton	+5
Nitrile rubber	+3		Wool	0
Polycarbonate	-5		Aervlie	-10
Epoxy	-32			25
PET (mylar) solid	-40		(SB) rubber	-35
Gum rubber	-60			
Polyimide	-70		EVA rubber	-55
Vinyl	-75		Polystyrene	-70
Polypropylene (PP)	-90		Silicones	-72
Cellulose nitrate	-93		LDPE	-90
(CN)	00		HDPE	-90
(PCP)	-98		UHMWPE	-95
Natural rubber	-105		PVC (rigid vinyl)	-100
(Latex)			Viton, filled	-117
Epichlorohydrin (ECH) rubber	-118		Santoprene rubber	-120
Hypalon rubber	-130		Butyl rubber, filled	-135
EDPM rubber	-140		PTFE (Teflon)	-190

Fig. 4. Triboelectric series for selection of materials in TENGs [80].

spinning. Two main types of yarns are commonly used: spun yarns, which consists of fibers with a specific length, and filament yarns, which are composed of continuous fibers. Filament yarns are typically made from synthetic fibers, while spun yarns are made from natural fibers. A twisting process takes place in the spinning of natural fibre-based yarns in order to improve the coherence force between the fibres to form a yarn. There are different spinning methods used to produce synthetic yarn, including melt spinning, wet spinning, and dry spinning. The melt spinning process starts with melted polymer chips being extruded into filaments by extruders. Wet spinning involves the coagulation of polymer solution using nonsolvent to form a filament. Then these yarns are used to make the textile fabric having a different structure like woven, knitted, braided (Fig. 5).

nonwoven, knitting, and braiding. Weaving is the process of interlacing two different types of yarn (warp and weft). The textile design affects how woven cloth is made. This design classifies woven cloth as plain, twill, satin, and many more terms. Similarly, knitting is the process of two yarns (wale and course) interloping with one another. There are many different knitting patterns, including plain, rib, purl, interlock, etc. On the other hand, by interlocking the fibres together, a nonwoven fabric structure can be created. (Fig. 5). One of the most promising processes for producing nonwoven fabrics is needle punching. Finally, a braiding machine is used to braid two or more yarns together in the braiding fabric production procedure. A classification of textile fibres, yarns, and fabric is shown in Table 2, in terms of their dimension, construction process, advantages and disadvantages.

There are four different ways to make textile fabric: weaving,

A nanocomposite is a material composed of at least one of the phases



Fig. 5. Textile fibre [83], yarn [84], and various textile fabrics (braided [85], nonwoven [86], woven (self-drawn), knitted [87]).

# Table 2

Classification of textile fibres, yarns, and fabrics in terms of their dimension	n, construction process, advantages, and disadvantages.
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Type of textiles			Constructions	Advantages	Disadvantages
Fibre (0D dimension)	Natural		Growing naturally	Easy availability, low cost, biodegradable, environment friendly	Low strength, heavy in weight, not durable, wrinkled structure
	Synthetic		Derived from chemicals	High strength and durability, lower moisture content, smooth surface	Cost effective, processing challenges, non- eco friendly
Yarn (1D dimension)	Spun		Made by twisting of the staple fibres (fibres having short length)	Low cost	Low strength, less softness, more hairiness
	Filament	Mono Multi	Consists of a single strand Consists of two or more strands	High strength, softer, less hairiness	Cost effective, hard to manufacture
Woven fabric (plain, twill, satin,	2D fabric		Made by combining two orthogonal yarns	Easy processing, lower fibre damage, higher dimensional stability	Heavy in weight, less productivity, rigid in structure
sateen, etc.)	3D fabric		Made by combining three orthogonal yarns	Light in weight, high productivity, softer, higher dimensional stability	Higher fibre damage tendency, complexity in manufacturing
Knitted Fabric (plain, rib, purl, interlock)	2D fabric 3D fabric		Made by interloping of the yarns	Good flexibility, soft to touch, higher extensibility	Lower dimensional stability, cost effective, shrinks easily, tendency to curl at the edges
Braided fabric (bi-axial and tri-axia	1)		Made by intwining of the yarns	Higher strength than fibre and yarn, low cost, easy processability, good for high performance applications	Only narrow fabric can develop, it can be stretched in one direction only
Nonwoven (electrospun, needle bonding, water entar bonding, etc.)	punch, spun 1glement, air		Made by entanglement of the fibres	Flexible, softer, low cost, light weight, higher specific surface area of the fibres (electrospun fibres)	Lower dimensional stability, low strength, low durability,

(matrix or reinforcing) having less than 100 nm dimensions [88]. Over the past years, the textile industry has become one of the emerging adopters of nanotechnology with numerous nanocomposite textile-based products available in the market [89,90]. Nanotechnology has found substantial application in textile manufacturing, particularly when incorporating nanofillers into textile materials, resulting in what is known as nanocomposite textiles. Compared to conventional textiles, nanocomposite textiles exhibit enhanced functional properties. For example, their piezoelectric or triboelectric characteristics are superior to those of regular textiles. In this review, we will delve into the details of nanocomposite textile-based piezoelectric and triboelectric nanogenerators.



**Fig. 6.** (a) (i) Digital photos of the PVDF/BT nanocomposite fibre based PENG [91], (ii) a bobbing of the PVDF/BT nanocomposite fibres [91]; (b) An output voltage and a output power generated by the PVDF/BT nanocomposite filament based PENG [91]; (c) The coil structure [94] and (d) Output voltage of the PVDF, PVDF/BT, PVDF/rGO, and PVDF/rGO/BT nanocomposite filaments [94], (e) Schematic illustration of the PVDF/KNN nanocomposite filament based PENG [39]. All essential copyrights and permissions received.

# 4. Nanocomposite textile based piezoelectric nanogenerators (NT-PENGs)

Significant research has already been conducted on textile-based and nanocomposite textile-based piezoelectric nanogenerators. In the case of conventional textiles, a poling method is typically required to achieve desirable piezoelectric properties. However, in nanocomposite textiles, the presence of nanofillers within the polymer matrix acts as a nucleating agent, resulting in higher piezoelectric properties compared to normal textiles. Consequently, there is sometimes no need for additional poling arrangements as required for conventional textiles. Alternatively, direct incorporation of piezoelectric fillers such as KNN, BT, PZT, etc., into the polymer matrix can be employed to enhance the piezoelectric properties. Nanocomposite textile-based piezoelectric nanogenerators can be fabricated in various forms, including single fibers or yarns, fabrics, and electrospun nanofibrous webs.

# 4.1. Nanocomposite single fibre or yarn based piezoelectric nanogenerators (NSF-PENGs or NY-PENGs)

In general, piezoelectric polymer-based single fibers or yarns in their pristine form exhibit lower piezoelectric performance. Moreover, pure polymer-based fibres or varns have poor mechanical properties. To overcome these limitations, researchers have attempted to develop nanocomposite-based piezoelectric fibers or varns. Although nanocomposite-based fibres or yarns possess superior properties, to date, only a limited number of studies have been conducted on nanocomposite single fibre or varn based piezoelectric nanogenerators. One such example is the work by Mokhtari and co-workers [91], who reported a PVDF/BT melt spun nanocomposite filament based piezoelectric nanogenerator (Fig. 6a). In this study, pure PVDF and PVDF/BT nanocomposite filaments were produced using a two-stage process: solution casting followed by melt spinning. The effect of BT nanoparticles on the piezoelectric properties of PVDF polymer was analyzed by varying the concentration of BT nanoparticles (0, 5, 10, and 20 wt%). It was observed that the fraction of  $\beta$ -phase increased in the case of PVDF/BT nanocomposite filaments (β-phase fraction-98%) compared to neat PVDF filament ( $\beta$ -phase fraction-51%). This increase can be attributed to the nucleation of  $\beta$ -crystalline nucleating effect of the BT nanoparticles in PVDF polymer. However, the  $\beta$ -phase fraction in the PVDF polymer is decreased beyond 10 wt% of BT nanoparticles. At higher concentration, BT nanoparticles get agglomerated and therefore, improper interaction takes place in between BT nanoparticle and PVDF polymer [92,93]. The PVDF/BT nanocomposite filament also showed better mechanical properties (tensile strength- 170% and young modulus- 130% higher than pure PVDF filament) compared to pure PVDF filament. PVDF/BT nanocomposite filament is a more suitable candidate for developing the piezoelectric nanogenerator due to higher dielectric constant (~200), lower cost, and ecofriendly nature of the BT nanoparticles. Further, it was found that the PVDF/BT nanocomposite filament based piezoelectric nanogenerator had an output voltage of 1.1 V which is comparatively high compared to the pure PVDF filament based piezoelectric nanogenerator (output voltage ~0.48 V). In addition, the nanocomposite filament based piezoelectric nanogenerator showed a maximum power of  $0.21\,\mu\text{W}$  with an external resistance of 400 k $\Omega$  (Fig. 6b). It was also found that the PVDF/10%BT nanocomposite filament based piezoelectric nanogenerator has shown higher output voltage than PVDF/20% BT. However, the piezoelectric voltage constant (g<sub>33</sub>) is lower in the case of PVDF/10% BT based PENG as compared to PVDF/20% BT based PENG. Finally, to prove practical ability of this as prepared PENG, capacitors having different capacities were charged. Later, in the year 2021, the same research group [94] has also reported the piezoelectric properties of a highly stretchable piezoelectric nanogenerator composed of PVDF/rGO/BT melt spun nanocomposite filament (Fig. 6c). For this study, pure PVDF, PVDF/BT, PVDF/rGO/ and PVDF/rGO/BT nanocomposite filaments were prepared

by a melt spinning process. During melt spinning, a twist of 12000 TPM (twist per meter) was inserted into the melt spun filament until a coil formed completely along the entire length of the filament. It was found that the  $\beta$ -phase fraction is increased in the case of PVDF/rGO/BT nanocomposite filament as compared to other counter parts. This is because of the nucleation effect of rGO which causes delocalization of  $\pi$ -electrons with the remaining oxygen groups in rGO which interact with the CH<sub>2</sub>, and CF<sub>2</sub> functional groups of the PVDF polymer chains. It was also found that the pure PVDF, PVDF/rGO/ and PVDF/BT nanocomposite filament-based PENG showed output voltages of 0.3 V, 0.4 V and 0.7 V, respectively (Fig. 6d). The PVDF/BT based PENG showed higher output than PVDF/rGO - this may be due to the rGO sheets having many free charges which did not find dipoles from the PVDF polymer. On the other hand, the developed PVDF/rGO/BT nanocomposite filament-based PENG showed an output voltage of  $\sim 1.2$  V, an output current of ~2.5 nA, and a power density of ~ 0.42  $\mu$ W/cm<sup>3</sup>, respectively. The optimized PENG device was employed as motion and strain sensors to detect the human finger bending angle and movements. The sensor output voltage was calibrated using different forces (0.5 - 10 N)applied during the sample compression test. Although BT has advantages in boosting the electrical performance of the nanocomposite based piezoelectric nanogenerator, BT has limited dielectric constant, piezoelectric constant  $(d_{33})$  and curie temperature as compared to the other ceramic based piezoelectric materials like PZT and KNN.

KNN is one of the most desirable piezoceramic materials because of its higher piezoelectric constant (150-500 pC/N), higher curie temperature (>400 °C), higher dielectric constant, and its ecofriendly nature [5,95]. Therefore, it is expected that KNN nanocomposite filament based piezoelectric nanogenerators will show better piezoelectric properties as compared to BT based piezoelectric nanogenerators. In a study, Bairagi and colleagues [39] prepared a piezoelectric nanogenerator using KNN nanorods incorporated PVDF polymer nanocomposite melt spun filaments (Fig. 6e). Herein PVDF/KNN nanocomposite filaments were developed by the melt spinning process with different concentrations of KNN nanorods (0%, 2%, 4% and 6%). The as prepared filaments were characterized using the FTIR and XRD to find out the structural characteristics of the filaments. It was observed that the beta fraction value for the nanocomposite filaments increases with increase in the KNN nanorod concentration (beta fraction for PVDF/0% KNN NRs is 9% while 26% for the PVDF/4% KNN NRs). This is due to the beta nucleating effect of the KNN nanorods in PVDF polymer. However, the beta fraction value reduces down for PVDF/6% KNN NRs due to the agglomeration of the KNN nanorods at higher concentration. The same trend was also found for the XRD analysis of the developed nanocomposite filaments. The dielectric constant of the filaments was also analyzed, and it was found that the PVDF/4% KNN NRs based nanocomposite filament showed 175 dielectric constant which is comparatively higher than the other filaments (pure PVDF-73, PVDF/2% KNN NRs-134, and PVDF/6% KNN NRs-131, respectively). Finally, the piezoelectric responses were evaluated with a digital oscilloscope using hand-tapping on the respective PENG device and interestingly, it was found that the PVDF/KNN nanocomposite filament based piezoelectric nanogenerator showed better electrical performance (output voltage ~3.7 V (Fig. 6 f) and output current~0.326 µA, respectively) compared to the pure PVDF filament based piezoelectric nanogenerator (output voltage ~1.9 V). The electrical performance of the PVDF/KNN nanocomposite filament-based PENG is higher compared to PVDF/BT based PENG.

Later in the same year (2020), Chowdhury and co-workers [96] prepared a PVDF/PU/rGO nanocomposite filament based piezoelectric nanogenerator. In this study, PVDF/PU/rGO nanocomposite filaments were engineered by varying the concentration of rGO from 0 wt% to 0.5 wt%. The FTIR characterization was carried out to find out the relative beta fraction value of the as prepared filaments and it was found that the beta fraction value is higher in the case of PVDF/PU/0.1%rGO (66%) nanocomposite filaments compared to other counterparts (for 0%

rGO, beta fraction is 40%, for 0.3% rGO, beta fraction is 38% and for 0.5% rGO, beta fraction is 39%, respectively). The relative beta fraction is high for the PVDF/PU filaments having lower concentration of rGO compared to higher rGO incorporated PVDF/PU filaments. This is because, at lower concentration of rGO, a suitable interaction takes place between the PVDF and PU polymer chains which results in a high relative beta fraction, but at higher concentration of rGO, that interaction between the PVDF and PU polymer chains gets restricted [97]. The PVDF/PU/0.3% rGO nanocomposite filament-based PENG had an open circuit voltage and a short circuit current of 0.349 V and 0.294  $\mu$ A, respectively while only the PVDF/PU filament-based PENG showed an open circuit voltage and a short circuit current of 0.318 V and 0.172  $\mu$ A, respectively. Moreover, 0.3% rGO incorporated PVDF/PU based PENG showed a power density of 66.39 mW/m<sup>2</sup> under a constant load and frequency of 3.82 N and 2.5 Hz, respectively.

All these above-mentioned studies provide a clear idea about nanocomposite fibre or yarn based piezoelectric nanogenerators. It can be found that the nanocomposite fibre/yarn based piezoelectric nanogenerators have shown better piezoelectric performance as compared to pristine polymer based piezoelectric nanogenerators. This is due to the additional piezoelectric characteristic of the nano fillers and nucleation effect of the nano fillers in polymer matrix. In addition, nanocomposite fibre/yarn showed higher mechanical strength compared to pristine polymeric fibre/yarn. However, to prepare nanocomposite based piezoelectric fibre/yarn one of the main challenges is the uniform distribution of filler in polymer matrix and therefore, to mitigate this problem, more research work needs to be explored. Although nanocomposite filament based PENGs are very flexible in nature and easy to process, they have very low piezoelectric outputs. Hence, the nanocomposite filament based PENGs would not be a suitable power source for wearable and portable electronics. Therefore, textile fabrics (woven, knitted, and braided) can be prepared by using these nanocomposite filaments to get enough piezoelectric signals.

# 4.2. Nano composite fabric based piezoelectric nanogenerators (NF-PENGs)

The piezoelectric outputs of nanocomposite single fibre or yarn



**Fig. 7.** (a) (i) Digital photos of the PVDF/BT nanocomposite fabric based PENG [91], (ii) an output voltage signal generated by PENG [91], and (iii) the digital photos of the PVDF/BT nanocomposite fabric based PENG (an output voltage signal at the inset) [91]; (b) Schematic diagram of the PVC/BT nanocomposite fabric based PENG [40], (ii) & (iii) an output voltage and current generated by the as fabricated PENG [40]; (c) The schematic illustration of the ZnO NWs nanocomposite fabric based PENG [99]; (d) The ZnO NWs incorporated nanocomposite based PENG (i) schematic diagram of the PENG [100], and (ii) an output voltage generated by the PENG in forward and reverse directions [100]. All essential copyrights and permissions received.

based PENGs, are very low due to limited number and smaller active area. Therefore, in order to enhance piezoelectric outputs, one encouraging approach is to combine multiple nanocomposite fibres or yarns into the 2D or 3D textile structure by different textile manufacturing methods (weaving, knitting, nonwoven, etc.). The nanocomposite fabric based PENGs are a more suitable candidate than nanocomposite fibres or yarn based PENGs, in terms of their higher energy conversion efficacy. Moreover, nanocomposite fabric based PENGs can also be integrated in textile structures for wearable applications. Hence, nanocomposite fabric based PENGs are a more suitable candidate to supply power to small portable and electronic devices. To date, many research works have already been done on nanocomposite fabric based PENGs. For a typical example, Mokhtari and co-workers [98] prepared a piezoelectric nanogenerator composed of nanocomposite braided yarn. The nanocomposite braided yarn was engineered by intertwining of the melt spun piezoelectric PVDF fibres and conductive silver coated nylon yarns. The PVDF fibres were braided around the silver coated nylon yarns to form the inner electrodes, while the whole assembly was braided further with the silver coated nylon yarns to form the outer electrode. This formed a triaxial structure that made the energy harvester quite robust and fatigue resistant. These energy harvesters under compressive or bending forces generated an output voltage of 380 mV with a power density of 29.62  $\mu$ Wcm<sup>-3</sup>.

In another study, Mokhtari and colleagues [91] also studied the piezoelectric properties of a nanocomposite fabric based piezoelectric nanogenerator (Fig. 7a). The nanogenerator was fabricated using the PVDF/BT nanocomposite filaments prepared by the melt spinning process. It was observed that, at a higher loading of the barium titanate nanoparticles, agglomeration of the nanoparticles took place. Moreover, at higher loading of nanoparticles, the nanocomposite preparation became difficult owing to the reduced mobility of the polymer chains. The electroactive phase content of PVDF increased with the increase in the barium titanate nanoparticle loading owing to the increased interaction between the filler and polymer. However, beyond a certain concentration, the segmental mobility of the polymer chains is restricted; that results in a decrease in the electroactive phase content. The fibres were then triaxially braided to form energy harvesters. The braided fibres were then fabricated into knitted structures to form wearable energy harvesters. These energy harvesters generated up to 4 V output voltage with a power density of 87  $\mu W cm^{-3}$  and excellent sensitivity. Likewise, Zhang et al. [40] fabricated a 2D nanocomposite fabric-based PENG composed of BaTiO<sub>3</sub> NWs/PVC nanocomposite varns, cotton varns and Cu wires (Fig. 7b). Here, BaTiO<sub>3</sub> NWs/PVC nanocomposite yarns were used as the piezoelectric component whereas cotton yarns for protecting short circuit between the Cu wires as well as to maintain wearability of the PENG device. To fabricate 2D fabric based piezoelectric nanogenerator, BaTiO<sub>3</sub> NWs/PVC nanocomposite yarns were used in the warp direction (length wise) while conductive Cu wires used in the weft direction (width wise). On the other hand, cotton yarns were utilized as the insulating spacers between the conductive Cu wires to protect from short circuit between Cu wires. The fabric was prepared by hand knitting using these above-mentioned yarns. For getting better piezoelectric response the prepared 2D fabric was poled by applying an external electric field of 4 kV/mm at a temperature of 20 °C for 20 h. Furthermore, for comparison purposes, a pure PVC polymer-based fabric was also developed. Finally, the as prepared piezoelectric nanogenerator was attached onto an elbow pad to harvest biomechanical energy from the human body movement. And interestingly, it was found that the fabricated piezoelectric nanogenerator showed an output voltage and short circuit current of 1.9 V and 24 nA, respectively, against the human hand bending motion.

In wearable piezoelectric nanogenerators (PENGs), biocompatibility is a crucial requirement. In this context, zinc oxide (ZnO) is an appealing alternative to lead zirconate titanate (PZT) or other filler materials due to its biocompatible nature. It has good mechanical and piezoelectric coupled semiconducting properties, which results in easy conversion of

body movement mechanical energy into electrical energy. Therefore, ZnO based nanocomposite fabrics have been studied widely for piezoelectric performance. One such example in 2019 (Rafique and coworkers [99]) reported a textile based piezoelectric nanogenerator which was developed by growing silver doped ZnO nanorods on cotton fabric (Fig. 7c). These nanorods were grown hydrothermally on the cotton substrate. The morphological studies of the substrate revealed good adhesion of the Ag-doped ZnO nanorods on cotton due to hydrogen bonding between them and the hydroxyl groups of the cotton fabric. Structural analysis of the hydrothermally grown Ag-doped ZnO nanorods revealed the replacement of Zn<sup>2+</sup>ions with the Ag<sup>+</sup> ions, evident from the shift in peaks towards higher degree for the doped nanorods compared to pristine nanorods. The improved piezoelectric performance of the Ag-ZnO nanorods on cotton fabric compared to pristine ZnO nanorods on cotton fabric was mainly due to the reduction in free charge carrier concentration due to doping with silver. In addition, the piezoelectric performance of the developed piezoelectric nanogenerator was carried out under a cyclic applying load of 3 kgf. The Ag-ZnO/cotton based piezoelectric nanogenerator showed an output voltage of 6.85 V and a short circuit 3.42 µA while the undoped ZnO/cotton based piezoelectric nanogenerator showed an output voltage and a short circuit current of 2.28 V and 1.16 µA, respectively. Therefore, the output voltage and current of the Ag doped ZnO grown cotton based piezoelectric nanogenerator is almost 3 times higher than the undoped ZnO coated cotton based piezoelectric nanogenerator. Moreover, the Ag doped ZnO based piezoelectric nanogenerator also showed a maximum power density of 1.45 mW/cm<sup>2</sup> at a load resistance of 31 M $\Omega$  and they also found that capacitive reactance also affects the charging capability of the PENG. Similarly, in another study, a nanocomposite fabric-based PENG was developed using two types of fibres, one was ZnO NWs grown fibres and other is ZnO NWs grown covered with the palladium (Pd) (Fig. 7d). The as-fabricated PENG could generate output voltage and current of 3 mV and 17 pA, respectively, when the PENG is slid by a linear motor [100]. A summary of some of the nanocomposite single fiber (NSF) or yarn (NY) and the nanocomposite fabric (NF) based piezoelectric nanogenerators (PENGs) is shown in Table 3.

From these above studies on nanocomposite textile fabric based piezoelectric nanogenerators, it can be seen that few well known piezoelectric materials such as PVDF, BaTiO<sub>3</sub>, and ZnO, are utilized to develop the nanocomposite textile fabric based PENGs. Often ZnO, and BaTiO<sub>3</sub>, either directly grow on the textile fabric or incorporate into the polymer matrix whereas PVDF is usually fabricated into the filament mostly by the melt spinning method. It can also be observed that the nanocomposite fabric based PENGs have shown better piezoelectric properties and durability as compared to the pristine fabric based PENGs. Although nanocomposite textile fabric based PENGs show better piezoelectric properties, still systematic studies need to be done.

To prepare nanocomposite fibre/yarn and fabric based piezoelectric materials the most important step is the poling method. Poling is a method where all randomly oriented dipoles (molecular dipoles for the polymer and ionic dipoles for the inorganic piezoelectric materials) in the nanocomposite get aligned in a particular direction by application of an external electric field. The poled piezoelectric properties. Therefore, it can be concluded that, to prepare nanocomposite piezoelectric fibre/ yarn or fabric, additional poling arrangement need to be added. This additional arrangement will increase the overall cost of the nanogenerator. To solve these problems, the electrospinning method comes into the picture to prepare electrospun nanocomposite piezoelectric materials. The electrospinning method for preparing electrospun nanocomposite based PENGs is discussed in detail in the next section.

#### Table 3

Summary of the NSF-PENGs /NY-TENGs and NF-PENGs.

Materials	Textile structure	Electrd.	wt%	Voc (V)	Isc (μA)	P <sub>d</sub> <sup>max</sup> (mW/m <sup>2</sup> )	Applications	Ref.
PVDF/BT	Melt spun filament	Al	10	1.1	-	2.1	Energy harvesting	[91]
PVDF/rGO/BT	Melt spun filament	Al	0.5 & 10	1.2	0.0025	36	Energy harvesting and LEDs glowing	[94]
PVDF/KNN NRs	Melt spun filament	Al	4	3.7	0.000326	-	Energy harvesting	[101]
PVDF/Ag-Nylon	Braided fabric	Ag	10	0.38	1750	1.6	Energy harvesting and capacitor charging	[98]
PVC/BT NWs	Knitted fabric	Cu	-	1.9	24	0.1	Energy harvesting and motion sensing	[40]
Cotton/Ag-ZnO	Woven fabric	Cu	-	6.85	3.42	14.5	Energy harvesting and capacitor charging	[99]

# 4.3. Nanocomposite electrospun web based piezoelectric nanogenerators (NEW-PENGs)

Electrospinning is a versatile and important process used to produce nanofibers. It involves the application of a high-voltage electric field to a polymer solution or melt, causing the formation of ultrafine fibers with diameters typically ranging from a few nanometers to several micrometers of a nonwoven like web. Here, the electrospinning process is described briefly. A flowing dielectric polymer solution bends as it leaves a needle or spinneret under the influence of an electric field to create a cone (known as a Taylor cone) from which small droplets lift off. The droplets lifting off will combine into a thin liquid jet if the voltage supplied is high enough. The viscosity and electrical resistivity of the polymer solution affect how the Taylor cone and thin liquid jet develop. While the jet travels to the collector as fibre jets for high viscosity solutions, it breaks up into droplets for low viscosity solutions. The narrow jets will form fibres as long as the solvent carrying the polymer evaporates quickly enough. Following initiation from the cone, the jets move chaotically or exhibit bending instability before being field-directed towards the grounded collector that is negatively charged and collects the charged fibres [102]. Electrospinning can be affected by different parameters which play a significant role in deciding the morphology, diameter, and formation and electromechanical properties of the resultant nanofibers or webs. The factors can be divided into solution parameters, process parameters and ambient condition. Among them, solution parameters such as viscosity, molecular weight and



**Fig. 8.** (a) Schematic illustration of the PVDF/PZT electrospun nanocomposite fibre based PENG (inset showing morphology of the PVDF/PZT nanofibres) [121]; (b) Schematic diagram of the SMPU/PZT electrospun nanocomposite fibre based PENG and a digital image of the developed PENG as depicted at the right hand side [122]; (c) (i) The P(VDF-TrFE)/PZN-PZT electrospun nanocomposite based PENG [123], (ii) & (iii) the output voltage and current generated by the developed PENG [123]; (d) (i) A diagram of the PVDF/KNN nanofibre based PENG [125]; (ii) an output voltage generated by the PENG [125]; (e) Schematic diagram of the PVDF/SM-KNN electrospun nanocomposite fibre based PENG along with its output voltage graph [126]. All essential copyrights and permissions received.

conductivity and major process parameters including applied electric field, flow rate and tip to collector distance are primary ones. Besides these, the morphology and diameter are also affected by ambient conditions such as temperature and humidity [103–107].

One of the highly promising research areas in recent years is the development of piezoelectric nanogenerators based on electrospun nanofibers. These nanogenerators offer several advantages over meltspun materials and solution cast films, primarily due to the in-situ poling process integrated into the electrospinning procedure. As a result, they exhibit greater output signals and eliminate the need for additional poling arrangements. During electrospinning, the dipoles within the nanofibers align themselves in a specific direction in response to the external electric field applied during the process. This aligning of dipoles ensures that the nanofibers possess a predetermined polarization, eliminating the requirement for supplementary poling apparatus [108–111]. Additionally, compared to solution-cast films or melt-spun filaments, electrospun nanofibre-based piezoelectric nanogenerators ought to be lighter, more flexible, and less expensive. Numerous researchers all over the world are using electrospun nanofibres as piezoelectric materials as a result of these benefits. However, piezoelectric pure polymer based electrospun nanofibres are not sufficient to harvest energy for self-powering applications due to their lower piezoelectric signals. To overcome drawbacks with the pure polymer based electrospun nanofibres, researchers have tried to develop electrospun nanocomposite fibres. In general, polymer (PVDF, PVDF-TrFE, PVDF-HFP, PP, Nylon11, PLA, and so on) [112-116] material has been used as a matrix whereas inorganic metal oxide (ZnO, KNN, PZT, and many more) [117–120] has been utilized as a filler component in the nanocomposite based piezoelectric material. And it is also expected that output signals of the nanocomposite electrospun web based PENGs will be high as compared to nanocomposite single fibres or yarns and fabric based PENGs. However, the durability of the electrospun nanofibrous webs still needs to be much improved. There are several research works on nanocomposite electrospun web based PENGs. Koc and colleagues [121] reported on PVDF/PZT electrospun nanocomposite fibres based flexible piezoelectric nanogenerators (Fig. 8a). Herein, PVDF/PZT electrospun nanocomposite fibres were developed by the electrospinning process where different concentrations (0 wt%, 10 wt%, 20 wt%, and 30 wt%) of PZT filler were incorporated into the PVDF polymer. The piezoelectric outputs for PVDF/PZT and pure PVDF electrospun fibres were evaluated by connecting a load in parallel to the piezoelectric device and also testing without connecting the load. It was found that the PVDF/PZT nanocomposite-based PENG had a more effective voltage of 35.81% more than the pure PVDF electrospun fibres-based PENG under a resistive load of 1 MΩ. Whereas, without resistive load, the PVDF/PZT based PENG only showed an effective voltage of 11.73% above the pure PVDF based PENG. Furthermore, it was also found that the PVDF/10% PZT nanocomposite-based PENG had a higher power output (6.35 µW) than the pure PVDF based PENG (3.44  $\mu$ W). This is due to the higher piezoelectric characteristics of the PZT. In another study, Guan et al. [122] also reported a piezoelectric nanogenerator based on the PZT incorporated shape memory polyurethane (SMPU) electrospun nanocomposite fibres (Fig. 8b). Two types of electrospun nanocomposites were prepared: random oriented and aligned nanocomposite fibres. Randomly oriented and aligned nanofibres were developed intentionally to observe the effect of fibre orientation upon the mechanical and piezoelectric properties of the resultant SMPU/PZT nanocomposite. Interestingly, it was found that the aligned (0  $^{\circ}$  aligned means nanocomposite fibres parallel to the tensile testing direction) nanocomposite fibres showed better mechanical and piezoelectric properties as compared to randomly oriented nanocomposite fibres. Mechanical properties of the aligned nanocomposite fibres are better due to alignment of the nanofibres along the load bearing direction. Specifically, the aligned nanocomposite fibres based piezoelectric nanogenerator had an output voltage of 421 mV which is 5.4 times higher than randomly oriented nanocomposite fibres based piezoelectric nanogenerator

(output voltage for the randomly oriented fibres based piezoelectric nanogenerator is 65.6 mV). This is because of the higher density and lower surface area of the aligned nanocomposite fibres. Likewise, Liu and colleagues [123] reported the piezoelectric outputs for a PZN-PZT (0.4Pb (Zn<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>-0.6Pb(Zr0.5Ti0.5)O<sub>3</sub>) nanoparticles incorporated P(VDF-TrFE) polymers electrospun nanocomposite based piezoelectric nanogenerator (Fig. 8c(i)). To develop electrospun nanocomposite fibres, various concentrations (0 wt%, 5 wt%, 10 wt%, and 20 wt%) of PZN-PZT fillers (diameter of 54 nm) were loaded into the P(VDF-TrFE) polymer during the electrospinning process. It can be seen from Fig. 8c(ii) and (iii), that the piezoelectric output signals (output voltage and current) are increasing with increase in the concentration of the PZN-PZT fillers - this is due to higher piezoelectric properties of PZN-PZT fillers compared to P(VDF-TrFE) polymer. The P (VDF-TrFE)/20% PZN-PZT nanocomposite fibre based piezoelectric nanogenerator had an output voltage and a short circuit current of 3.4 V and 240 nA, respectively, which is much higher than the pure P (VDF-TrFE) polymer based piezoelectric nanogenerator. Therefore, it can be concluded from this section that PZT is one of the most lucrative piezoelectric fillers for getting better piezoelectric output from electrospun nanocomposite based piezoelectric nanogenerators. This is because PZT has very good piezoelectric properties. However, in recent years, there is a tendency to avoid PZT in piezoelectric applications since it contains more than 60% lead which is non-eco-friendly.

To address the issue of lead toxicity, various lead-free piezoelectric materials such as KNN, ZnO, BT, etc., have been employed to develop environmentally friendly electrospun nanocomposite fibers for piezoelectric applications. Among these lead-free piezoelectric fillers, KNN stands out as a promising material due to its high piezoelectric constant and Curie temperature compared to other alternatives. In 2018, Teka and co-authors [124] reported a flexible, lightweight piezoelectric nanogenerator based on the PVDF/KNN electrospun nanocomposite fibres. Here, pure PVDF and PVDF/5%KNN nanocomposite fibres were prepared by an electrospinning process. It was found that the PVDF/KNN nanocomposite fibres based piezoelectric nanogenerator showed an output voltage of 1.9 V, while an output voltage in the range of 50-100 mV was reported for the pure PVDF based piezoelectric nanogenerator. This is due to the  $\beta$  nucleating effect of KNN nanostructures in PVDF polymer. Later, in the year 2019, Bairagi and colleagues [125] also studied the piezoelectric properties of the PVDF/KNN electrospun nanocomposite fibre-based piezoelectric materials (Fig. 8d (i)). Interestingly, it was found that the PVDF/3% KNN NRs nanocomposite based piezoelectric nanogenerator showed a maximum output voltage and current (17.5 V and 0.522 µA, respectively) (Fig. 8d (ii)), whereas PVDF/0% KNN NRs, PVDF/1% KNN NRs, and PVDF/5% KNN NRs based piezoelectric nanogenerators have shown an output voltage of 0.5 V, 1.9 V, 2.4 V and a short circuit current of 0.089 µA,  $0.109 \,\mu\text{A}, 0.197 \,\mu\text{A}$ . Therefore, it can be seen that the output voltage and current was increased with increase in the KNN NRs concentration upto a certain limit (i.e 3% KNN NRs) which could be attributed to the  $\beta$ nucleating effect of KNN NRs in PVDF polymer and in-situ poling of PVDF and KNN NRs during the electrospinning method. After a certain limit, the piezoelectric output signals deteriorate because, at higher concentration, KNN NRs get agglomerated and choke the needle tip during the electrospinning method for which a much reduced number of KNN NRs are expected to reach into PVDF electrospun fibres. Further, to solve the agglomeration problem of the KNN NRs, Bairagi and co-workers [126] recently reported a surface modified KNN NRs (SM-KNN NRs) incorporated PVDF electrospun nanocomposite based piezoelectric nanogenerator (Fig. 8e). Here, as synthesized KNN NRs were modified using silane coupling agent (3-aminopropyltriethoxy silane). Herein, different concentrations (0-5%) of SM-KNN NRs were utilized to develop PVDF/SM-KNN NRs based electrospun nanocomposite fibres. The 3% SM-KNN NRs/PVDF nanocomposite fibre-based piezoelectric nanogenerator had an output voltage and a short circuit 21 V (Fig. 8e) and 22 µA, respectively, which is higher compared to other counterparts (0.5 V for P-PVDF, 18 V for PVDF/1% SM-KNN NRs, and 2.5 V for PVDF/5% SM-KNN NRs). Therefore, it can be concluded from these above studies that, after surface modification of KNN NRs, there is a small increment in piezoelectric outputs for the SM-KNN NRs incorporated PVDF polymer nanocomposite fibres based PENG compared to the untreated KNN NRs based PENG. Although KNN is one of the most comparable piezoceramic materials, it has various drawbacks including lower stability at higher temperatures, it can show better piezoelectric properties at its morphotopic phase boundary (MPB), and it has a low piezoelectric constant i.e 90 pC/N [63,127].

ZnO has also been explored extensively in making piezoelectric nanocomposite materials [128]. ZnO nanomaterials can be synthesized simply by the hydrothermal method. The synthesis of ZnO is quite easy as compared to KNN and PZT. It has suitable piezoelectric as well as semiconducting properties, which make it a suitable candidate for piezoelectric applications [129]. However, dispersion of ZnO nanomaterials in polymer solution is a challenging task to obtain uniform distributed ZnO throughout the polymer matrix. Therefore, doping or using other additional dispersing agents with ZnO nanomaterials is a must to get uniform distribution of ZnO nanomaterials in the polymer matrix [130]. The piezoelectric nanocomposite having uniform ZnO nanomaterial distribution could exhibit a better piezoelectric property. One such example, where a piezoelectric nanogenerator was fabricated from electrospun membranes of PVDF-HFP combined with cellulose nanocrystals and Fe-doped nano ZnO is shown in Fig. 9a. The Fe-ZnO nanostars were synthesized hydrothermally while the cellulose nanocrystals were prepared through hydrolysis reactions. Electrospinning helps to align the domains of the filler material in the nanocomposite material in a specific direction that helps to enhance the overall

piezoelectric performance of the material. The prepared nanogenerator produces different voltages under various mechanical stimulations: 12 V upon mechanical vibration, 5.5 V for elbow movement and 1.1 V upon cloth folding movements. It also had the ability to harvest energy from ultrasonic waves, which proves its efficacy in energy harvesting from various energy sources. Such type of piezoelectric nanogenerators can be incorporated into wearables for energy harvesting purposes [130]. Likewise, in another study, the PVDF-TrFE/ZnO/exfoliated graphene oxide (EGO) electrospun nanocomposite fibres were utilized to develop a flexible piezoelectric nanogenerator (PENG) [131] (Fig. 9b). The as prepared PENG, using hybrid nanocomposite fibres (PVDF-TrFE/ZnO/EGO) exhibited better piezoelectric performance compared to pristine PVDF-TrFE and PVDF-TrFE/ZnO based PENGs. The output voltage and current were found to be  $\sim 0.40$  V &  $\sim 0.23$  nA (for the hybrid nanocomposite-based PENG), ~0.20 V & ~0.18 nA (for the PVDF-TrFE/ZnO based PENG) and ~ 0.07 V & 0.12 nA, respectively, against an irregular finger tapping on the piezo devices. The hybrid nanocomposite-based PENG has shown better performance because of the higher  $\beta$  nucleating effect in PVDf-TrFE polymer in the presence of ZnO nanomaterials and EGO. The  $\beta$  nucleating action is limited in the case of PVDF-TrFE/ZnO nanocomposite as ZnO nanomaterials are agglomerated, while ZnO nanomaterials were distributed uniformly in the presence of EGO. Therefore, a uniform distribution of the nano filler materials in polymer matrix is a promising requirement to obtain better energy conversion efficiency from the nanocomposite-based PENG. Similarly, Parangusan [132] prepared a piezoelectric nanogenerator based on cobalt- ZnO incorporated PVDF-HFP electrospun nanocomposite fibres (Fig. 9c). It was found that the nanocomposite fibres had a dielectric constant value of 38, which is comparatively higher than



**Fig. 9.** (a) (i) P(VDF-HFP)/Fe-ZnO electrospun nanocomposite fibre [130], (ii) & (iii) P(VDF-HFP)/Fe-ZnO electrospun nanocomposite fibre-based PENG [130]; (b) (i) An electrospinning setup [131], (ii) PVDF-TrFE/ZnO/GO nanocomposite fibre deposited on PDMS electrode [131], (iii) & (iv) fabricated PVDF-TrFE/ZnO/GO nanocomposite fibre based PENG [132]; (c) (i) Schematic illustration of a PVDF-TrFE/Co-ZnO nanocomposite fibre based PENG [132]; (d) Schematic diagram of a PVDF-TrFE/BT nanocomposite fibre based PENG [132]; (d) Schematic diagram of a PVDF-TrFE/BT nanocomposite fibre based PENG [132]; (e) (i) Schematic diagram of a PVDF-TrFE/MWCNTs nanocomposite fibre based PENG [133], (ii) image of the developed PENG [133], (iii) the developed PENG under releasing mode [133] and (iv) the developed PENG under bending mode [133]. All essential copyrights and permissions received.

the dielectric constant of the pure PVDF-HFP polymer (i.e. 8). In addition, the output voltage of the PVDF-HFP/cobalt-ZnO based PENG was found to be 2.8 V, whereas output voltage of the pure polymer-based PENG is 120 mV. This is due to the  $\beta$  nucleating and charge induction characteristics of the cobalt-ZnO nanomaterials in PVDF-HFP polymer.

The durability of the electrospun nanofibre based PENG is an important factor. In general, pristine polymer based nanofibre has low mechanical strength. Therefore, making nanocomposite fibre could be a great approach to enhance the durability of PENGs. Although nanocomposite fibre based PENGs would show good durability, studies still need to be done. In another study, a piezoelectric nanogenerator was fabricated based on PVDF-TrFE/MWCNTs electrospun nanocomposite fibres [128] (Fig. 9d). The Young's modulus of pristine PVDF-TrFE polymer and PVDF-TrFE/MWCNTs nanocomposite fibres was found to be 34.36 MPa and 47.60 MPa, respectively. The nanocomposite fibres show higher Young's modulus because iso-directionally distributed CNTs and polymer chains have shared the stress while, for the pristine polymer, only polymer chains experience the stress. Along with this, the piezoelectric performance is also enhanced in the case of PVDF-TrFE/MWCNTs nanocomposite fibre-based PENG. The nanocomposite fibre-based PENG had an output voltage and current of 18.23 V and 2.14 µA, respectively, while pure PVDF-TrFE nanofibre based PENG had an output voltage and current of 8.68 V and 0.89 µA, respectively. The nanocomposite fibre-based PENG has shown an increasing trend of the output signals with increasing MWCNTs concentration up to a certain limit (3 wt%). Whereas, further increasing CNTs concentration actually drops the output signals. The reason is, at lower concentration of CNTs, the polar  $\beta$  phase formation takes place during the electrospinning process. In addition, CNTs can entrap charges injected in the electrospinning process at the interfaces. These entrapped charges will help to enhance polarization in the polymer matrix by inducing more dipoles and providing more inductive charges on the outside surface of the nanofibres. However, at higher concentration of CNTs, the entanglement of the CNTs weakens which results in lower crystallinity in PVDF-TrFE polymer. Although, some of the CNTs which are oriented coaxially will trap charges and will add more inductive charges on the nanofibre surface, with the agglomerated CNTs, the total inductive charges will be reduced as CNT will link each other and will neutralize a part of opposite inductive charges through the conductive path. Therefore, concentration of the conductive nano-filler in nanocomposite is an important parameter to get higher mechanical and functional properties.

Similarly, Siddiqui and colleagues [133] studied the manufacturing of piezoelectrically active nanofibers by the electrospinning process using a nanocomposite of barium titanate nanoparticles (BT NPs) implemented in poly(vinylidene fluoride-trifluoroethylene) (VDF-TrFE)) and this resulted in the successful fabrication of a strong, effective, flexible-lead free nanogenerator. A nanofiber PENG (NF-PENG) was fabricated by encompassing nanofibers in an elastomeric film (Fig. 9e). The fabricated device was demonstrated by the moment of footsteps. Packing of the nanofiber supported device (loaded with nf-PENG containing BT NPs of weight 15%) into a shoe generated 25 V during walking at a walking frequency of 0.6 Hz and the higher load of 600 N. Such nanofiber-supported device has the potential to charge a capacitor of 4.7  $\mu$ F after walking about seventy-two times. A strain sensor can function from such preserved charge without utilizing any power supply. Such excellent characteristics of the NF-PENG are primarily due to the automatically poled composite nanofibres. Moreover, these devices have a strong ability to protect nanofibres from mechanical damage. The applications of such devices comprise a wide area of prospect ranging from electronics that are wearable to aesthetics, and comfort related applications.

Yang and co-workers [134] prepared an aluminum nitride (AIN) incorporated PVDF-TrFE electrospun nanocomposite based flexible piezoelectric nanogenerator. The AIN/PVDF-TrFE electrospun nano-composite fibres were prepared by an electrospinning method by

varying the concentration of AIN from 0% to 0.2%. The peak intensity corresponding to ferroelectric  $\beta$  crystalline phase in PVDF-TrFE polymer was increased with increasing concentration of AIN from 0% to 0.1%. Peak intensity (corresponding to  $\beta$  crystalline phase) then decreased with further increase of the AIN concentration from 0.1% to 0.2%. This may be due to the agglomeration of AIN nanoparticles at higher concentration of the AIN. However, FTIR spectrums of the AIN/PVDF-TrFE nanocomposite fibres show no such significant changes when AIN was incorporated in PVDf-TrFE polymer. The influence of AIN on the piezoelectric properties of the nanocomposite fibres was further clarified by performing perpendicular and transverse piezoelectric characteristics of the AIN/PVDF-TrFE nanocomposite fibres. For the perpendicular piezoelectric measurement, the developed 0/1% AIN/PVDF-TrFE electrospun nanocomposite fibre-based piezoelectric nanogenerator showed an open circuit voltage and a short circuit current of 106 mV and 1.1 nA, respectively, under a 4 N load and 5 Hz frequency; whereas, for the transverse measurement, the same piezoelectric nanogenerator showed an open circuit voltage of 82.1 V and a maximum peak power density of 1.07  $\mu$ W/cm<sup>2</sup> at 12 mm displacement and 0.5 Hz frequency. Finally, the as prepared piezoelectric nanogenerator was used as a motion sensor and a pulse sensor.

From these above studies it can be seen that different inorganic piezoelectric fillers such as PZT, KNN, ZnO, and BT, etc are used to prepare electrospun nanocomposites whereas PVDF, and PVDF-TrFE are used as a polymer matrix for the same. PZT incorporated PVDF and PVDF-TrFE electrospun nanocomposite based PENGs have shown better piezoelectric properties compared to the pure polymer based PENGs. This is due to higher piezoelectric characteristics of PZT. On the other hand, ZnO, KNN and BT are also widely used to prepare nanocomposite electrospun web owing to their ecofriendly nature. In some studies, conductive fillers such as CNTs, MW-CNTs are also additionally used with the piezoelectric fillers (PZT, KNN, BT, etc) to prepare more efficient electrospun nanocomposite based PENGs. Although electrospun nanocomposite based PENGs show better piezoelectric properties, the lower durability and stability of the electrospun web limit their applications. However, these studies provide a clear direction and framework for future research work on the textile based piezoelectric nanogenerators. At the end, different electrospun nanocomposite fibres based piezoelectric nanogenerators (PENGs) are summarized in Table 4.

# 5. Nanocomposite textiles for triboelectric nanogenerators (NT-TENGs)

The textile based TENG (t-TENG) is being researched extensively for mechanical energy harvesting from different sources such as from human body movements, wind flows, rain drops, etc. Although the triboelectric performance of the t-TENG has been improved by introducing different approaches, it still lags significantly behind the performance of non-textile TENGs and considerable improvement is needed. The electrical performance of the t-TENG is mainly governed by the transfer of charges caused by contact or sliding of the triboelectric fabric/yarn/fibre materials and electrostatic induction of charges onto the electrodes. Two approaches have been explored to enhance the performance of triboelectric materials or textile-TENG (t-TENG). Firstly, the decoration or modification of the chemical composites or surface structure of the triboelectric materials has been investigated. This involves introducing additives, coatings, or functionalizing the material surfaces to optimize their triboelectric properties. Secondly, another approach focuses on designing a well-engineered structure for the triboelectric materials. By carefully designing the structure, such as the morphology, alignment, and configuration of the materials, the overall triboelectric performance can be significantly improved. These two approaches offer promising strategies to enhance the efficiency and reliability of triboelectric materials and t-TENG devices [139]. During triboelectrification, positive and negative charges result on the contacting surfaces. The generated charges may occupy a certain position

#### Table 4

Summary and comparison of the different electrospun nanocomposite fibre based PENGs.

Materials	Area (cm²)	Freq. (Hz)	Load (N)	Electrd.	<b>W</b> t%	Voc (V)	Isc (μA)	P <sup>max</sup> (mW/m <sup>2</sup> )	P <sup>max</sup> /N (mW/m <sup>2</sup> /N)	Applications	Ref.
PVDF/PZT P(VDF-TrFE)/ Pb(Zn <sub>1</sub> / 3 Nb <sub>2/3</sub> )O <sub>3</sub> –Pb (Zr0.5Ti0.5)O <sub>3</sub>	16 5	15 -	-	Al tape ITO coating	10 20	0.44 3.4	- 0.24	3.967 1.632	-	Energy harvesting Energy harvesting and capacitor charging	[121] [123]
PVDF/KNN nanostructures	6.25	-	-	Al tape	5	1.9	-	-	-	Energy harvesting	[124]
PVDF/KNN NRs	4	7	20	Al tape	3	17.5	0.522	22.8375	1.141875	Energy harvesting and LEDs glowing	[125]
PVDF/SM-KNN NRs	4	7	20	Al tape	3	21	22	1155	57.75	Energy harvesting and LEDs glowing	[126]
PVDF-HFP/CNC/Fe-ZnO	-	45	2.5	Ag coating	2 & 2	12	2.5	490	196	Energy harvesting	[130]
PVDF-TrFE/ZnO/EGO	-	-	-	Ti & Au coating	1&0.01	0.40	0.00023	-	-	Energy harvesting	[131]
PVDF-HFP/Co-ZnO	-	50	2.5	Al foils	2	2.8	-	-	-	Energy harvesting	[132]
PVDF-TrFE/MWCNTs	3	1	-	Cu foils	3	18.23	2.14	65.3	-	Energy harvesting and LEDs glowing	[128]
PVDF-TrFE/BT NPs	1	0.6	20	ITO coating	15	3.4	0.523	22.8	1.14	Energy harvesting and capacitor charging	[133]
PVDF-TrFE/AlN	1.5	5	4	Al tape	0.1	0.106	0.0011	10.7	2.675	Energy harvesting	[134]
PVDF/KNN/CNT	4	7	20	Al tape	3 & 0.1	23.24	9	522.9	26.145	Energy harvesting and LEDs glowing	[135]
PVDF/KNN/ZnO	4	7	20	Al tape	3 & 2	25	1.81	113.125	5.65625	Energy harvesting and LEDs glowing	[136]
PVDF/KNN-ZS	4	7	20	Al tape	3	25	2.11	131.875	6.59375	Energy harvesting and LEDs glowing	[127]
PAN/CuO	4	7	20	Al tape	0.5	5	0.172	2.15	0.1075	Energy harvesting	[137]
PVA/Casein	4	7	20	Al tape	-	20	37	1850	92.5	Energy harvesting and LEDs glowing	[138]

on the textile triboelectric layer causing new charges to face problems entering the triboelectric surface and therefore, the electrostatic induction process can be affected negatively. The porous nature of textiles is also likely to reduce the effectiveness of electrostatic induction (in the fabric-coated electrode case) when compared with induction through solid electrode-coated films. On the other hand, over time, accumulated charges on the surface of the triboelectric material can also decay due to drift or diffusion of the charges. To resolve these various problems, functional and optimized materials incorporated at the triboelectric layer is one approach that has been utilized to generate and maintain high surface charge density. In this context, nanocomposite textiles are a promising option for the textile triboelectric nanogenerator (t-TENG). In this section, nanocomposite textiles in the form of single fibre or yarns, fabric, and electrospun web, are discussed systematically [140].

# 5.1. Nanocomposite single fibre or yarn based triboelectric nanogenerators (NSF-TENGs or NY-TENGs)

Mechanical energy is one of the most available renewable energy sources as already mentioned earlier and it is often wasted abundantly in our surrounding environment. Amongst these, human activity is one of the promising sources of mechanical energy and hence this energy harvesting has become of great interest to researchers. Various studies have already been done by the different research groups on the development of the most capable energy harvesting devices. The triboelectric energy harvesting approach is more efficient than some of the other energy harvesting methods (e.g. compared to piezoelectric energy harvesting). Therefore, a surge of research has also been reported on triboelectric energy harvesting devices in different forms like fibre, yarn, and fabrics. The fibre based (especially nanocomposite fibre based) triboelectric nanogenerator is more advantageous than other textile forms because of its light weight, flexibility, small structure, and easy washability. In the case of fibre based or nanocomposite fibre based TENGs, various approaches have been attempted such as dip coating, spray coating or functional layer on the textile fibres, electrospun fibres and thermal drawn fibres, to develop the FTENG [140].

electric nanogenerator is composed of nylon (as positive triboelectric component) and PTFE (as negative triboelectric component) fibres (Fig. 10a). In this study, nylon and PTFE fibres were wrapped on a copper (Cu) fibre. Here, copper fibre acts as an electrode material and nylon, PTFE fibre as the friction layered. After wrapping nylon and PTFE fibres on a copper fibre, these two core-shell fibres (nylon and PTFE) were twisted into a coaxial double helix structure. Then various characterizations were carried out to know its potential. Since the developed triboelectric nanogenerator has been utilized to harvest mechanical energy during human activities like walking, moving, stretching, and so forth, the tensile property should be an essential requirement. Interestingly, the developed twined structure has shown tensile strength in excess of 200 MPa and strain to failure of about 180% (see Fig. 10a). To test triboelectric performance of the developed twined nylon and PTFE structure, the first two ends of the fibre based triboelectric nanogenerator (FTEN) were fixed and a polyester thread having diameter of 0.20 mm was rubbed against the FTENG. It was found that, when polyester fibre comes in contact with the nylon fibre, they rub each other. As per the triboelectric series, polyester fibre gains electrons and becomes negatively charged, while the nylon fibre becomes negatively charged. In this way, when the polyester thread moves toward the PTFE thread from nylon fibre, the PTFE fibre shows negative charge and polyester becomes positively charged. As a result, electrons flow from PTFE electrode surface to nylon electrode surface through the external circuit. The same phenomenon takes place when polyester thread moves from the right to the left side (PTFE thread side to nylon thread side). In this way, the FTENG can generate output voltage and current of ~850.20 mV and ~19.52 nA, respectively.

having potential energy harvesting capability. The fibre based tribo-

He and co-workers [33] developed a highly stretchable silicon fibre-based triboelectric nanogenerator (TENG) (Fig. 10b). Where silicon rubber was used as core fibre and CNT loaded polymer sheet was used as one electrode material. Then again, silicon rubber was wrapped over the surface of CNT/polymer based conductive layer and finally a micron size copper wire was convoluted over the silicon rubber layer. The CNT/polymer sheet is not only working as an electrode material but also provides mechanical strength in the TENG. Compared with the

Liu et al. [141] reported a fibre based triboelectric nanogenerator



**Fig. 10.** (a) (i) Nylon and PTFE composite fibre based TENG (FTNG) [141], (ii) digital image of the FTNG [141], (iii) a magnified view of the marked part in Fig. (a) ii [141], (iv) stress-strain curves of the nylon coated copper fibre, PTFE coated copper fibre and nylon/PTFE composite fibre [141]; (b) (i) Schematic diagram of the fibre like TENG [33], (ii) magnified view of the fibre like TENG ( insets showing SEM images of the TENG fibre [33], (iii) & (iv) image of the fibre like TENG under stretched and released conditions [33], (v) – (vii) digital images of the fibre-like TENG under stretched, bendable and twistable conditions [33]; (c) (i) Fabrication flow chart [20], (ii) SEM images [20], and (iii) images of the fully stretchable TENG based on the silicon rubber and a composite conductive electrode [20]; (d) Schematic illustration of the stretchable TENG based on Ag coated nylon yarn and PVDF-TrFE electrospun nanofibre [142]. All essential copyrights and permissions received.

previous study done by Liu et al., herein the contact area between convoluted copper wire and silicon rubber layer is much higher, therefore it can be expected that this TENG will generate higher triboelectric output (because charge density is higher owing to the greater contact area between the silicon rubber and copper wire). In this case, contact-separation takes place between the silicon rubber and copper wire surface by stretching-releasing operation. When the whole fibre is stretched, the silicon rubber surface gets separated from the copper wire surface and vis-versa. The developed TENG is not only stretchable but also bendable and twistable. Here, authors evaluate the electrical performance of the TENG against three different factors like number of coils of the copper wire on the rubber surface, stretching-releasing frequency, and stretching strain percentage. Interestingly, it was found that output voltage and current of the fibre-like TENG was changed proportionally (output voltage: 13-140 V and current: 0.005-0.075 µA/cm) with increasing the number of coils from 6/cm to 22/cm, at constant stretching frequency (2 Hz) and strain (50%). Similarly, when number of coils and strain were constant (i.e. 22/cm and 50%), the output current of the fibre-like TENG was only increased (from 0.01 to  $0.18 \,\mu\text{A/cm}$ ) with increase in the stretching frequency (0.5–5 Hz). This is due to higher charge transfer cycles taking place at a certain time, which results in a higher output current by the TENG. While output

voltage of the TENG has not changed since voltage depends on the structure and materials selection. Also, when the number of coils and stretching frequency were constant, output voltage and current were changed with increase of the strain rate. Therefore, this fibre-like TENG can easily be utilized to supply power in various wearable and portable electronic goods.

Park et al. [20] also reported similar kinds of fibre-like TENG, where silicon rubber was used as a triboelectric surface and a conductive thread was used as an electrode (Fig. 10c). The conductive thread was fabricated by silver-coated copper and polyester fibre to maintain the tensile properties of the TENG. The working mechanism of the developed TENG can be explained in such a way that, when the fibre-like TENG comes in contact with the human skin, electrons move from the skin to the silicon rubber surface. Because, as per the triboelectric series, silicon rubber has more electron affinity than human skin. And when the two surfaces have separated from each other, a positive charge is induced in the conductive thread due to the negative charge on the silicon rubber surface. This results in a current flow from human skin to the conductive thread. Again, when human skin comes in contact with the silicon rubber, induced positive charge in the conductive thread gets decreased and a reverse current is generated. In this way an electrical output will be generated by the fibre-like TENG by continuous contact-separation phenomenon. It was found that this TENG having dimensions of 8.5 cm length and 1.2 mm diameter, can generate output voltage and current of  $\sim 28$  V and 0.56  $\mu A$ , respectively against 1 kgf force induced between the human skin and silico rubber.

In another study, Sim et al. [142] studied the triboelectric effect of a stretchable fibre-like TENG (STEF) (Fig. 10d). The fibre like TENG is composed of a multilayered core-shell structure, where a silver-coated nylon filament was wrapped on a polyurethane (PU) core followed by an electrospun PVDF-TrFE web and CNT sheet placed on the surface of the silver-coated PU core. Here, silver-coated nylon filament acts as the inner electrode as well as the positive triboelectric material, whereas the electrospun PVDF-TrFE web and CNT sheet were used as the other triboelectric material and outer electrode, respectively. The developed stretchable fibre-like TENG was worked in such a way that, at initial condition the electrospun PVDF-TrFE web contacts the silver-coated nylon filament surface. This mechanical contact results in a negative charge accumulation on the surface of PVDF-TrFE web since it has higher electron affinity compared to silver-coated nylon filament as per

the triboelectric series. When the resultant fibre structure is stretched by an external force, the PVDF-TrFE electrospun web surface gets separated from silver-coated nylon filament due to their different Poisson's ratio and eventually electrons flow from inner electrode (silver-coated nylon filament) to outer electrode (CNT sheet). Again, when the external force is released, all the components (PVDF-TrFE electrospun web, silver-coated nylon filament) get back in their original position and a reverse signal is generated. The electrical performance of the STEF based triboelectric nanogenerator was evaluated against a mechanical stretching to develop a strain sensor by calibrating triboelectric output performance and resistance change of the silver-coated nylon/PU fibers. The fabricated sensor showed good sensitivity to direction of strain (50% at various frequencies up to 10 Hz).

All these four studies provide an idea regarding preparation of the fibre/yarn based triboelectric nanogenerators. It can be seen from these studies that core-sheath structure-based fibre/yarn is used as one of the triboelectric layers. The silicon rubber is mostly used to prepare fibre/yarn based TENGs. Although fibre/yarn based TENGs are quite



**Fig. 11.** (a) PVDF/CNC nanocomposite fabric based TENG (i) as prepared PVDF/CNC nanocomposite yarn [143], (ii) PVDF/CNC nanocomposite fabric [143], and (iii) the charge and power density against the external load resistance [143]; (b) The functional elastomer layer composite fabric (made from Ecoflex and CNTs) based TENG [140]; (c) (i) Schematic illustration of the NPCO/silicone and Mxene/silicone nanocomposite fabric based TENG [144], and (ii) an output voltage and current density of the plain silicone and nanocomposite fabric based TENGs [144]; (d) Schematic diagram, output signals and different applications of the porous nanocomposite fabric based TENG [145]; (e) The AgNWs and graphene nanocomposite fabric based TENG [146]. All essential copyrights and permissions received.

advantageous due to their flexibility and stretchability, they can generate low triboelectric outputs.

# 5.2. Nanocomposite fabric based triboelectric nanogenerators (NF-TENGs)

In general, textile fabric-based triboelectric nanogenerators (TENGs) often exhibit lower electrical performance due to structural limitations. This is primarily attributed to the small contact area between textile fabrics when they come into contact with each other. To address these challenges, several approaches have been explored, including fabric surface modification, microstructure introduction on fabric surfaces, and the development of nanocomposite textile fabrics using functional fillers.

However, surface modification techniques are not suitable for longterm applicability of TENGs. In contrast, nanocomposite textile fabrics offer a promising approach to enhance both the electrical performance and durability of TENGs. Several research groups have studied nanocomposite fabric-based triboelectric nanogenerators (NF-TENGs). For example, Lee et al. [143] developed an NF-TENG using a nanocomposite fabric composed of PVDF/cellulose nanocrystal (CNC) nanocomposite varns (Fig. 11a). The nanocomposite varns exhibited higher mechanical strength compared to pristine PVDF varns. The NF-TENG demonstrated an output voltage, current density, and power density of approximately  $2 \text{ V}, -155 \text{ nA/cm}^2$ , and  $219 \text{ nW/cm}^2$ , respectively, at a pressure of 9.8 kPa. Furthermore, the NF-TENG was utilized as a real-time pressure sensor to evaluate its feasibility as a smart sensing textile with CNC1 fibers. Interestingly, it was observed that the triboelectric pressure sensor was capable of sensing pressures ranging from 0.98 kPa to 98 kPa (sensing area of  $10 \times 10 \text{ mm}^2$ ).

A nanocomposite NF-TENG based on Ecoflex/CNT modified conductive fabric has been reported (Fig. 11b) in another study. It was found that the NF-TENG can generate a high output voltage of  $\sim$ 490 V, a current of 43  $\mu$ A, and a power density of ~1.6 mW/cm<sup>2</sup>, respectively [140]. Similarly, a silicon/nanoporous cobalt oxide and silicon/MXene decorated woven fabric-based NF-TENG has been reported by Rahman et al. (Fig. 11c) [144]. The NF-TENG showed a power density of 10.4  $W/m^2$ , which is 23 times higher than the silicon only based TENG. Likewise, a cellulose acetate (CA) and  $Al_3O_3$  treated conductive fabric-based NF-TENG has also been fabricated for mechanical energy harvesting and motion sensing Bai et al. [145]. The porous nanocomposite fabric (PNF) was developed by dry casting method (Fig. 11d). The PNF based TENG having 10% Al<sub>2</sub>O<sub>3</sub> concentration, showed a maximum open circuit voltage of 448 V and a power density of  $2.5 \text{ mW/cm}^2$ , respectively. Wu et al. [146] have also prepared a NF-TENG by utilizing a transparent nanocomposite textile (Fig. 11e). The NT-TENG was engineered using polyester/AgNWs/GO/PMMA, polyester/AgNWs/GO/PI, and polyester/AgNWs/GO/PDMS, as a positive and negative triboelectric layer, respectively. For developing polyester/Ag NWs/GO/PMMA triboelectric layer, the polyester fabric was first modified with the AG NWs and GO aqueous suspensions by the blade-coating method. Thereafter, PMMA solution was coated on polyester/Ag NWs/GO fabric followed by drying process. In the same way, PI solution was coated on polyester/Ag NWs/GO fabric, and PDMS solution was coated on polyester/Ag NWs/GO fabric, to obtain polyester/Ag NWs/GO/PI and polyester/Ag NWs/GO/PDMS triboelectric layers. The electrical performance was estimated by finger movement, and it was found that a single NF-TENG can generate an output voltage of  $\sim$ 4 mV, an output current of  $\sim 2 \mu A$ , and a power density of  $\sim 7 \text{ nW/cm}^2$ , respectively.

In wearable electronics, textiles are a more attractive candidate due to their outstanding deformability and breathability. Textile based TENGs (T-TENGs) can easily be incorporated with clothing to harvest mechanical energy from daily human body motions. Although the T-TENG can sense every small motions of the body, induced friction between textiles, fibres and yarns inevitably cause wear and tear, mainly due to strong repetitive mechanical stress during human activity and the durability of the textile can be deteriorated in the presence of moisture or light. For this reason, still durability of the T-TENG is a challenging task and therefore, in recent years researchers are trying to develop more durable T-TENGs by different means such as modification of textiles, making durable textiles using nanocomposite yarns, etc. As an example, Xiong et al. [43] developed a durable skin-actuated wearable NF-TENG based on the black phosphorus (BP) and the hydrophobic cellulose oleoyl ester nanoparticles (HCOENPs) modified PET fabric. The NF-TENG based on BP/HCOENPs/PET, was found to generate higher electrical performance (an open circuit voltage of ~1860 V, short circuit current density of  $\sim 1.1 \,\mu\text{A/cm}^2$ , and a power density of ~0.52 mW/cm<sup>2</sup>) as compared to pristine PET, BP/PET, and HCOENPs/PET fabrics. This is due to the synergistic effect of BP and HCOENPs in the case of the BP/HCOENPs/PET based NF-TENG. BP can entrap charges which results in lower charge loss from the TENG device and HCOENPs protects the BP from the environment. In addition, HCOENPs can induce charges on the material surface resulting in higher electrical performance.

There are various studies that have been explored to enhance triboelectric signals of the textile based TENGs by different processes such as surface modification, modification of the fibre surface before weaving, and many more. In general, surface modification or other treatment of the textile clothes will improve surface contact area during energy generation eventually which results enhancement in triboelectric signals of the textile based TENGs. Feng et al. [147] reported a fabric based triboelectric nanogenerator (Fig. 12a), where surface modified velvet fabric and PTFE material were used as positive and negative triboelectric materials. In this study, the main goal was to improve triboelectric performance of the fabric based triboelectric nanogenerator through modification of the velvet fabric by acylated CNT and polyetherimide (PEI). Surprisingly, it was found that the output voltage and current were 10 times higher than the unmodified velvet fabric. For this study, the velvet fabric was first treated by oxygen plasma followed by dipping in an acylated CNT solution with continuous ultrasonication for 20 min at 50 °C. Then CNT modified fabric was dipped into the PEI solution followed by drying at 60 °C overnight. Finally, the TENG device was fabricated by using both the triboelectric materials having dimensions of  $2 \times 2$  cm<sup>2</sup> (modified velvet fabric as positive triboelectric material and PTFE fabric as negative triboelectric material). It was found that developed fabric based TENG could generate output voltage of 119 V, output current of 12.6  $\mu$ A, and power density of 3.2 W/m<sup>2</sup>, respectively. They explored TENG as a force and tactile sensor by calibrating the peaks of the voltage level that correspond to the stepping on the TENG and the departure of the foot from the TENG device. Further, woollen gloves were used as a sensing platform for human-computer interaction applications.

Seung et al. [148] fabricated a flexible nanopattern textile based triboelectric nanogenerator (Fig. 12b). Ag coated woven fabric was purchased from the market and this Ag coated fabric was used as a base fabric to develop nanopatterns. For developing the nanopattern, first Ag coated woven fabric was washed thoroughly followed by seeding and growing of the ZnO NRs on the fabric surface. Then PDMS polymer solution was covered on the ZnO NRs array-based Ag coated textile fabric. Finally, the triboelectric nanogenerator was assembled where Ag coated woven fabric and PDMS pattern were utilized as tribo-positive and tribo-negative materials, respectively. The developed TENG showed output voltage and current of 120 V and 65  $\mu$ A, respectively, which is higher than the non-nanopatterned PDMS flat film based TENG (30 V and 20  $\mu$ A).

Dong et al. [149] developed a knitted textile based self-charging device. This power device is made of a fibre based triboelectric nanogenerator as well as a fibre-based supercapacitor. The knitted fabric structure was chosen in this study since it has suitable flexibility and stretchability. Due to this characteristic (flexibility and stretchability) of the knitted structure, mechanical energy from human motion can be



**Fig. 12.** (a) The CNT/PEI nanocomposite fabric based TENG (surface treatment flow chart, schematic diagram of the TENG, and output signal of the developed CNT/ PEI based TENG [147]; (b) The ZnO NRs modified nanocomposite fabric based TENG (schematic illustration of the TENG, a flowchart of the ZnO NRs modification, output signals, and various applications of the TENG) [148]; (c) The carbon nanofibres and H<sub>3</sub>PO<sub>4</sub>/PVA nanocomposite knitted fabric based TENG for self-charging wearable electronics [149]; (d) The PA 6-Ag, PA66-Ag and PTFE nanocomposite knitted fabric based TENG, (i) –(iii) microscopic images of the PA 6-Ag multifilament, PA66-Ag filament, and PTFE-Ag yarn [150], (iv) & (v) cross-sectional images of the PA66-Ag and PTFE-Ag yarns [150], (vi) digital image of a flat knitting machine used in this work [150], (vii) the state of loop during knitting method [150], (viii) the as prepared TENG having dimensions of 8 cm × 8 cm [150], and (ix) a magnified view of the TENG device shown in Fig. 10 (d), (viii) [150]. All essential copyrights and permissions received.

easily harvested as electrical energy. In this device, the triboelectric fabric was used to harvest mechanical energy from human motion again and the knitted supercapacitor (SC) was used to store the harvested energy to supply power in various wearable devices. The knitted triboelectric fabric was developed by using a single yarn. The yarn was made of silicon rubber coated three-plie stainless steel/polyester yarn. In this coated yarn, silicon rubber acts as triboelectric material while stainless steel/polyester acts as electrode material, respectively. And the supercapacitor (SC) yarn was fabricated by dip coating of a bundle of carbon fibres in a solution containing carbon nanofiber (CNF) and poly(3, 4-ethylenedioxythiophene)-poly-(styrenesulfonate) (PEDOT: PSS). Finally, SC yarn was incorporated in the triboelectric fabric by a knitting process. The developed integrated system of knitted self-charging power textile is symmetrically depicted in Fig. 12c. For the preparation of the knitted textile based triboelectric nanogenerator (TENG), the commercially available three-plie stainless steel/polyester yarn was coated by four different types of silicon rubber having parameters of 0-10, 0-20, 0-30 and 0-50 hardness, and 14,000, 3000, 3000, and 800 cps viscosity, respectively. Thereafter, the electrical performance of the silicon rubber coated yarns was investigated under a constant force of 5 N with frequency ranging from 1 to 5 Hz. When frequency is increased from 1 to 5 Hz having constant force (5 N), it was found that the short circuit current value is increasing with increasing frequency, but open circuit voltage (V<sub>OC</sub>) and short circuit charges were almost the same at all the various frequencies. On the other hand, the electrical performance of the yarns was also tested under a constant frequency (2 Hz) with contact force varying from 0.25 N to 9.8 N. Here, as expected (due to the contact-force dependence of TENGs), it was found that the electrical performance of the yarns was improved continuously. This may be due to the increase in contact area between the silicon rubber and an active

layer (i.e., acrylic plate) as the applied contact force was increased. Finally, it was found that the 0-20 hardness-based silicon rubber coated yarn showed better electrical performance as compared to the other counterparts. This is because a moderate hardness and lower viscosity-based silicon rubber (0-20 and 3000 cps) can enable a higher contact area and contact separation distance between the silicon rubber coated yarn and acrylic plate rather than a higher hardness and viscus silicon rubber. For this better performance of the 0-20 hardness and 3000 cps viscosity-based silicon rubber coated yarn, it was used to fabricate the TENG fabric in this study. Further, the electrical performance of knitted TENG was evaluated under standard testing equipment driven by linear motor. And interestingly, it was found that this TENG can generate output voltage and current of  $\sim 150$  V and  $\sim \! 0.55$   $\mu A$  to  $\sim$ 2.9  $\mu$ A, respectively at constant force of  $\sim 11$  N with varying the frequency from 1 Hz to 5 Hz. In another study, Dong et al. [150] also fabricated a tubular knitted fabric based TENG (Fig. 12d). Where PTFE-Ag and nylon66-Ag braided yarns were used as negative and positive triboelectric materials. A tubular knitted fabric based TENG was fabricated by using a computerized flat knitting machine. In the developed tubular TENG, PTFE-Ag and nylon66-Ag knit layer knitted in such a way that two layers are alternatingly parallel to each other and cross each other at the junctions. It was found that this t-TENG has enough stretchability (up to 110%), but for measuring the electrical performance of the developed t-TENG, 15% and 60% strain was chosen, whereas the frequency was varied from 1 to 3 Hz. It was found that this t-TENG can generate output voltage and current of 50 V and 0.9 µA, respectively.

The summary and current nanocomposite fibre and nanocomposite fabric based triboelectric nanogenerators are recorded in Table 5. From these studies it can be seen that the nanocomposite fibre and fabric

#### Table 5

A comparison of electrical performance of the NF-TENGs or NY-TENGs and NF-TENGs.

(+) layer	(-) layer	Area (cm <sup>2</sup> )	Freq. (Hz)	Press. (p) (Pa)	V <sub>oc</sub> (V)	Isc (μA)	J <sub>s</sub> (μA/ cm <sup>2</sup> )	P <sup>max</sup> (mW/ m <sup>2</sup> )	P <sup>max</sup> /p (mW/ N)	Applications	Ref.
Nylon/Cu	PTFE/Cu	-	1.8	-	2.15	0.0693	0.142	-	-	Energy harvesting	[141]
Cu	silicon rubber	-	2.0	50% stretch strain	140	0.075	-	-	-	Energy harvesting, capacitor charging, acceleration sensing	[33]
Human skin	Silicon rubber	-	-	9.8	28	0.56	-	-	-	Energy harvesting	[20]
Nylon/Ag	PVDF-TrFE	-	10 N force	50% strain	0.024	0.008	-	-	-	Energy harvesting and motion sensing	[142]
Cotton	PVDF/CNC	1	-	-	2.0	0.155	0.155	2.19	-	Energy harvesting	[143]
nitrile-butadiene rubber	Ecoflex/CNT/ LTV	6.25	2400	16	490	43	6.88	16000	6.667	Energy harvesting	[140]
Nitrile glove	NPCO/silicone/ MXene/silicone	-	5 N force	10	1680	117	10.555	10400	-	Energy harvesting and motion sensing	[144]
CA/Al <sub>2</sub> O <sub>3</sub> / Conductive fabric	LTV	9	1666.67	18	480	45	5	25000	14.99	Energy harvesting	[145]
Polyester/ AgNWs/GO/ PMMA	Polyester/Ag NWs/GO/PDMS	-	-	-	0.004	2	-	0.07	-	Energy harvesting	[146]
Human skin	BP/HCOENPs/ PET	49	1020.40	10	1860	-	1.1	5200	5.09	Energy harvesting	[43]

based triboelectric nanogenerators are prepared by the different approaches (core-sheath fibre structure, surface modification of the fabrics, nanocomposite fibre/yarn-based fabrics, etc.). The as developed TENGs have shown significant triboelectric outputs. But it is rather difficult to compare good or bad fibre or fabric based TENGs since triboelectric performance has been carried out at different loading conditions and environmental factors. However, these studies can provide a direction for future research in nanocomposite fibre or fabric based triboelectric nanogenerators.

# 5.3. Nanocomposite electrospun web based triboelectric nanogenerators (NEW-TENGs)

Among the materials being considered for TENGs, polymers have gained considerable attention - particularly electrospun polymer-based TENGs due to their high specific surface area, and excellent flexibility [151]. Polyvinylidene fluoride (PVDF) is the most commonly used polymer for preparing TENGs, primarily owing to its exceptional dielectric properties and the presence of strong electronegative fluorine groups [152]. However, TENGs prepared using polymers usually have relatively low triboelectric output that are insufficient for practical applications [153]. Doping nanofillers into the bulk of the friction materials has been shown as an effective way to enhance the triboelectric performance by providing charge trapping sites, static induction, triboelectrification and enhancing the relative permittivity of the original bulk material. Furthermore, it has already been reported that electrospun webs have good triboelectric performance but have major problems with durability. Therefore, nanocomposite electrospun webs are being considered in order to boost mechanical strength and durability. Electrospun fibrous mats can also serve to increase the contact area available for charge transfer. Various recent works on electrospun web based TENGs will now be discussed in detail.

One recent study used of a copolymer of PVDF (i.e hexafluoropropylene with liquid metal Galinstan), acting as a conductive filler, to prepare TENGs by the process of electrospinning (Fig. 13a) [154]. They stated that such electrospun nanocomposites can further enhance the triboelectric performance of the nanogenerators. It was found that by using 2 wt% liquid metal in the PVDF copolymer nanofibrous film (acting as a negative triboelectric layer with thermoplastic polyurethane as the positive layer), that the peak open-circuit voltage was recorded as 1680 V with a power density of 24 W/m<sup>2</sup> across a resistive load of 150 MΩ. These values were found to be higher when compared with the conventionally used PVDF nanocomposite electrospun web based triboelectric nanogenerators (NEW-TENGs). This excellent performance is attributed to numerous factors such as: the enhanced surface potential, capacitance, charge trapping capability, and the secondary polarization inside the PVDF-copolymer based nanofibrous web with liquid metal droplets incorporated in the web. It was further found by the scientists that incorporating 2 wt% liquid metal in the nanofibrous membrane, demonstrated generation of 12 times higher output voltage along with highest multifunctional efficiency of mechanical performance and electrical generation of TENG - in comparison to only the PVDF-copolymer sample with aluminium foil as the positive friction layer and a small air gap ( $\sim$ 75 µm) in between. Furthermore, it was reported that the tensile properties of liquid metal modified nanocomposite membranes were also enhanced. It was found that the modified TENGs could illuminate seven segment digit light emitting diode (LED) modules. Therefore, it can be concluded that liquid metal modified PVDF-copolymer nanofibrous membranes can be promising materials in high performance TENGs as the negative tribo-layer. Similarly, another research work carried out by He et al. [155] reported a new approach for hybridizing nanomaterials to build Schottky based junctions for controlling triboelectric charges by dynamic movement in the friction layer and improving the electricity generation of the flexible TENGs (Fig. 13b). For preparing such flexible TENGs, researchers have used the same above stated material, i.e. poly (vinylidene fluoride-co-hexafluoropropylene) composite nanofibrous mat, acting as the negative triboelectric layer. The previously stated study can be differentiated from the present one by the introduction of a combination of conductive silver nanowires (AgNWs) and perovskite oxide Mn-doped (Bi<sub>0.5</sub>Na<sub>0.5</sub>)TiO<sub>3</sub>-BaTiO<sub>3</sub> nanomaterials into the PVDF-copolymer web. It was found that incorporating conductive fillers in to the electrospun web showed a significant enhancement of output voltage of approximately 386% over the pristine PVDF-copolymer based nanogenerator. Furthermore, a peak open-circuit voltage of about 2170 V and a power density of about 47 W/m<sup>2</sup> were stated, which were remarkably superior to the output performance of only PVDF-based TENGs. Such superior electrical properties conferred by the nanofibrous mat may be attributed to enhanced dielectric properties, increased capacitance of the TENG, increased charge trapping capability, and enlarged work function of the composite electrospun fibrous mat. Furthermore, it was found that the Schottky junction incorporated in between the conductive perovskite oxide and metal nanowires accelerates the charge movement from AgNWs to PVDF-copolymer mat and slows down the electron



**Fig. 13.** (a) Schematic illustration of the PVDF-HFP/LM based NEW-TENG along with a photograph and testing set up [154]; (b) (i) Schematic diagram of the PVDF-HFP/AgNWs/Mn-BNT-BT nanocomposite electrospun web based TENG [155], (ii) output power corresponding to the various load resistances [155]; (c) Morphology of the (i) PVDF-ZnO NW [157], and (ii) nylon-ZnO NW electrospun nanocomposite webs [157], (iii) schematic illustration of the PVDF and nylon electrospun nanocomposite web based TENG [157]; (d) Schematic diagram of the PVDF-PVDF/BT-PVDF three-layer nanocomposite electrospun web based TENG with (i) continuous sprayed [158], (ii) separately sprayed [158], and (iii) separately sprayed with gold sputter coating [158]; (e) (i) Fabrication flow chart of the PVDF/GO electrospun nanocomposite based TENG [160], and (ii) the different states (initial, compressed, and released) of the TENG [160]; (f) (i) Schematic of the PVDF/MXene electrospun nanocomposite web based TENG [161], (ii) open circuits voltage ( $V_{OC}$ ) and short circuit current of the fabricated TENG with different external loads [161], and (iii) the output power of the TENGs made from PVDF/MXene electrospun nanocomposite web having different concentration of MXene corresponding to the different external loads [161]. All essential copyrights and permissions received.

dissipation, thereby helping to retain a higher and long-lasting potential. Therefore, it can be concluded that such modified TENGs show renewability, biocompatibility, and wearability of the supplied energy useful for physiological monitoring, human machine interactions and healthcare in this rapidly evolving information age. Another similar work used the same material (i.e. PVDF/AgNW composite and nylon nanofibers) to prepare TENGs as the top and bottom triboelectric layers. Such ferroelectric polymer–metallic nanowire composite nanofiber triboelectric layers is described for use in high-performance TENGs. Electrospinning process further facilitated the uniaxial stretching of the polymeric chains, which improves the formation of the highly oriented crystalline  $\beta$ -phase that forms the most polar crystalline phase of PVDF. The addition of AgNWs further promotes the  $\beta$ -phase crystal formation by introducing electrostatic interactions between the surface charges of the nanowires and the dipoles of the PVDF chains. The ability of trapping the induced tribocharges increases upon the addition of nanowires to the PVDF matrix. The enhanced surface charge potential and the charge trapping capabilities of the PVDF-AgNW composite nanofibers

significantly enhance the TENG output performance. Finally, the mechanical stability of the electrospun nanofibers is dramatically enhanced while maintaining the TENG performance by applying thermal welding near the melting temperature of PVDF [156]. In this context, another similar study involving electrospun PVDF and a nylon 11 fibrous web with zinc oxide nanowires (ZnO NWs) was investigated to prepare flexible TENGs (Fig. 13c) [157]. It was found that ZnO NWs were aligned along the fibre axis during electrospinning. Such cooperative and mutual alignment of polymer chains with ZnO NWs were achieved because of electrospinning, which led to the formation of highly polar crystalline  $\beta$ -phase of PVDF and  $\delta$ ' phase of nylon 11. It was found that the maximum power density reached 3.0 W/m<sup>2</sup> under an external load of 10-20 MQ. Furthermore, the TENG exhibited output performance capable of directly lighting more than 100 LEDs. Additionally, the incorporation of ZnO NWs enhanced both thermal stability and mechanical properties, such as tensile strength and elastic modulus of PVDF and nylon 11 fibrous membranes. So, this work can be considered as a potent and sustainable power source for portable electronic devices. On the other hand, ceramic doped-polymeric structures as organic and inorganic hybrid structures create a new field in terms of advanced materials for flexible along with stretchable sensors and actuators. Herein, another recent study explored the use of a uniform ceramic-based polymer composite made up of BaTiO<sub>3</sub> and PVDF using solution casting to enhance the pressure sensitivity (Fig. 13d) [158]. The incorporation of BaTiO3 nanoparticles into the PVDF structure has been shown to enhance piezoelectricity and pressure sensitivity. Moreover, the research group presented a novel flexible and stretchable multi-layered pressure sensor composed of electrospun nanocomposite fibers, which exhibited a high electrical sensitivity of approximately 6 mV N-1. In comparison, the solution-cast PVDF sensors mentioned earlier demonstrated an electrical sensitivity of 1.88 mV N-1 when subjected to cyclic loads at a frequency of 2.5 Hz and a constant load of 0.5 N. This work presents a promising approach for the fabrication of nanostructures for pressure sensors in various wearable devices and technologies. Similarly, another study utilized PVDF/BaTiO3 to create hybrid nanogenerators through the process of electrospinning [159]. The charge density of the hybrid tribo and piezoelectric nanogenerator was recorded as 2.1 times higher than that of the sum value of the pure electrospun hybrid nanogenerators, which can reach up to 105.6 µC  $m^{-2}$ . Improved performance of the hybrid nanogenerator was due to the enhanced synergistic coupling for triple effects of pore dipole, triboelectricity and piezoelectricity. This work showed novelty due to the fact that the researchers here have developed a hybrid nanogenerator, which is able to harvest biomechanical energies generated from various action related to our daily life. Another novel work developed the technique of preparing a book-shaped TENG made up of electrospun PVDF and poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) nanofibres to effectively harvest mechanical energy (Fig. 13e). In addition, they have dispersed graphene oxide (GO) in the PVDF nanofibres, which act as charge trapping sites, thereby increasing the interface for charge storage as well as the output performance of the TENG. Also, this modified TENG was used as a direct power source to drive small electronics such as LED bulbs. This kind of study demonstrates the possibility of improving the performance of TENGs using composite materials [160]. Similarly, in another work, the use of electrospun PVDF functionalized with MXene (Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub>) composite nanofiber was proposed as a promising negative triboelectric layer for enhancing the performance of TENGs (Fig. 13f) [161]

The fillers were dispersed into the PVDF matrix by the process of electrospinning to enhance the dielectric property and surface charge density of the nanogenerators. It was found that the dielectric modulation by inclusion of such conductive nanosheets significantly enhanced the dielectric constant and the surface charge density of by 270% and 80%, respectively. Therefore, it was found that the TENG made up of PVDF/MXene along with Nylon 6/6 nanofibre can deliver a peak power of 4.6 mW (having a power density of 11.2 Wm<sup>-2</sup>) at a load of 2 MΩ,

which was found to be 1.58 times higher than that of pure PVDF electrospun nanofibre. In addition, the fabricated TENG shows excellent performance under low-frequency impact motions with high durability up to more than 60k cycles of output signals, quick charge storage capacity, and can easily illuminate around 100 commercial LEDs. Finally, this work successfully demonstrated a self-powered TENG used in sensing foot motion capable of controlling automatically the step lights based on the human foot motion over the stairs. Another study by Rana et al. [162] employed the same material, namely PVDF-TrFE/MXene nanocomposite (Fig. 14a). The prepared material exhibited an exceptional dielectric constant and a high surface charge density. The fabricated NEW-TENG achieved a maximum power density of 4.02 W/m<sup>2</sup> at an external load resistance of 4 MΩ. Furthermore, the nanocomposite demonstrated an enhanced output performance that was four times greater than that of pure PVDF-based TENGs. It was found that the fabricated nanocomposite could successfully be used to power an electronic stopwatch and thermohygrometer by using harvested energy released by human finger tapping. Furthermore, the device was used in smart home applications as a self-powered switch for controlling electrical home appliances, including fire alarms, fans, and smart doors. So, this study presents an effective and new approach toward self-powered systems, human-machine interface and smart home applications. Another new kind of TENG based on electrospun PVDF and nylon nanowires was reported in literature by Zheng et al. [163] (Fig. 14b). It was found that the nanogenerator exhibited easy fabrication, low cost and high output. The TENG showed open-circuit voltage of around 1163 V and short-circuit current density up to  $11.5 \,\mu\text{A/cm}^2$  at a frequency and amplitude of 5 Hz and 20 mm, respectively. In addition, the nanogenerator released power density of around 26.6 Wm<sup>-2</sup>. The novel feature of this work was that the as fabricated TENG could directly power a direct current (DC) motor without using any energy storage system. Therefore, such a TENG could be used as a fully self-powered ultraviolet radiation detection device without using any additional components. In this context, another research work aiming to harvest mechanical energy using a green technology for applications in self-powered portable electronics, wireless sensing, implanted devices, and security systems was reported by Haung et al. [164]. The same group of researchers have previously explored a NEW-TENG fabricated using PVDF nanofibres. This prepared device exhibited an output voltage of around 210 V. Therefore, to obtain a device showing more output voltage, these researchers doped PVDF nanofibres with silica (SiO<sub>2</sub>) nanoparticles, which enhanced the output electrical generation of the resultant TENGs (Fig. 14c). It was found that the peak voltage was increased to 370 V as the weight % of SiO2 nanoparticles increased to 0.6 with further increase in the nanoparticle content showing a deterioration in electrical properties. The researchers further stated that, when such hydrophobic nanoparticles were modified by an octanol treatment (mSiO<sub>2</sub>), this drastically improved the output performance of the prepared TENGs. It was stated that an output peak voltage of around 430 V was obtained with 0.8 wt% of mSiO<sub>2</sub> demonstrating an increase of about 48% when compared with the output value represented by a PVDF/SiO<sub>2</sub> nanofibrous TENG. This TENG was used in a self-powered digital thermometer for temperature measurement without the use of a battery. Similarly, in another study [165] TENGs based on PVDF having a high degree of crystallinity were reported (Fig. 14d). The electrospinning process was used to grow PVDF nanofibres. Commercially obtained printer ink (PI) nano-fillers were incorporated in a PVDF electrospun nanofibrous structure to further enhance the degree of crystallinity to obtain a high output voltage TENG. Crystallinity of around 88%, as quantified using FTIR spectroscopy, was obtained for PVDF-PI nanofibers grown over a period of five hours. Furthermore, the nanogenerator showed output power of 22  $W/m^2$ , more than double that of PVDF nanofibres, which, at the time, was considered the highest recorded value for PVDF nanofibre based TENGs. The reason for such outstanding performance is due to ferroelectric domain alignment, surface charge density, and nanofibre medium properties. Also, as it is



**Fig. 14.** (a) The PVDF/MXene electrospun nanocomposite fibre based TENG (schematic diagram of the TENG along with its applications flowchart, dielectric constant of the nanocomposite fibre with respect to MXene volume fraction, and the charge density vs volume fraction of the MXene [162]; (b) (i) A schematic illustration of the PVDF/Nylon electrospun nanocomposite fibre based TENG [163], (ii) morphology of the PVDF nanofibres [163], and (iii) morphology of the nylon nanofibres [163]; (c) The PVDF/mSiO<sub>2</sub> electrospun nanocomposite fibre based TENG (a schematic diagram of the TENG and its applications) [164]; (d) A TENG based on PVDF/PI electrospun nanocomposite fibre (schematic illustration of the TENG, PVDF and PVDF/PI nanofibres and electrospinning setup [170]; (e) (i) A schematic diagram of the PVDF/graphene electrospun nanocomposite fibre based TENG [42], (ii) digital image of the TENG [42], (iii) output signals vs graphene concentration and external load resistance [42]. All essential copyrights and permissions received.

known that printer ink contains magnetic materials, the relation of magnetic properties to the  $\beta$  phase and the power output as a function of growth time have been discussed. Such nanogenerators can be used as humidity sensors, easily integrated into flexible electrical and optoelectronic systems. Another work in this context [166] reported the fabrication of a 20  $\times$  20 mm<sup>2</sup> TENG device made up of PVDF/Graphene nanofibers along with films made up of polyamide-6 (PA6). The resulting TENG displayed enhanced triboelectric performance with an output voltage of around 1511 V, a short-circuit current density of about 189 mA  $\mathrm{m}^{-2},$  and a maximum peak power density of approximately 130.2  $Wm^{-2}$ , which is nearly eight times higher than that of the pure PVDF-PA6 TENGs (Fig. 14e). Additionally, the modified nanocomposite TENG can harvest energy of about 74.13 µJ/cycle, with an output power density of about 926.65 mW/m<sup>2</sup>. This work therefore provides an effective strategy of simultaneously optimizing the chemical composition and surface microstructure of triboelectric materials to significantly improve the output performance of TENGs. Min et al. [167] developed high-performance ferroelectric-assisted TENGs using electrospun fibrous surfaces based on PVDF-TrFE incorporated with a conductive

filler i.e. BaTiO<sub>3</sub> in either cubic or tetragonal form. Three types of TENG were tested i.e. pristine PVDF-TrFE), PVDF-TrFE)/cubic BaTiO3 and PVDF-TrFE)/tetragonal BaTiO<sub>3</sub>. When tested in contact with polyethylene terephthalate, the results showed increases in output electrical output from pristine  $(0.75 \text{ W/m}^2)$  to cubic BaTiO<sub>3</sub>  $(2 \text{ W/m}^2)$  and to tetragonal BaTiO<sub>3</sub> (2.75  $W/m^2$ ). Furthermore, electrospinning increases the maximum output voltage to 315 V for TENGs when compared with the spin-coated PVDF-TrFE)/ BaTiO<sub>3</sub>. The reason is attributed to the increase in alignment of the dipoles due to the application of high voltages, which helps in the formation of a highly oriented crystalline β-phase via uniaxial stretching. Essentially, transfer of triboelectric charge is enhanced because of increased surface potential owing to enhanced ferroelectric polarization. It was further stated by the researchers that higher output voltage is produced by PVDF-TrFE)/ tetragonal BaTiO<sub>3</sub> compared to PVDFTrFE)/ cubic BaTiO<sub>3</sub> even though permittivity is almost the same. Finally, the TENG was integrated with a graphene based flexible electrode supercapacitor and produced 1.25 V in a very short duration of about 5 min. These results demonstrate that this technology is helpful in wearable applications where higher power output, more efficient charging and flexibility is of utmost importance. In this context, another flexible and lightweight TENG was fabricated using MoS<sub>2</sub>/carbon nanotube (MC)-doped PVDF as the friction substrate using the electrospinning technique for harvesting energy from human body motions under various mechanical deformations. It was found that the charge density of PVDF nanofibers increased significantly with the introduction of an electron acceptor of the MC composite, and nylon as a clothing material, acting as a friction layer that simplifies the device structure. The authors stated that by optimising electrospinning parameters, an output voltage of more than 300 V can be reached and can generate power up to 0.484 mW. Also, the device was found to remain durable up to 3000 cycles and has the capability for charging a capacitor. Finally, it can be concluded from the results that this flexible device can easily convert mechanical energy to electrical energy and can be applied in the field of power supply for portable electronic devices [168]. Another novel work related to PVDF based TENGs was demonstrated by Fatma et al. [169]. They stated that by incorporating maghemite ( $\gamma$ -Fe<sub>2</sub>O<sub>3</sub>) fillers in PVDF with polyethylene terephthalate by the process of electrospinning, the output electrical performance of the TENGs can be enhanced significantly and are applicable in powering wearable electronic devices. Results showed that addition of nanofillers in the PVDF matrix produced an open circuit voltage of around 250 V and short circuit current of 5  $\mu$ A, which is substantially higher than that of pristine-PVDF-based TENG. Furthermore, it was found that the device can induce a maximum power output of 0.17 mW with a power density of 0.117 W/m<sup>2</sup> by the application of



**Fig. 15.** A. Schematic illustration of the PAN/LM based NEW-TENG [171]; B. NEW-TENG based on graphene nanoplates (GNPs) and tetraethyl orthosilicate (TEOS) incorporated polyvinyl butyral (PVB) electrospun nanocomposite [172]; C. Silk fibroin (SF) fibrous layer and a polycaprolactone (PCL)/graphene oxide (GO) based flexible TENG (i) schematic illustration of different triboelectric layers [173], (ii) & (iii) an output voltage and current for the different PCL/GO based TENGs [173]; D. PCL/GO electrospun nanocomposite fibres based TENG (i) different layers structures of the developed TENG [174], (ii) & (iii) an output voltage and current of the TENGs [174]; E. PA66/MWCNTs NF and PVDF NF based TENGs (i) structure of the as developed TENG [175], (ii) output voltage of the TENGs [175], and (iii) short circuit current of the TENGs [175]. All essential copyrights and permissions received.

manual force. Also, the device is extremely robust with excellent long-term stability for approximately 3000 s. The novel outcome for this work was that the device could harvest biomechanical motion in the form of slow and fast foot movement by attaching it to the footwear sole. Besides, the device could continuously supply power to light up to 108 series-connected LEDs, without the use of a capacitor and has potential applications in self-powered wearable and portable electronics obviating the use of batteries. Moreover, this device could harvest energy from a rotary pump to charge a 1  $\mu$ F capacitor to a value of around 30 V in a very short duration of time i.e. 90 s. Finally, a thick magnetic  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub>/PVDF based nanocomposite film was also successfully investigated as a magneto-triboelectric nanogenerator (M-TENG) in noncontact mode showing potential for harvesting of the stray magnetic field.

In addition to PVDF polymer, studies have also explored the use of polyacrylonitrile (PAN) polymer incorporated with conductive fillers for fabricating nanowire-based triboelectric nanogenerators (NEW-TENGs). In the NEW-TENG, PAN nanofibrous web incorporated with LM serves as the positive triboelectric material, while a thin film of PTFE serves as the negative triboelectric material (Fig. 15a) [171]. The NEW-TENG demonstrated its maximum electrical performance when the PAN nanofibrous web with a 1.5% incorporation of LM was used. It was observed that the electrical performance improved with higher LM concentration. This improvement can be attributed to the higher dielectric constant of PAN polymer in the presence of LM and the charge trapping ability of LM. The PAN/ML-PTFE-based NEW-TENG exhibited an output voltage of approximately 375 V and a current density of approximately 35 mA/m<sup>2</sup>, while the PAN-PTFE-based NEW-TENG showed an output voltage of around 190 V and a current density of about 20 mA/m<sup>2</sup> when subjected to an applied force of 30 N at a frequency of 3 Hz [172]. In this case, the electrospun PAN acted as the negative triboelectric layer, while the electrospun nanocomposite PVB served as the positive triboelectric layer (Fig. 14b). The resulting NEW-TENG generated an output voltage of 810 V, an output current of 263  $\mu$ A, and a power density of 20 W/m<sup>2</sup>. Notably, the power density of the NEW-TENG was approximately six times higher than that of the pristine PVB-based TENG. This improvement can be attributed to the higher surface roughness and surface area achieved with the nanocomposite GNPs/TEOS/PVB electrospun web compared to the pristine PVB electrospun web.

Parandeh et al. [173], in 2019, published a work on a hybrid triboelectric nanogenerator based on graphene oxide (GO) incorporated polycaprolactone (PCL) and cellulose paper. Here GO incorporated PCL electrospun nanocomposite web and cellulose paper were used as the triboelectric negative and positive layer, respectively. The PCL/GO electrospun nanocomposite was developed by using different concentration of GO. The electrical performance of the NEW-TENG is increased with increasing GO concentration up to 4%. However, further increasing the GO content (8%) results in lower electrical performance: this may be due to the aggregation of the GO or increasing of the fibre diameter with higher concentration of GO. It was found that the output voltage, output current, and power were increased from 0.473 V to 22.73 V, 4-165 nA, and 0.473 nW to 938 nW, respectively, at 8 N applied load and 2 Hz frequency, as shown in Fig. 15c. In another study, authors have also reported a NEW-TENG using PCL/GO triboelectric material too [174]. The NEW-TENG was fabricated by PCL/GO electrospun web as the negative tribo layer and nanofibrous silk fibroin as the positive tribo layer. The NEW-TENG has also followed the same electrical performance as the previous work. The output voltage of the NEW-TENG is increased from 23 V (0% GO) to 100 V (6% GO) and the output current is increased from 0.16 µA (0% GO) to 2.52 µA (6% GO), respectively, under constant force and frequency of 13 N and 3 Hz. (Fig. 15d). However, the electrical performance of the NEW-TENG is decreased with further increasing the GO concentration in PCL polymer. At lower concentration of GO, lower electrostatic induction takes place due to lower conductivity; whereas, at higher concentration, aggregation takes place which results in lower electrical performance of the NEW-TENG.

Sun et al. [175], in 2021, reported a waterproof, breathable, and washable nanocomposite electrospun web based triboelectric nanogenerator (NEW-TENG), for wearable applications (Fig. 15e). The NEW-TENG is composed of electrospun PA66/MWCNTs (positive tribo layer) and the electrospun PVDF nanofibrous web (negative tribo layer), respectively. MWCNTs is one of the good conductive materials and therefore, in this study MWCNTs are used to enhance the conductivity of the PA66 nanofibrous web. The conductive characteristic of the PA66 web could facilitiate electrostatic induction during the triboelectric performance between PA66/MWCNTs and PVDF electrospun layers. For evidence, it was found that the output signals are increased with increasing the MWCNTs concentration up to 0.5%. However, further increasing of MWCNTs caused lower electrical performance due to higher conductivity. At high electrical conductivity, the tribo layers will lose more electrons. The NEW-TENG showed an output voltage of 142 V, an output current of 15.5  $\mu$ A, and a power density of 1.30 W/m<sup>2</sup> at an applied force and frequency of 15 N and 5 Hz, respectively.

Similar to nanocomposite fibre/yarn or nanocomposite fabric based TENGs, nanocomposite electrospun web based triboelectric nanogenerators are summarized in Table 6. The electrospun nanocomposite based TENGs are developed by different approaches (various materials aspect, various TENGs design aspect, etc.). Throughout the above studies on the nanocomposite electrospun web based TENGs, it can be found that the as developed TENGs have shown very good triboelectric outputs. But triboelectric performance of the nanocomposite electrospun TENGs is difficult to distinguish due to varied loading conditions and environmental factors. Although electrospun TENGs show better triboelectric properties than the fibre or fabric based TENGs, the electrospun fibre has lower durability and stability. In addition, a comparison of different techniques or methods for the preparation of textile based piezoelectric and triboelectric nanogenerators is shown in Table 7. Table 7 provides an in-depth insight for the research community regarding different methods or techniques for preparation of the textile based nanogenerators.

#### 6. Challenges and prospects

The nanocomposite-based textile-PENGs and TENGs have the ability to generate higher output than traditional PENGs and TENGs for wearable smart sensing and device applications. However, these devices possess ongoing challenges like low electrical output performance, low contact surface area, washability, breathability, device adaptability to fabrics, charge extractions and energy storing, and commercialization.

### 6.1. Low electrical output performance

While nanocomposite textile-based (PENGs) and triboelectric nanogenerators (TENGs) exhibit superior electrical performance compared to conventional textile-based PENGs and TENGs, further enhancements are still required for real-time applications. These nanocomposite textilebased devices are ideal candidates for wearable electronic applications due to their enhanced flexibility and wearability. However, it is important to note that they generate lower energy levels compared to traditional solid-state material-based nanogenerators (NGs). Nevertheless, these nanocomposite textiles based NGs offer improved output performance and other relevant properties due to the synergistic characteristics of the nanocomposite materials and textiles in wearable conditions. For instance, Xiong et al. [43] demonstrated enhanced output performance (Voc of approximately 1860 V) by incorporating combinations of BP/HCOENPs/PET nanocomposites, surpassing that of traditional TENGs. Nonetheless, further research is still needed to enhance the stable and continuous electrical output of nanocomposite textile based NGs. To enhance the electrical output performance of T-TENGs, optimizing the electrode design within the yarns or textile is fundamental. Employing conductive materials with high electrical conductivity, coupled with an efficient electrode geometry, is essential

#### Table 6

Summary and comparison of the different electrospun nanocomposite fibre based TENGs.

(+) layer	(-) layer	Area (cm²)	Pressure (p) (kPa)	Freq. (Hz)	V <sub>oc</sub> (V)	I <sub>SC</sub> (μΑ)	J <sub>s</sub> (μA/ cm²)	Pd <sup>max</sup> (mW∕ m²)	Load (MΩ)	P <sup>max</sup> /p (mW/N)	Ref.
TPU film	PVDF-HFP/LM nanofiber	4	250	5	1680	12.8	3.2	24000	150	0.096	[176]
Al tape	PVDF-HFP/5%AgNWs/5%Mn- BNT-BT	4	250	4	2172	30	7.5	47.3	70	0.00018	[155]
Nylon/ZnO NWs	PVDF/ZnO NWs	10	100	3.2	330	10	1.0	3.0	10	0.00003	[157]
PVDF/GO nanocomposite electrospun fibre	Poly(3-hydroxybutyrate-co-3- hydroxyvalerate) (PHBV) nanofibers	20	-	1.8	200	50	2.5	2.3	8	-	[160]
Nylon 66 nanofibre	PVDF/MXene nanocomposite fibre	4	125	8	710	162	40.5	11.213	2	0.00008	[161]
Nylon 11 nanofibre	PVDF-TrFE)/MXene nanocomposite fibre	-	7 N force	6	190	35	-	4.02	4	-	[162]
PVDF/mSiO <sub>2</sub> nanocomposite electrospun fibre	Poly(3-hydroxybutyrate-co-3- hydroxyvalerate) (PHBV) nanofibers	20	-		430	85	-	-	-	-	[164]
PVDF nanofibre	PVDF/printer ink (PI) nanocomposite fibre	9	0.45	2–3	1600	130	14.44	22	500	0.0489	[170]
PA 66 film	PVDF/graphene nanocomposite fibre	4	125	5	1511	75.5	18.9	130.2	200	0.00104	[42]
PET film	P(VDF-TrFE)/ BaTiO <sub>3</sub> nanocomposite fibre	6.25	12.8	4	315	42	6.7	2.75	100-500	0.00021	[177]
Nylon cloth	PVDF-MoS <sub>2</sub> /CNTs nanocomposite fibre	36	13.88	1.6	300	11.5	0.319	0.134	100	0.000009	[168]
PAN/LM nanocomposite fibre	PTFE thin film	4	75	3	375	-	3.5	-	-	-	[171]

#### Table 7

Comparison between the different methods for preparation of textile based PENGs and TENGs.

Methods	Merits	Demerits		
Textile fibre/yarn based NGs (PENGs and TENGs) Nanocomposite fibre/ yarn based NGs (PENGs and TENGs)	Easy to incorporate into the cloth since they are flexible in nature ✓ High electrical outputs compared to conventional fibre/yarn based NGs (PENGs and TENGs) ✓ High mechanical strength and stability	Low electrical outputs due to the small resulting contact area Difficult to maintain uniform distribution of the piezoelectric fillers in polymer matrix		
Electrospun fibre based NGs (PENGs and TENGs)	High electrical outputs compared to other methods ( due to in-situ poling and high mechanical stretching for the PENGs and higher surface area for the TENGs)	Low durability and stability		
Nanocomposite electrospun fibre based NGs (PENGs and TENGs)	Comparatively high electrical outputs compared to conventional electrospun fibre based NGs ( due to the additional piezoelectric effect of the inorganic fillers for the PENGs and additional dielectric characteristics of the fillers for the TENGs)	Low durability and stability, non-uniform distribution of the fillers		
Textile fabric based NGs (PENGs and TENGs)	High dimensional stability, high electrical outputs compared to fibre/yarn based NGs (PENGs and TENGs)	High complexity in the preparation method		
Nanocomposite textile fabric based NGs (PENGs and TENGs)	Comparatively high electrical outputs compared to conventional textile fabric based NGs	Complex method, tendency of non-uniform distribution of the nanofillers		

for maximizing charge collection and transfer. Recent advancements have shown promise in the development of multi-layered textile structures, each serving a distinct function. For instance, one layer can be dedicated to energy harvesting, another for energy storage (utilizing components like supercapacitors), and a third layer designed to enhance user comfort. The inclusion of capacitors, supercapacitors, or energy-efficient power management circuits can significantly improve the storage and utilization of harvested energy. Efficient power conversion electronics are equally critical in converting and conditioning the low-voltage, low-current output of the NGs into a more practical form suitable for charging devices or powering low-energy electronics. The integration of self-charge extraction and circuits, employing charge trapping and self-excitation methods, is emerging as a key component of power management electronics within nanocomposite NGs. Furthermore, facilitating the effective transfer of charge from nanocomposite NGs to practical wearable applications demands further research, primarily due to the impedance mismatch between the power source and the load. Enhancing the electrical output of low-output textile TENGs and PENGs necessitates a comprehensive, multidisciplinary approach encompassing materials science, textile engineering, and electronics. Experimentation and rigorous testing remain crucial for refining the design, tailoring it to specific applications, be it wearable technology, smart textiles, or energy-efficient clothing.

### 6.2. Non-uniform filler distribution in the polymer matrix

Nanocomposite textiles, which incorporate triboelectric or piezoelectric materials, offer the potential for higher permittivity or dielectric constant. This is primarily due to their increased ability to trap charges compared to conventional pure textile materials. Consequently, nanogenerators (NGs) constructed using these nanocomposite textiles are expected to outperform their counterparts in terms of electrical performance. In nanocomposite textiles, the inclusion of inorganic fillers and organic polymers can create a substantial interface between materials, further enhancing charge trapping capacity. However, achieving a consistent and continuous distribution of nanofillers within nanocomposite textiles presents a challenge, as nanomaterials tend to agglomerate. Such agglomeration can lead to unstable performance in nanocomposite piezoelectric and triboelectric nanogenerators (PENGs and TENGs). To tackle this issue effectively, it is more suitable to develop stable layers that consist of composite nanostructures (such as nanorods, nanofibers, or nanowires) made of metal oxide/polymers (e. g., ZnO, PVDF) on pure textile materials. To advance uniform filler distribution, it is essential to explore various techniques, including

preparation methods like solution mixing, in situ polymerization, and mechanical mixing, surface modification such as surface treatment and the use of coupling agents, controlled processing involving temperature control and shear forces, optimized mixing time and speed, the incorporation of nanofillers, the masterbatch approach, additive manufacturing, and others. By implementing these strategies, you can significantly enhance the uniform distribution of fillers within the polymer matrix of textile based PENGs and TENGs. This improvement leads to enhanced device performance and greater energy harvesting capabilities while preserving the washability and comfort properties of the textiles.

# 6.3. Washability

A key practical requirement of wearable textile NGs is washability. The washability of wearable nanocomposite textile-based piezoelectric and triboelectric nanogenerators (NGs) presents additional challenges due to the need to maintain the device's functionality and performance after washing. Wearable textiles need to be repeatedly washed either by machine or any other traditional method by using water (H<sub>2</sub>O), soap, and other chemical detergents. The nanocomposite textile NGs are aimed at producing a sustainable energy generator and this device is a complete component of the wearable textile or maybe a part of the textile. Therefore, they should be washable like normal clothes with a removal electronics device to protect the electronic circuits Hence, device components, including the nanocomposite materials, electrodes, and interconnects, need to be water-resistant to prevent water penetration and potential damage to the device. Water ingress can lead to degradation of the materials, loss of electrical connectivity, or changes in the device's electrical properties. In order to test this ability, many papers report that they are evaluated in such a way that multiple machine washing and drying processes are being performed with respect to time. However, due to the interfering reaction of chemical detergents with nanocomposites at the outer surface, NGs erode the capability of nanocomposites due to multiple washes. The washing process involves mechanical agitation, such as spinning and tumbling, which subjects the NGs to mechanical stress. The device therefore should be mechanically robust enough to withstand the rigours of washing without undergoing significant deformation, delamination, or structural damage. Maintaining the integrity of the nanocomposite textile and the interfaces between different components is crucial for long-term device performance. Washability is a significant challenge in exposing the nanocomposite and electrode material layer to soap or chemical detergents. Some detergents contain chemicals that can interact with the device materials, leading to material degradation or altered electrical properties. Ensuring the compatibility of the nanocomposite textiles and other device components with detergents is essential for maintaining the NG's performance after washing. The exposure to water and detergents may also cause changes in the conductivity of the electrodes, alter the piezoelectric or triboelectric properties of the nanocomposite materials, or induce changes in the device's electrical connectivity. It is crucial to ensure that the NG retain their electrical stability and performance even after multiple washing cycles. Frequent washing and exposure to harsh conditions can accelerate material degradation, fatigue, or wear and tear. The device should be designed to withstand repeated washing cycles while maintaining its functionality and performance over an extended period. The development of an entire fabric from nanocomposite PENG and TENG is still a big challenge due to the composite material properties (toxicity level, biocompatible, biodegradability etc.) and its stability for long term use (washing, drying and etc.) of the fabrics. Addressing the washability challenge requires careful selection of materials that are water-resistant and chemically stable, optimization of device architectures to enhance mechanical robustness, and development of fabrication techniques that ensure long-term durability. Additionally, implementing protective coatings or encapsulation methods can help enhance the washability and longevity of wearable

NGs by providing a barrier against water and chemicals.

### 6.4. Durability and stability

The reliable and stable electrical output of NGs is an essential requirement to operate real-time wearable sensing devices. Even though NGs are proven to be sustainable green energy alternatives, they still struggle to achieve reliable and stable outputs for long term use. The materials used in these devices should possess sufficient mechanical strength, flexibility, and resilience to ensure long-term durability. However, nanocomposite textiles can experience fatigue and mechanical failure over time, especially when subjected to extensive bending, stretching, or twisting. Another essential factor is nanocomposite textile material degradation with respect to multiple washing and extreme environmental conditions including temperature, humidity, and chemical exposure. These factors can deteriorate the performance and stability of the device over time. For example, moisture absorption in the nanocomposite textiles can lead to dimensional changes and can alter the device output performance and hence its stability. Similarly, exposure to chemicals or UV radiation can cause degradation of the materials, reducing the device's efficiency and lifespan. However, the NGs typically involve interfaces between the nanocomposite textiles with electrodes and interconnects. Ensuring stable and reliable interfaces is crucial for long-term device performance. Weak interfaces can lead to delamination, increased resistance, or loss of electrical connectivity, affecting the overall durability and functionality of the NGs. The output performance of nanocomposite TENGs tends to decrease with respect to time due to composite material degradation within the fabrics. This is due to the repeated mechanical stress, that leads to cracks, fractures, or degradation of the nanocomposite textile, resulting in reduced piezoelectric or triboelectric conversion efficiency. The electrical output of the device should remain consistent and predictable over time. However, factors like material aging, degradation, or wear can affect the stability of the NGs, resulting in a decline in their energy conversion efficiency. Therefore, the integration of standard power management circuits with machine learning predictive systems is imperative, adapting to changes occurring within the composite layers. Additionally, adopting a comprehensive strategy is vital. This includes the selection of robust and wear-resistant textile materials fortified with fibers to withstand mechanical stress, the application of protective coatings to both the textile surface and NG components to shield against moisture and contaminants, and the optimization of electrode design with corrosionresistant materials and secure adhesion.

### 6.5. Energy conversion efficiency

Achieving high energy conversion efficiency is a crucial challenge for wearable nanocomposite textile-based piezoelectric and triboelectric nanogenerators (NGs). The choice of materials used in nanocomposite textiles plays a significant role in determining the energy conversion efficiency. Finding materials with high piezoelectric or triboelectric coefficients is essential to maximize the conversion of mechanical into electrical energy. However, identifying and developing materials that simultaneously exhibit desirable mechanical, electrical, and piezoelectric or triboelectric properties is challenging. Further the interfaces between the nanocomposite textile and the device components (power management circuits, storage, charge extractor and etc), can significantly impact the energy conversion efficiency. Interface engineering techniques, such as surface modifications, interfacial layers, or electrode optimization, are necessary to enhance charge generation, collection, and transport, thus improving energy conversion efficiency. Apart from materials properties and circuits, the design and architecture such as electrode configuration, device geometry, interconnectivity, and structural parameters can also affect the distribution of stress or charge density within the device - directly impacting the energy conversion process. Optimizing device design to maximize the utilization of mechanical energy and minimize energy losses is a challenge that requires careful consideration. Ultimately, suitable power management and energy storage systems are essential to harness and utilize the generated electrical energy effectively. Wearable NGs often produce intermittent and variable electrical outputs, requiring appropriate power conditioning, regulation, and storage (flexible supercapacitors), and solving these challenges requires advances in material science, device engineering, and system integration.

# 6.6. Commercialization and industrialization

Considerable progress would be required to enable textile nanocomposite NGs to be practical for real-time wearable energy harvesting applications. Issues such as low output power, washability, reliability, stability, and comfort etc. would have to be addressed. Researchers are trying to develop core spun yarn based NGs, which consist of bundles of nano or microfibers and current fabrication technology is often focused on high voltage-electrospinning. The major challenge is the complexity of the fabrication process, high-power consumption, and low production rate and so considerable progress would be required to produce nanofibers on a large scale to enable industrialization. New advanced material combinations and structures and more efficient nanocomposite textile NGs are very often complex and sometimes toxic in nature having multiple chemical and yarn-making processing steps; it should be remembered that this reduces the cost efficiency and sustainability of some nanocomposite Textile NGs. Biocompatible and degradable materials selection will play an important role in the fabrication of stable and eco-friendly energy devices. A lot of work is required for assessing the lifetime response of nanocomposite textile NGs in real time environments to make them commercialized products. Another major issue is the industrialization of the fabrication process in connection with commercialization. The adaptability of nanocomposite textile NGs into complete fabrics is very limited, and primarily researchers have only integrated the materials at joints in the body where clear displacement of layers can occur. On the other hand, nanocomposite-based core spun yarn-based woven or knitted NGs offer good adaptability throughout the fabrics, and they have the simplest route to scale-up its output performance in terms of output power, washability and reliability. These will also create facilitate conduction with wearable smart sensors. However, they have currently the lowest output performance and it is very difficult to produce core-spun yarns by using existing technology. Researchers need to concentrate on these issues to scale up the small-scale development of nanocomposite textile NGs to large-scale industrial production.

Nanocomposite-based textile Piezoelectric Nanogenerators (PENGs) and Triboelectric Nanogenerators (TENGs) exhibit promising capabilities for generating substantial power in wearable smart sensing and device applications. However, they encounter several challenges, including low electrical output performance, non-uniform filler distribution within the polymer matrix, washability, durability, stability, and energy conversion efficiency. Ensuring a consistent and reliable electrical output over time is a pressing concern, necessitating strategies such as optimizing electrode design, integrating efficient power management circuits, and addressing material degradation. The issue of washability presents another hurdle, as these devices must maintain functionality after washing, demanding water-resistant materials and compatibility with detergents. Furthermore, enhancing energy conversion efficiency and progressing toward industrial-scale production are critical prospects. In summary, effectively tackling these challenges is essential for harnessing the potential of nanocomposite textile based NGs in the realm of wearable energy harvesting applications. Fig. 16 describes the current challenges and prospects for nanocomposite textile based piezoelectric nanogenerators (PENGs) and triboelectric nanogenerators (TENGs).



Fig. 16. Schematic illustration of current challenges and prospects for nanocomposite textile based piezoelectric nanogenerators (PENGs) and triboelectric nanogenerators (TENGs).

# 7. Prospects

In this review, we explored the potential of wearable nanocomposite textile-based piezoelectric and triboelectric nanogenerators for mechanical energy harvesting and smart sensing applications. We discussed the latest advancements in piezoelectric and triboelectric materials that enable the development of wearable nanocomposite textile NGs. We then presented a comprehensive overview of nanocomposite textiles used in piezoelectric nanogenerators (single fibers or yarns/fabrics/ electrospun webs) and triboelectric nanogenerators (single fibers or yarns/fabrics/electrospun webs). Additionally, we covered textile-based PENG and TENG sensing and electronic applications for smart sensing purposes. Consequently, we next identify some exciting future opportunities and potential applications for nanocomposite textile NGs in wearable sensing and monitoring.

# 7.1. Progress in the development of nanostructured composite-nanofiber materials

The development of nanostructured composite-nanofiber materials holds great prospects across various fields due to nanostructured composite-nanofiber materials offering improved mechanical properties compared to traditional materials. By incorporating nanofibers into composite matrices, it is possible to enhance strength, stiffness, toughness, and fatigue resistance. This makes them attractive for applications in structural engineering, aerospace, automotive, and sports equipment, where lightweight materials with high mechanical performance are desired. For example, by incorporating nanoparticles or nanowires into nanofibers, it is possible to create materials with unique properties, such as enhanced conductivity, optical properties, or catalytic activity. These functional nanocomposite-nanofiber materials have applications in electronics, energy storage, sensors, and catalysis. PVDF exhibits exceptional piezoelectric activity, making it a preferred material for the electrospinning process of nanofibers. Additionally, materials like nylon and PTFE possess favorable triboelectric properties, leading to their extensive use in nanofiber materials. However, there is a need to go beyond the selection of single-contact materials and focus on developing biocompatible metal oxide/polymer nanocomposite-based fibers with uniform nanostructure orientation throughout the fibers. Moreover, there is a strong demand for the production of core electrospun composite fibers that possess not only triboelectric and piezoelectric

properties but also properties such as mechanical flexibility, biocompatibility, durability, and washability. In the context of textile nanogenerators (NGs), a key objective is to develop core electrodes with porosity and conductivity in order to enhance charge extraction and electron carrying capacity. By modifying the surface chemistry or topography of nanofibers, it is possible to achieve desired functionalities such as hydrophobicity, anti-fouling properties, or bioactive surfaces. This opens up opportunities in areas like filtration, tissue engineering, drug delivery, and biosensors. To fully realize the prospects of nanostructured composite-nanofiber materials, ongoing research focuses on advancing fabrication techniques, optimizing material properties, and exploring novel applications. This includes advancements in electrospinning, self-assembly, nanocomposite synthesis, and scalable manufacturing methods.

### 7.2. Smart sensing capability of nanocomposite textile based NGs

The field of Smart Sensing Nanotextiles (SSN) is an emerging area that requires support to enhance wearable human healthcare monitoring systems, addressing the health challenges faced by the growing global population [178]. Wearable sensing devices play a crucial role in

tracking health and treating illnesses by enabling real-time monitoring of vital body parameters. Nanocomposite textile based NGs can be integrated into wearable garments or medical textiles to monitor vital signs and physiological parameters. The NGs can sense and convert mechanical motions, such as body movements or muscle contractions, into electrical energy, enabling continuous health monitoring. This technology has the potential to revolutionize personalized healthcare by providing real-time data on ECG, heart rate, respiration rate, body posture, and muscle activity. The effectiveness of real-time health monitoring relies on wearable sensors and electronics, which provide reliable data for healthcare data analytics platforms. Nanocomposite textile based NGs can be employed for environmental sensing applications. By incorporating sensors that respond to specific environmental factors like temperature, humidity, pressure, or chemical exposure, these NGs can provide real-time data on the surrounding environment. This can be valuable for applications such as pollution monitoring, workplace safety, and smart buildings. By embedding sensors into textiles, NGs can detect touch, pressure, or gestures, allowing for intuitive and natural interaction with electronic devices or virtual interfaces. This can have applications in virtual reality, augmented reality, gaming, and human-computer interfaces. Further, by integrating NGs into textiles



Fig. 17. Principle of the self-charge excitation triboelectric nanogenerator. (a) The fundamental scheme of the self-charge excitation triboelectric nanogenerator (SCE-TENG), the auto-switch can change capacitors from parallel to serial connection during the operation cycle. (b) The systematic electric circuit scheme of the SCE-TENG. (c) The input/output node and scheme of self-voltage-multiplying circuit (SVMC). (d) The charge distribution of SCE-TENG in initial state. (e–g) Charge excitation process during period contact-separation cycle. (h, i) Air breakdown caused by high charge density and discharge protection circuit, respectively [179]. All essential copyrights and permissions received.

used in structural components, it is possible to detect and monitor mechanical stresses, vibrations, or deformations in real-time. This can contribute to early damage detection, predictive maintenance, and ensuring the structural integrity of critical systems. Nanocomposite textile based NGs can monitor and analyze human performance in sports, fitness, and rehabilitation applications. By sensing and analyzing body movements, NGs can provide data on biomechanics, gait analyzis, posture, and muscle activity. This information can be utilized for performance optimization, injury prevention, and rehabilitation programs. To realize the full potential of smart sensing capabilities in nanocomposite textile based NGs, ongoing research focuses on improving the sensitivity, selectivity, and reliability of the embedded sensors, developing flexible and stretchable electronics, optimizing integration techniques, and enhancing data processing algorithms.

# 7.3. Design and development charge feedback and pumping mechanisms

Reliable and stable energy output from NGs are essential requirements for the wearable device in real time scenarios. In order to generate a reliable output, there should be frequent mechanical tractions or force in between the contact layers.

However, continuous mechanical friction is less in wearable fabrics. and this will reduce NG output performance; thereby, limiting real-time wearable applications. In order to enhance the flow of continuous charge density, much research has been focused on materials selection, surface modification, contact improvement, and charge storage system, which can increase the charge density up to some extent. However, very recently a self and external charge pump method has been reported for thin film based TENGs, referring to electromagnetic generators and op amp-based feedback amplifier principles which is promising [179] (Fig. 17). In this comprehensive review, we have thoroughly examined the charge feedback mechanism as a means of achieving self-excitation in nanocomposite textile based NGs. This approach holds great promise for achieving both higher and more stable electrical output by seamlessly integrating electronic components into the textile structure. Addressing critical challenges like lower electrical output performance, this technique amplifies the output signal, ultimately enhancing its stability for wearable devices.

# 7.4. Integration of ML assisted IoT system with nanocomposite Textile based NGs

The building blocks of textiles are fibers and yarns; their outstanding chemical, electric, and mechanical properties enable these textiles to be used for sensing and NGs for wearable, self-driving electronics, and wireless sensor system applications. Whilst self-powered smart textile devices are attractive; they also come with challenges such as performance degradation with time. In this regard, machine learning-based predictive systems can be attractive as they can reveal the hidden relationship between the body parameters and the sensing signals through data transformations, normalization, elimination of baseline drifts, and compression. Additionally, they can cut down sensor optimization costs and time, provide unexpected insights into the experimental data, and predict the outcome by deciphering non-linear analytical input signals without mathematical fitting, readily integrated into an IoT setting. ML algorithms also facilitate adaptive energy management in IoT systems powered by nanocomposite textile based NGs. These algorithms can dynamically allocate and prioritize the available energy to different IoT devices based on their energy requirements, usage patterns, and priority levels. This ensures efficient utilization of the harvested energy and extends the operational lifetime of the IoT system. By analysing patterns and trends in the sensor data, ML models can identify early signs of wear, degradation, or potential failures in the NGs. This enables proactive maintenance actions, minimizing downtime and ensuring continuous operation of the IoT system. Hence ML systems enabled with context-aware applications by utilizing data from

nanocomposite textile-based NGs and other sensors within the IoT system will be desirable. By analyzing the sensor data in real-time, ML models can understand the context of the user or the environment, enabling personalized and adaptive functionalities. For example, ML algorithms can interpret body movement patterns captured by NGs to provide context-aware activity tracking or gesture recognition. Thus, it will lead to enhanced user experiences. This allows for personalized recommendations, intelligent automation, and tailored feedback, improving the overall usability and satisfaction of the IoT system. To leverage the benefits of integrating ML-assisted IoT systems with nanocomposite textile-based NGs, it is crucial to address challenges such as data privacy and security, computational constraints in resourcelimited devices, and the need for robust and scalable ML models.

In this section, we investigated the potential of wearable nanocomposite textile-based piezoelectric and triboelectric nanogenerators (PENGs and TENGs) for mechanical energy harvesting and smart sensing. We discusseed advancements in materials, such as incorporating nanofibers into composites to improve mechanical properties and create materials with unique attributes like enhanced conductivity. The potential applications span various fields, including structural engineering, aerospace, environmental sensing, and healthcare monitoring. Smart Sensing Nanotextiles (SSN) were introduced as a promising area for real-time health monitoring and environmental sensing. Additionally, the text highlighted the importance of addressing challenges in achieving reliable energy output, such as developing charge feedback mechanisms. Furthermore, the integration of Machine Learning (ML) with IoT systems powered by nanocomposite textile based NGs was discussed, emphasizing its potential for personalized recommendations, adaptive energy management, and proactive maintenance. Challenges like data privacy and computational constraints were noted as areas requiring attention to fully leverage ML in these applications.

# 8. Conclusion

With the advancement and miniaturization of technology, demand for self-powered wearable and portable electronics has risen dramatically in recent years. Although batteries are one of the most promising power sources for those applications in terms of power supply, the limited life span, bulkiness, and environmental concerns make them less than ideal. Therefore, in recent years, wearable textile based nanogenerators (piezoelectric and triboelectric) have been proposed as a good solution to directly harness the energy available in daily human motions and remove the need for batteries. However, pure textile based triboelectric nanogenerators still generate relatively small output signals due to the small active contact area at textile interfaces. In contrast, wearable nanocomposite textile based triboelectric nanogenerators could be a great option for supplying power for wearable electronic goods. Nanocomposite textiles can have higher dielectric constants and higher active contact area, and this can boost output for nanocomposite textile based TENGs. Nanocomposite composition of fibres has also been shown to increase the strength and durability of fibres. However, there still remains many challenges and these include comparatively low stability, low durability, low electrical output and lack of standards. Commercialization and power management are also issues that need to be addressed for nanocomposite TENG designs. The present review started by exploring the basics of textile materials as well as the concept and operation of piezoelectric and triboelectric nanogenerators. It then explored the many recent advances that have been made in nanocomposite textiles (fibre, yarn, fabric and electrospun web based) for piezoelectric and triboelectric nanogenerators. Finally, present challenges and their future outlook have also been identified. It is almost ten years since the publication of the first paper proposing the textile TENG in 2014 [180]. In this time, output has improved but still lags significantly behind that of solid film based TENGs. In summary, low power and current output and challenges like durability, wearability and washability remain major obstacles to widespread or commercial usage.

Nanocomposite fibres have shown promise in mitigating some of these issues and more development is expected in the next 10 years.

### CRediT authorship contribution statement

Satyaranjan Bairagi: Conceptualization, Investigation, Methodology, Software, Validation, Formal analysis, Writing – original draft, Writing – review & editing. Shahid-ul-Islam: Investigation, Methodology, Software, Validation, Writing – review and editing. Charchit Kumar: Investigation, Methodology, Software, Validation, Writing – review & editing. Aswathy Babu: Investigation, Methodology, Software, Validation, Writing – review & editing. Akshaya Kumar Aliyana: Investigation, Methodology, Software, Validation, Writing – review & editing. George Stylios: Investigation, Methodology, Software, Validation, Writing – review & editing. Suresh C. Pillai: Investigation, Methodology, Software, Validation, Writing – review & editing. Daniel M. Mulvihill: Lead Supervision, Conceptualization, Resources, Writing – original draft, Writing – review & editing, Project administration, 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

Data will be made available on request.

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#### References

- [1] Y. Yang, H. Zhang, G. Zhu, S. Lee, Z.H. Lin, Z.L. Wang, Flexible hybrid energy cell for simultaneously harvesting thermal, mechanical, and solar energies, ACS Nano 7 (2013) 785–790, https://doi.org/10.1021/nn305247x.
- [2] S. Garain, T. Kumar Sinha, P. Adhikary, K. Henkel, S. Sen, S. Ram, C. Sinha, D. Schmeißer, D. Mandal, Self-poled transparent and flexible UV light-emitting cerium complex-PVDF composite: a high-performance nanogenerator, ACS Appl. Mater. Interfaces 7 (2015) 1298–1307, https://doi.org/10.1021/am507522r.
- [3] P. Adhikary, S. Garain, D. Mandal, The co-operative performance of a hydrated salt assisted sponge like P(VDF-HFP) piezoelectric generator: an effective piezoelectric based energy harvester, Phys. Chem. Chem. Phys. 17 (2015) 7275–7281, https://doi.org/10.1039/c4cp05513f.
- [4] D. Mandal, K. Henkel, D. Schmeisser, Comment on "preparation and characterization of silver-poly(vinylidene fluoride) nanocomposites: formation of piezoelectric polymorph of poly(vinylidene fluoride), J. Phys. Chem. B. 115 (2011) 10567–10569, https://doi.org/10.1021/jp201335j.
- [5] S. Bairagi, S.W. Ali, Effects of surface modification on electrical properties of KNN nanorod-incorporated PVDF composites, J. Mater. Sci. 54 (2019) 11462–11484, https://doi.org/10.1007/s10853-019-03719-x.
- [6] V.S. Mallela, V. Ilankumaran, S.N. Rao, Trends in cardiac pacemaker batteries, Indian Pacing Electrophysiol. J. 4 (2004) 201–212.
- [7] R. Riemer, A. Shapiro, Biomechanical energy harvesting from human motion: theory, state of the art, design guidelines, and future directions, J. Neuroeng. Rehabil. 8 (2011) 1–13, https://doi.org/10.1186/1743-0003-8-22.
- [8] X. Peng, K. Dong, Y. Zhang, L. Wang, C. Wei, T. Lv, Z.L. Wang, Z. Wu, Sweatpermeable, biodegradable, transparent and self-powered chitosan-based electronic skin with ultrathin elastic gold nanofibers, Adv. Funct. Mater. 32 (2022), https://doi.org/10.1002/adfm.202112241.
- [9] C. Dagdeviren, B.D. Yang, Y. Su, P.L. Tran, P. Joe, E. Anderson, J. Xia, V. Doraiswamy, B. Dehdashti, X. Feng, B. Lu, R. Poston, Z. Khalpey, R. Ghaffari, Y. Huang, M.J. Slepian, J.A. Rogers, Conformal piezoelectric energy harvesting and storage from motions of the heart, lung, and diaphragm, Proc. Natl. Acad. Sci. U. S. A. 111 (2014) 1927–1932, https://doi.org/10.1073/pnas.1317233111.
- [10] C. Dagdeviren, S.W. Hwang, Y. Su, S. Kim, H. Cheng, O. Gur, R. Haney, F. G. Omenetto, Y. Huang, J.A. Rogers, Transient, biocompatible electronics and

energy harvesters based on ZnO, Small 9 (2013) 3398-3404, https://doi.org/10.1002/smll.201300146.

- [11] Z.L. Wang, Triboelectric nanogenerators as new energy technology for selfpowered systems and as active mechanical and chemical sensors, ACS Nano 7 (2013) 9533–9557.
- [12] A. Ali, Y. Chen, H. Liu, L. Yu, Z. Baloch, S. Khalid, J. Zhu, L. Chen, Starch-based antimicrobial films functionalized by pomegranate peel, Int. J. Biol. Macromol. 129 (2019) 1120–1126, https://doi.org/10.1016/j.ijbiomac.2018.09.068.
- [13] Z.L. Wang, J. Chen, L. Lin, Progress in triboelectric nanogenerators as a new energy technology and self-powered sensors, Energy Environ. Sci. 8 (2015) 2250–2282.
- [14] S.J. Kim, C.J. Lee, A.Y. Choi, J. Park, C. Choi, H.J. Sim, Y.T. Kim, Erratum: triboelectric generator for wearable devices fabricated using a casting method (RSC Advances (2016) 6 (10094-10098)), RSC Adv. 6 (2016) 14223, https://doi. org/10.1039/c6ra90010k.
- [15] V. Leonov, R.J.M. Vullers, Wearable thermoelectric generators for body-powered devices, J. Electron. Mater. 38 (2009) 1491–1498, https://doi.org/10.1007/ s11664-008-0638-6.
- [16] Y.K. Ramadass, A.P. Chandrakasan, A battery-less thermoelectric energy harvesting interface circuit with 35 mV startup voltage, IEEE J. Solid-State Circuits 46 (2011) 333–341, https://doi.org/10.1109/JSSC.2010.2074090.
- [17] D. Das, P. Kalita, O. Roy, Flat plate hybrid photovoltaic- thermal (PV/T) system: a review on design and development, Renew. Sustain. Energy Rev. 84 (2018) 111–130, https://doi.org/10.1016/j.rser.2018.01.002.
- [18] D. Das, U. Bordoloi, H.H. Muigai, P. Kalita, A novel form stable PCM based bio composite material for solar thermal energy storage applications, J. Energy Storage 30 (2020), 101403, https://doi.org/10.1016/j.est.2020.101403.
- [19] Y. Mao, P. Zhao, G. McConohy, H. Yang, Y. Tong, X. Wang, Sponge-like piezoelectric polymer films for scalable and integratable nanogenerators and selfpowered electronic systems, Adv. Energy Mater. 4 (2014) 1–7, https://doi.org/ 10.1002/aenm.201301624.
- [20] J. Park, A.Y. Choi, C.J. Lee, D. Kim, Y.T. Kim, Highly stretchable fiber-based single-electrode triboelectric nanogenerator for wearable devices, RSC Adv. 7 (2017) 54829–54834, https://doi.org/10.1039/c7ra10285b.
- [21] V.U. Somkuwar, A. Pragya, B. Kumar, Structurally engineered textile-based triboelectric nanogenerator for energy harvesting application, J. Mater. Sci. 55 (2020) 5177–5189, https://doi.org/10.1007/s10853-020-04359-2.
- [22] G. Zhu, C. Pan, W. Guo, C.Y. Chen, Y. Zhou, R. Yu, Z.L. Wang, Triboelectricgenerator-driven pulse electrodeposition for micropatterning, Nano Lett. 12 (2012) 4960–4965, https://doi.org/10.1021/nl302560k.
- [23] Z.L. Wang, Triboelectric nanogenerators as new energy technology and selfpowered sensors - principles, problems and perspectives, Faraday Discuss. 176 (2014) 447–458, https://doi.org/10.1039/c4fd00159a.
- [24] S. Lee, W. Ko, Y. Oh, J. Lee, G. Baek, Y. Lee, J. Sohn, S. Cha, J. Kim, J. Park, J. Hong, Triboelectric energy harvester based on wearable textile platforms employing various surface morphologies, Nano Energy 12 (2015) 410–418, https://doi.org/10.1016/j.nanoen.2015.01.009.
- [25] A.Y. Choi, C.J. Lee, J. Park, D. Kim, Y.T. Kim, Corrugated textile based triboelectric generator for wearable energy harvesting, Sci. Rep. 7 (2017) 7–12, https://doi.org/10.1038/srep45583.
- [26] C. Kumar, J. Perris, S. Bairagi, G. Min, Y. Xu, N. Gadegaard, D.M. Mulvihill, Multiscale in-situ quantification of the role of surface roughness and contact area using a novel Mica-PVS triboelectric nanogenerator, Nano Energy 107 (2023), 108122, https://doi.org/10.1016/j.nanoen.2022.108122.
- [27] X. Pu, L. Li, H. Song, C. Du, Z. Zhao, C. Jiang, G. Cao, W. Hu, Z.L. Wang, A self-charging power unit by integration of a textile triboelectric nanogenerator and a flexible lithium-ion battery for wearable electronics, Adv. Mater. 27 (2015) 2472–2478.
- [28] W. Yang, J. Chen, G. Zhu, J. Yang, P. Bai, Y. Su, Q. Jing, Harvest. Energy Nat. Vib. Hum. Walk. (2013) 11317–11324.
- [29] F. Yi, X. Wang, S. Niu, S. Li, Y. Yin, K. Dai, G. Zhang, L. Lin, Z. Wen, H. Guo, J. Wang, M. Yeh, Y. Zi, Q. Liao, Z. You, Y. Zhang, Z.L. Wang, A highly shape-adaptive, stretchable design based on conductive liquid for energy harvesting and self-powered biomechanical monitoring, (2016).
- [30] Y. Zi, S. Niu, J. Wang, Z. Wen, W. Tang, Z.L. Wang, Standards and figure-of-merits for quantifying the performance of triboelectric nanogenerators, Nat. Commun. 6 (2015), https://doi.org/10.1038/ncomms9376.
- [31] K. Dong, Z. Wu, J. Deng, A.C. Wang, H. Zou, C. Chen, D. Hu, B. Gu, B. Sun, Z. L. Wang, A stretchable yarn embedded triboelectric nanogenerator as electronic skin for biomechanical energy harvesting and multifunctional pressure sensing, Adv. Mater. 30 (2018) 1–12, https://doi.org/10.1002/adma.201804944.
- [32] P. Xu, T. Gu, Z. Cao, B. Wei, J. Yu, F. Li, J.H. Byun, W. Lu, Q. Li, T.W. Chou, Carbon nanotube fiber based stretchable wire-shaped supercapacitors, Adv. Energy Mater. 4 (2014), https://doi.org/10.1002/aenm.201300759.
  [33] X. He, Y. Zi, H. Guo, H. Zheng, Y. Xi, C. Wu, J. Wang, W. Zhang, C. Lu, Z.L. Wang,
- [33] X. He, Y. Zi, H. Guo, H. Zheng, Y. Xi, C. Wu, J. Wang, W. Zhang, C. Lu, Z.L. Wang, A highly stretchable fiber-based triboelectric nanogenerator for self-powered wearable electronics, Adv. Funct. Mater. 27 (2017), https://doi.org/10.1002/ adfm.201604378.
- [34] J. Zhong, Y. Zhang, Q. Zhong, Q. Hu, B. Hu, Z.L. Wang, J. Zhou, Fiber-based generator for wearable electronics and mobile medication, ACS Nano 8 (2014) 6273–6280, https://doi.org/10.1021/nn501732z.
- [35] X. Li, Z.H. Lin, G. Cheng, X. Wen, Y. Liu, S. Niu, Z.L. Wang, 3D fiber-based hybrid nanogenerator for energy harvesting and as a self-powered pressure sensor, ACS Nano 8 (2014) 10674–10681, https://doi.org/10.1021/nn504243j.
- [36] P. Zhang, L. Deng, H. Zhang, J. He, X. Fan, Y. Ma, Enhanced performance of triboelectric nanogenerator with micro-rhombic patterned PDMS for self-powered

wearable sensing, Adv. Mater. Interfaces 9 (2022) 1-8, https://doi.org/10.1002/admi.202201265.

- [37] K. Shi, X. Huang, B. Sun, Z. Wu, J. He, P. Jiang, Nano Energy Cellulose / BaTiO 3 aerogel paper based fl exible piezoelectric nanogenerators and the electric coupling with triboelectricity, Nano Energy 57 (2019) 450–458, https://doi.org/ 10.1016/j.nanoen.2018.12.076.
- [38] S. Tian, X. Wei, L. Lai, B. Li, Z. Wu, Y. Dai, Frequency modulated hybrid nanogenerator for efficient water wave energy harvesting, Nano Energy 102 (2022), 107669, https://doi.org/10.1016/j.nanoen.2022.107669.
- [39] S. Bairagi, S.W. Ali, A unique piezoelectric nanogenerator composed of melt-spun PVDF/KNN nanorod-based nanocomposite fibre, Eur. Polym. J. 116 (2019) 554–561, https://doi.org/10.1016/j.eurpolymj.2019.04.043.
- [40] M. Zhang, T. Gao, J. Wang, J. Liao, Y. Qiu, Q. Yang, H. Xue, Z. Shi, Y. Zhao, Z. Xiong, L. Chen, A hybrid fibers based wearable fabric piezoelectric nanogenerator for energy harvesting application, Nano Energy 13 (2015) 298–305, https://doi.org/10.1016/j.nanoen.2015.02.034.
- [41] S. Bairagi, S.W. Ali, Influence of high aspect ratio lead-free piezoelectric fillers in designing flexible fibrous nanogenerators: demonstration of significant high output voltage, Energy Technol. 7 (2019) 1–10, https://doi.org/10.1002/ ente.201900538.
- [42] L. Shi, H. Jin, S. Dong, S. Huang, H. Kuang, H. Xu, J. Chen, W. Xuan, S. Zhang, S. Li, X. Wang, J. Luo, High-performance triboelectric nanogenerator based on electrospun PVDF-graphene nanosheet composite nanofibers for energy harvesting, Nano Energy 80 (2021), 105599, https://doi.org/10.1016/j. nanoen.2020.105599.
- [43] J. Xiong, P. Cui, X. Chen, J. Wang, K. Parida, M.F. Lin, P.S. Lee, Skin-touchactuated textile-based triboelectric nanogenerator with black phosphorus for durable biomechanical energy harvesting, Nat. Commun. 9 (2018) 1–9, https:// doi.org/10.1038/s41467-018-06759-0.
- [44] Y. Cheng, X. Lu, K. Hoe Chan, R. Wang, Z. Cao, J. Sun, G. Wei Ho, A stretchable fiber nanogenerator for versatile mechanical energy harvesting and self-powered full-range personal healthcare monitoring, Nano Energy 41 (2017) 511–518, https://doi.org/10.1016/j.nanoen.2017.10.010.
- [45] J. Curie, Piezoelectricity 5 (1920), https://doi.org/10.1016/B978-0-12-818835-4.00005-5.
- [46] L. G, Principe de la conservation de l'elecricité, Ann. Chem. e Phys. (1881) 1881.
  [47] D. Damjanovic, Piezoelectricity, Encycl, Condens. Matter Phys. (2005) 300–309, https://doi.org/10.1016/B0-12-369401-9/00433-2.
- [48] M. Smith, S. Kar-Narayan, Piezoelectric polymers: theory, challenges and opportunities, Int. Mater. Rev. 67 (2022) 65–88, https://doi.org/10.1080/ 09506608.2021.1915935.
- [49] A. Ledoux, J.J. Connor, Theory of Piezoelectric Materials and Their Applications in Civil Engineering By Certified by: Theory of Piezoelectric Materials and Their Applications in Civil Engineering By Abstract, (2011).
- [50] J. Takahashi, Introd. Theory Soc. Control (1960), https://doi.org/10.4057/ jsr.11.2 51.
- [51] IEC 60050 International Electrotechnical Vocabulary Details for IEV number 121–11-40: "electric flux density," (n.d.). (https://www.electropedia.org/iev/iev. nsf/display?openform&ievref=121–11-40) (accessed February 23, 2023).
- [52] Fundamentals of Piezoelectricity (Oxford Science Publications) by Ikeda, Takuro, (n.d.). (https://www.biblio.com/book/fundamentals-piezoelectricity -oxford-science-publications-ikeda/d/1174583361) (accessed February 23, 2023)
- [53] R.S. Dahiya, M. Valle, Robotic tactile sensing: technologies and system, Robot. Tactile Sens. Technol. Syst. 9789400705 (2014) 1–245, https://doi.org/10.1007/ 978-94-007-0579-1.
- [54] Y. Wang, Y. Yang, Z.L. Wang, Triboelectric nanogenerators as fl exible power sources, Npj Flex. Electron 2017 (2017) 1–9, https://doi.org/10.1038/s41528-017-0007-8.
- [55] Z.L. Wang, On Maxwell's displacement current for energy and sensors: the origin of nanogenerators, Mater. Today 20 (2017) 74–82, https://doi.org/10.1016/j. mattod.2016.12.001.
- [56] R. Zhang, H. Olin, Material choices for triboelectric nanogenerators: a critical review, EcoMat 2 (2020), e12062.
- [57] S. Gokhool, S. Bairagi, C. Kumar, D.M. Mulvihill, Reflections on Boosting Wearable Triboelectric Nanogenerator Performance via Interface Optimisation, (n.d.).
- [58] K. Dong, X. Peng, Z.L. Wang, Fiber/fabric-based piezoelectric and triboelectric nanogenerators for flexible/stretchable and wearable electronics and artificial intelligence, Adv. Mater. 32 (2020) 1–43, https://doi.org/10.1002/ adma.201902549.
- [59] S. Bairagi, M. Shahadat, D.M. Mulvihill, W. Ali, Nano Energy Mechanical energy harvesting and self-powered electronic applications of textile-based piezoelectric nanogenerators: a systematic review, Nano Energy 111 (2023), 108414, https:// doi.org/10.1016/j.nanoen.2023.108414.
- [60] A. Babu, I. Aazem, R. Walden, S. Bairagi, D.M. Mulvihill, C. Suresh, Multifunctional triboelectric nanogenerators fabricated by electrospinning methods, (n.d.) 1–63.
- [61] G. Khandelwal, R. Dahiya, Self-powered active sensing based on triboelectric generators, Adv. Mater. 34 (2022), https://doi.org/10.1002/adma.202200724.
- [62] H. Wang, A. Jasim, Piezoelectric energy harvesting from pavement, LTD (2020), https://doi.org/10.1016/B978-0-12-818981-8.00014-X.
- [63] S. Banerjee, S. Bairagi, S. Wazed Ali, A critical review on lead-free hybrid materials for next generation piezoelectric energy harvesting and conversion, Ceram. Int. (2021), https://doi.org/10.1016/j.ceramint.2021.03.054.

- [64] S.W. Ali, S. Bairagi, P. Shankar, Biomaterials-Based Nanogenerator: Futuristic Solution for Integration Into Smart Textiles, (n.d.) 189–201.
- [65] P. Devices, U. Aluminum, F. May, FEZOSELECTRC EDEVICES (UTLANG ALURAINURANTERIDE, (1963) 1–4.
- [66] W.R. Cook, D.A. Berlincourt, F.J. Scholz, Thermal expansion and pyroelectricity in lead titanate zirconate and barium titanate, J. Appl. Phys. 34 (1963) 1392–1398, https://doi.org/10.1063/1.1729587.
- [67] A.W. Warner, M. Onoe, G.A. Coquin, Determination of elastic and piezoelectric constants for crystals in class (3 m), J. Acoust. Soc. Am. 42 (1967) 1223–1231, https://doi.org/10.1121/1.1910709.
- [68] T. Yamada, H. Iwasaki, N. Niizeki, Piezoelectric and elastic properties of LiTaO 3: temperature characteristics, Jpn. J. Appl. Phys. 8 (1969) 1127, https://doi.org/ 10.1143/jjap.8.1127.
- [69] D.O.N. Berlincourt, H. Japze, Elastic and Piezoelectric Coefficients of Single-Crystal Barium Titanate, 108 (1958) 143–148.
- [70] D.W. Xianwu Tang, Jianming Dai, Xuebin Zhu, Jianchao Lin, Qing Chang, Y. S. Wenhai Song, Thickness-dependent dielectric, ferroelectric, and magnetodielectric properties of BiFeO 3 thin films derived by chemical solution depositio, J. Am. Ceram. Soc. 544 (2012) 538–544, https://doi.org/10.1111/j.1551-2916.2011.04920.x.
- [71] K. Uchino, Gen. View Ferroelectr. (2019).
- [72] Q.M. Zhang, J. Zhao, Electromechanical Properties of Lead Zirconate Titanate Piezoceramics Under the Influence of Mechanical Stresses, 46 (1999) 1518–1526.
- [73] W. and K.Z. Xuming Pang, Jinhao Qiu, Morphotropic phase boundary of sodium–potassium niobate lead-free piezoelectric ceramicsmorphotropic phase boundary of sodium–potassium niobate lead-free piezoelectric ceramics, J. Am. Ceram. Soc. 801 (2011) 796–801, https://doi.org/10.1111/j.1551-2916.2010.04143.x.
- [74] H.-Y. Park, et al., Microstructure and piezoelectric properties of 0.95(Na0.5K0.5) NbO3–0.05BaTiO3 ceramics, Appl. Phys. Lett. 95 (2021) 3–6, https://doi.org/ 10.1063/1.2335816.
- [75] S.C. List, Piezoelectricity in polyvinylidenefluoride, J. Acoust. Soc. Am. 1608 (1981) 1596–1608.
- [76] H.S. Nalwa, Recent developments in ferroelectric polymers, J. Macromol. Sci. Part C. 31 (1991) 341–432, https://doi.org/10.1080/15321799108021957.
- [77] F. Mokhtari, Z. Cheng, C.H. Wang, J. Foroughi, Advances in Wearable Piezoelectric Sensors for Hazardous Workplace Environments, 2300019 (2023). https://doi.org/10.1002/gch2.202300019.
- [78] S. Wang, L. Lin, Z.L. Wang, Triboelectric nanogenerators as self-powered active sensors, Nano Energy 11 (2015) 436–462, https://doi.org/10.1016/j. nanoen.2014.10.034.
- [79] K. Dong, Z.L. Wang, Self-charging power textiles integrating energy harvesting triboelectric nanogenerators with energy storage batteries/supercapacitors, J. Semicond. 42 (2021), https://doi.org/10.1088/1674-4926/42/10/101601.
- [80] S. Liu, T. Hua, X. Luo, N. Yi Lam, X. Ming Tao, L. Li, A novel approach to improving the quality of chitosan blended yarns using static theory, Text. Res. J. 85 (2015) 1022–1034, https://doi.org/10.1177/0040517514559576.
- [81] S. Of, T. Fibres, Sources of Textile Fibres Definition of Terms, (n.d.).
- [82] Basics of Textile Fibers Textile Blog, (n.d.). (https://www.textileblog.co m/basics-of-textile-fibers/) (accessed April 22, 2023).
- [83] Textile Fibres Cass Materials, (n.d.). (https://cassmaterials.com/fibres/) (accessed August 29, 2023).
- [84] No.104 Thin Natural Cotton Twine, (n.d.). (https://henrywinning.co.uk/products /no-104-thin-natural-cotton-twine?currency=GBP&variant=32447054479540 &gclid=CjwKCAjwrranBhAEEiwAzbhNtWllJvy9JnsuRxojmxsif8ctYtxw nhlEqj9ikyw755Jx2OH3xHPouhoCX4gQAvD\_BwE> (accessed August 29, 2023).
- [85] Terms used in Balloon Fabric Creation, (n.d.). (https://balloonhq.com/faq/weav e-faq/terms-sources/joseph/) (accessed August 29, 2023).
- [86] 160cm 60g Spunbond Polypropylene Non Woven Fabric, (n.d.). (https://www. mh-chine.com/products/white-60gsm-recycled-polypropylene-nonwoven-fabri c-6405–0151) (accessed August 29, 2023).
- [87] Knitting Terms and Definition | Textile Study Center, (n.d.). (https://textilest udycenter.com/knitting-terms-and-definition/) (accessed September 4, 2023).
- [88] B.D. Malhotra, M.A. Ali, Nanomaterials for biosensors: fundamentals and applications, Nanomater. Biosens. Fundam. Appl. (2017) 1–321, https://doi.org/ 10.1016/C2015-0-04697-4.
- [89] D. Karst, Y. Yang, Potential advantages and risks of nanotechnology for textiles, AATCC Rev. 6 (2006) 44–48.
- [90] S. Riaz, M. Ashraf, T. Hussain, M.T. Hussain, A. Younus, Fabrication of robust multifaceted textiles by application of functionalized TiO2 nanoparticles, Colloids Surf. A Physicochem. Eng. Asp. 581 (2019), 123799, https://doi.org/10.1016/j. colsurfa.2019.123799.
- [91] F. Mokhtari, G.M. Spinks, C. Fay, Z. Cheng, R. Raad, J. Xi, J. Foroughi, Wearable electronic textiles from nanostructured piezoelectric fibers, Adv. Mater. Technol. 5 (2020), https://doi.org/10.1002/admt.201900900.
- [92] P. Hu, L. Yan, C. Zhao, Y. Zhang, J. Niu, Double-layer structured PVDF nanocomposite film designed for flexible nanogenerator exhibiting enhanced piezoelectric output and mechanical property, Compos. Sci. Technol. 168 (2018) 327–335, https://doi.org/10.1016/j.compscitech.2018.10.021.
- [93] K. Shi, B. Sun, X. Huang, P. Jiang, Synergistic effect of graphene nanosheet and BaTiO3 nanoparticles on performance enhancement of electrospun PVDF nanofiber mat for flexible piezoelectric nanogenerators, Nano Energy 52 (2018) 153–162, https://doi.org/10.1016/j.nanoen.2018.07.053.
- [94] F. Mokhtari, G.M. Spinks, S. Sayyar, Z. Cheng, A. Ruhparwar, J. Foroughi, Highly Stretchable Self-Powered Wearable Electrical Energy Generator and Sensors, Adv. Mater. Technol. 6 (2021), https://doi.org/10.1002/admt.202000841.

- [95] S. Bairagi, S.W. Ali, Poly (vinylidine fluoride) (PVDF)/Potassium Sodium Niobate (KNN) nanorods based flexible nanocomposite film: Influence of KNN concentration in the performance of nanogenerator, Org. Electron (2019), 105547, https://doi.org/10.1016/j.orgel.2019.105547
- [96] A. Chowdhury, S. Bairagi, S.W. Ali, B. Kumar, Leveraging shape memory coupled piezoelectric properties in melt extruded composite filament based on polyvinylidene fluoride and polyurethane, Macromol. Mater. Eng. 305 (2020) -12, https://doi.org/10.1002/mame.202000296
- [97] P. Martins, A.C. Lopes, S. Lanceros-Mendez, Electroactive phases of poly (vinylidene fluoride): Determination, processing and applications, Prog. Polym. Sci. 39 (2014) 683–706, https://doi.org/10.1016/j.progpolym
- [98] F. Mokhtari, J. Foroughi, T. Zheng, Z. Cheng, G.M. Spinks, Triaxial braided piezo fiber energy harvesters for self-powered wearable technologies, J. Mater. Chem. A. 7 (2019) 8245-8257, https://doi.org/10.1039/c8ta10964h.
- S. Rafique, A.K. Kasi, J.K. Kasi, M. Aminullah, Z. Bokhari, Shakoor, Fabrication of [99] silver-doped zinc oxide nanorods piezoelectric nanogenerator on cotton fabric to utilize and optimize the charging system, Nanomater. Nanotechnol. 10 (2020) 1-12, https://doi.org/10.1177/1847980419895741.
- [100] S. Bai, L. Zhang, Q. Xu, Y. Zheng, Y. Qin, Z.L. Wang, Two dimensional woven nanogenerator, Nano Energy 2 (2013) 749-753, https://doi.org/10.1016/j. nanoen 2013 01 001
- [101] S. Bairagi, S.W. Ali, A unique piezoelectric nanogenerator composed of melt-spun PVDF/KNN nanorod-based nanocomposite fibre, Eur. Polym. J. 116 (2019) 554-561, https://doi.org/10.1016/j.eurpolymj.2019.04.043.
- [102] F. Mokhtari, M. Latifi, M. Shamshirsaz, Electrospinning/electrospray of polyvinylidene fluoride (PVDF): Piezoelectric nanofibers, J. Text. Inst. 107 (2016) 1037-1055, https://doi.org/10.1080/00405000.2015.1083300.
- [103] V.S. Reddy, Y. Tian, C. Zhang, Z. Ye, K. Roy, A. Chinnappan, S. Ramakrishna, W. Liu, R. Ghosh, A review on electrospun nanofibers based advanced applications: from health care to energy devices, Polymers 13 (2021) 1-39, tps://doi.org/10.3390/polym13213746.
- [104] J. Xue, T. Wu, Y. Dai, Y. Xia, Electrospinning and electrospun nanofibers: Methods, materials, and applications, Chem. Rev. 119 (2019) 5298-5415, https://doi.org/10.1021/acs.chemrev.8b00593.
- [105] G.J. Kim, K.O. Kim, Novel glucose-responsive of the transparent nanofiber hydrogel patches as a wearable biosensor via electrospinning, Sci. Rep. 10 (2020) 1-12, https://doi.org/10.1038/s41598-020-75906-9.
- [106] Y. Xu, G. Shi, J. Tang, R. Cheng, X. Shen, Y. Gu, L. Wu, K. Xi, Y. Zhao, W. Cui, L. Chen, ECM-inspired micro/nanofibers for modulating cell function and tissue generation, Sci. Adv. 6 (2020) 1-18, https://doi.org/10.1126/sciadv.abc2036.
- [107] H. Ma, B.S. Hsiao, B. Chu, Functionalized electrospun nanofibrous microfiltration membranes for removal of bacteria and viruses, J. Memb. Sci. 452 (2014) 446-452, https://doi.org/10.1016/j.memsci.2013.10.047. [108] D. Sun, C. Chang, S. Li, L. Lin, -Field Electro (2006).
- [109] C. Chang, K. Limkrailassiri, L. Lin, Continuous near-field electrospinning for large area deposition of orderly nanofiber patterns, Appl. Phys. Lett. 93 (2008) 1-4, oi.org/10.1063/1.2975834
- [110] J. Fang, X. Wang, T. Lin, Electrical power generator from randomly oriented electrospun poly(vinylidene fluoride) nanofibre membranes, J. Mater. Chem. 21 (2011) 11088–11091, https://doi.org/10.1039/c1jm11445j.
- [111] J. Fang, H. Niu, H. Wang, X. Wang, T. Lin, Enhanced mechanical energy harvesting using needleless electrospun poly(vinylidene fluoride) nanofibre webs, Energy Environ. Sci. 6 (2013) 2196-2202, https://doi.org/10.1039/c3ee24230g.
- [112] S.C. Mathur, J.I. Scheinbeim, B.A. Newman, Piezoelectric properties and ferroelectric hysteresis effects in uniaxially stretched nylon-11 films, J. Appl. Phys. 56 (1984) 2419-2425, https://doi.org/10.1063/1.334294.
- [113] L. Huang, X. Zhuang, J. Hu, L. Lang, P. Zhang, Y. Wang, X. Chen, Y. Wei, X. Jing, Synthesis of biodegradable and electroactive multiblock polylactide and aniline pentamer copolymer for tissue engineering applications, Biomacromolecules 9 (2008) 850-858, https://doi.org/10.1021/bm7011828
- [114] D.J.B, J.B.T, S.A.D, D.D.H, D.J.T, D.L.W, I.C. Summerhayes, Enhanced peripheral nerve regeneration through a poled bioresorbable poly(lactic-co-glycolic acid) guidance channel, J. Neural Eng. 1 (2004) 91. (http://stacks.iop.org/1741-255 1/i=2/a=004
- [115] E. Pukada, History and recent progress in piezoelectric polymers, IEEE Trans. Ultrason. Ferroelectr. Freq. Control. 47 (2000) 1277-1290, https://doi.org/ 0.1109/58.883516.
- [116] A.J. Lovinger, Ferroelectric polymers, Science 220 (1983) 1115-1121. (http://sc nce.sciencemag.org/content/220/4602/1115.abstract
- [117] E.S. Nour, A. Khan, O. Nur, M. Willander, A flexible sandwich nanogenerator for harvesting piezoelectric potential from single crystalline zinc oxide nanowires, Nanomater. Nanotechnol. 4 (2014) 24, https://doi.org/10.5772/5906
- [118] S. Lee, B. Yeom, Y. Kim, J. Cho, Layer-by-layer assembly for ultrathin energyharvesting films: piezoelectric and triboelectric nanocomposite films, Nano Energy 56 (2019) 1-15, https://doi.org/10.1016/j.nanoen.2018.11.024.
- [119] R. Guo, Y. Guo, H. Duan, H. Li, H. Liu, Synthesis of orthorhombic perovskite-type ZnSnO3 single-crystal nanoplates and their application in energy harvesting, ACS Appl. Mater. Interfaces 9 (2017) 8271-8279, https://doi.org/10.1021/ sami.6b16629
- [120] H. Xu, M. Joung, J. Kim, S. Nahm, M. Kang, C. Kang, S. Yoon, Synthesis of homogeneous ( Na 1 À x K x) NbO 3 nanorods using hydrothermal and post-heat treatment processes, Chem. Eng. J. 211-212 (2012) 16-21, https://doi.org/ 10.1016/j.cej.2012.09.052.
- [121] M. Koç, L. Paralı, O. Şan, Fabrication and vibrational energy harvesting characterization of flexible piezoelectric nanogenerator (PEN) based on PVDF/

PZT, Polym. Test. 90 (2020), 106695, https://doi.org/10.1016/j. polymertesting.2020.10669

- [122] X. Guan, H. Chen, H. Xia, Y. Fu, J. Yao, Q.Q. Ni, Flexible energy harvester based on aligned PZT/SMPU nanofibers and shape memory effect for curved sensors, Compos. Part B Eng. 197 (2020), 108169, https://doi.org/10.1016/ compositesb.2020.108169
- [123] K. Liu, H.J. Choi, B.K. Kim, D. Bin Kim, C.S. Han, S.W. Kim, H.B. Kang, J.W. Park, Y.S. Cho, Piezoelectric energy harvesting and charging performance of Pb(Zn1/ 3Nb2/3)O3-Pb(Zr0.5Ti0.5)O3 nanoparticle-embedded P(VDF-TrFE) nanofiber composite sheets, Compos. Sci. Technol. 168 (2018) 296-302, https://doi.org/ 10.1016/j.compscitech.2018.10.012.
- [124] A. Teka, S. Bairagi, M. Shahadat, M. Joshi, S. Ziauddin Ahammad, S. Wazed Ali, Poly(vinylidene fluoride) (PVDF)/potassium sodium niobate (KNN)-based nanofibrous web: a unique nanogenerator for renewable energy harvesting and investigating the role of KNN nanostructures, Polym. Adv. Technol. 29 (2018) 2537-2544, https://doi.org/10.1002/pat.436
- [125] S. Bairagi, S.W. Ali, Influence of high aspect ratio lead-free piezoelectric filler in designing flexible fibrous nanogenerator: demonstration of significant high output voltage, Energy Technol. 7 (2019) 201900538, https://doi.org/10.1002/
- [126] S. Bairagi, S.W. Ali, Flexible lead-free PVDF/SM-KNN electrospun nanocomposite based piezoelectric materials: Significant enhancement of energy harvesting efficiency of the nanogenerator, Energy 198 (2020), 117385, https://doi.org/ 10.1016/i.energy.2020.11738
- [127] S. Banerjee, S. Bairagi, S.W. Ali, A lead-free flexible piezoelectric-triboelectric hybrid nanogenerator composed of uniquely designed PVDF/KNN-ZS nanofibrous web, Energy 244 (2022), 123102, https://doi.org/10.1016/j energy.2022.123102
- [128] C. Zhao, J. Niu, Y. Zhang, C. Li, P. Hu, Coaxially aligned MWCNTs improve performance of electrospun P(VDF-TrFE)-based fibrous membrane applied in wearable piezoelectric nanogenerator, Compos. Part B Eng. 178 (2019), 107447, https://doi.org/10.1016/j.compositesb.2019.107447
- [129] S. Bairagi, S.W. Ali, A hybrid piezoelectric nanogenerator comprising of KNN / ZnO nanorods incorporated PVDF electrospun nanocomposite webs, 44 (2020) 1-19. https://doi.org/10.1002/er.5306.
- [130] D. Ponnamma, H. Parangusan, A. Tanvir, M.A.A. AlMa'adeed, Smart and robust electrospun fabrics of piezoelectric polymer nanocomposite for self-powering electronic textiles, Mater. Des. 184 (2019), 108176, https://doi.org/10.1016/j. natdes.2019.108176.
- [131] S. Cherumannil Karumuthil, S. Prabha Rajeev, U. Valiyaneerilakkal, S. Athiyanathil, S. Varghese, Electrospun poly(vinylidene fluoridetrifluoroethylene)-based polymer nanocomposite fibers for piezoelectric nanogenerators, ACS Appl. Mater. Interfaces 11 (2019) 40180-40188, https:// doi.org/10.1021/acsami.9b17788.
- [132] H. Parangusan, D. Ponnamma, M.A.A. Al-Maadeed, Stretchable electrospun PVDF-HFP/Co-ZnO nanofibers as piezoelectric nanogenerators, Sci. Rep. 8 (2018) 1-11, https://doi.org/10.1038/s41598-017-19082-3
- [133] S. Siddiqui, D. Il Kim, E. Roh, L.T. Duy, T.Q. Trung, M.T. Nguyen, N.E. Lee, A durable and stable piezoelectric nanogenerator with nanocomposite nanofibers embedded in an elastomer under high loading for a self-powered sensor system, Nano Energy 30 (2016) 434-442, https://doi.org/10.1016/j. nanoen.2016.10.034
- [134] J. Yang, F. Xu, H. Jiang, C. Wang, X. Li, X. Zhang, G. Zhu, Piezoelectric enhancement of an electrospun AlN-doped P(VDF-TrFE) nanofiber membrane, Mater. Chem. Front. 5 (2021) 5679-5688, https://doi.org/10.1039/ d1am00550b
- [135] S. Bairagi, S.W. Ali, Investigating the role of carbon nanotubes (CNTs) in the piezoelectric performance of a PVDF/KNN-based electrospun nanogenerator, Soft Matter 16 (2020) 4876–4886, https://doi.org/10.1039/d0sm00438c
- [136] S. Bairagi, S.W. Ali, A hybrid piezoelectric nanogenerator comprising of KNN/ ZnO nanorods incorporated PVDF electrospun nanocomposite webs, Int. J. Energy Res. 44 (2020) 5545-5563, https://doi.org/10.1002/er.5306
- S. Bairagi, A. Chowdhury, S. Banerjee, A. Thakre, A. Saini, S.W. Ali, Investigating [137] the role of copper oxide (CuO) nanorods in designing flexible piezoelectric nanogenerator composed of polyacrylonitrile (PAN) electrospun web-based fibrous material, J. Mater. Sci. Mater. Electron. 33 (2022) 13152-13165, https:// doi.org/10.1007/s10854-022-08254-1.
- [138] S. Bairagi, S. Banerjee, P. Kalyaniyan, G. Dhanalakshmi, S.W. Ali, New insights toward casein/polyvinyl alcohol electrospun nanofibrous webs as a piezoelectriccum-triboelectric energy harvester, ACS Appl. Electron. Mater. 3 (2021) 4348-4361, https://doi.org/10.1021/acsaelm.1c00532.
- [139] C. Kumar, T. Speck, V. Le Houérou, Local contact formation during sliding on soft adhesive surfaces with complex microstructuring, Tribol. Int. 163 (2021) 0-2, nttps://doi.org/10.1016/j.triboint.2021.107180
- [140] Z. Bai, Z. Zhang, J. Li, J. Guo, Textile-based triboelectric nanogenerators with high-performance via optimized functional elastomer composited tribomaterials as wearable power source, Nano Energy 65 (2019), 104012, https://doi.org/ 10.1016/j.na noen.2019.104012
- [141] J. Liu, N. Cui, T. Du, G. Li, S. Liu, Q. Xu, Z. Wang, L. Gu, Y. Qin, Coaxial double helix structured fiber-based triboelectric nanogenerator for effectively harvesting mechanical energy, Nanoscale Adv. 2 (2020) 4482-4490, https://doi.org/ 10.1039/d0na0053
- [142] H.J. Sim, C. Choi, S.H. Kim, K.M. Kim, C.J. Lee, Y.T. Kim, X. Lepró, R. H. Baughman, S.J. Kim, Stretchable triboelectric fiber for self-powered kinematic sensing textile, Sci. Rep. 6 (2016) 1-7, https://doi.org/10.1038/srep35153.

- [143] J.E. Lee, Y.E. Shin, G.H. Lee, J. Kim, H. Ko, H.G. Chae, Polyvinylidene fluoride (PVDF)/cellulose nanocrystal (CNC) nanocomposite fiber and triboelectric textile sensors, Compos. Part B Eng. 223 (2021), 109098, https://doi.org/10.1016/j. compositesb.2021.109098.
- [144] M.T. Rahman, S.M.S. Rana, M. Salauddin, M.A. Zahed, S. Lee, E.S. Yoon, J. Y. Park, Silicone-incorporated nanoporous cobalt oxide and MXene nanocomposite-coated stretchable fabric for wearable triboelectric nanogenerator and self-powered sensing applications, Nano Energy 100 (2022), 107454, https:// doi.org/10.1016/j.nanoen.2022.107454.
- [145] Z. Bai, Y. Xu, J. Li, J. Zhu, C. Gao, Y. Zhang, J. Wang, J. Guo, An eco-friendly porous nanocomposite fabric-based triboelectric nanogenerator for efficient energy harvesting and motion sensing, ACS Appl. Mater. Interfaces 12 (2020) 42880–42890, https://doi.org/10.1021/acsami.0c12709.
- [146] C. Wu, T.W. Kim, F. Li, T. Guo, Wearable electricity generators fabricated utilizing transparent electronic textiles based on polyester/ag nanowires/graphene coreshell nanocomposites, ACS Nano 10 (2016) 6449–6457, https://doi.org/10.1021/ acsnano.5b08137.
- [147] P.Y. Feng, Z. Xia, B. Sun, X. Jing, H. Li, X. Tao, H.Y. Mi, Y. Liu, Enhancing the performance of fabric-based triboelectric nanogenerators by structural and chemical modification, ACS Appl. Mater. Interfaces 13 (2021) 16916–16927, https://doi.org/10.1021/acsami.1c02815.
- [148] W. Seung, M.K. Gupta, K.Y. Lee, K. Shin, J. Lee, T.Y. Kim, S. Kim, J. Lin, J.H. Kim, S. Kim, Nanopatterned Textile-Based, (2015) 3501–3509.
- [149] K. Dong, Y.C. Wang, J. Deng, Y. Dai, S.L. Zhang, H. Zou, B. Gu, B. Sun, Z.L. Wang, A highly stretchable and washable all-yarn-based self-charging knitting power textile composed of fiber triboelectric nanogenerators and supercapacitors, ACS Nano 11 (2017) 9490–9499, https://doi.org/10.1021/acsnano.7b05317.
- [150] S. Dong, P. Xu, Y. Sheng, Z. Guo, X. Pu, Y. Liu, Seamlessly knitted stretchable comfortable textile triboelectric nanogenerators for E-textile power sources, Nano Energy 78 (2020), 105327, https://doi.org/10.1016/j.nanoen.2020.105327.
- [151] X. Hu, L. Xu, X. Lin, M. Pecht, Battery lifetime prognostics, Joule 4 (2020) 310–346, https://doi.org/10.1016/j.joule.2019.11.018.
- [152] Z. Lin, B. Zhang, H. Guo, Z. Wu, H. Zou, J. Yang, Z.L. Wang, Super-robust and frequency-multiplied triboelectric nanogenerator for efficient harvesting water and wind energy, Nano Energy 64 (2019), 103908, https://doi.org/10.1016/j. nanoen.2019.103908.
- [153] Z.L. Wang, W. Wu, Nanotechnology-enabled energy harvesting for self-powered micro-/nanosystems, Angew. Chem. Int. Ed. 51 (2012) 11700–11721, https://doi. org/10.1002/anie.201201656.
- [154] Z. Sha, C. Boyer, G. Li, Y. Yu, F.-M. Allioux, K. Kalantar-Zadeh, C.-H. Wang, J. Zhang, Electrospun liquid metal/PVDF-HFP nanofiber membranes with exceptional triboelectric performance, Nano Energy 92 (2022), 106713.
- [155] Y. He, H. Wang, Z. Sha, C. Boyer, C.H. Wang, J. Zhang, Enhancing output performance of PVDF-HFP fiber-based nanogenerator by hybridizing silver nanowires and perovskite oxide nanocrystals, Nano Energy 98 (2022), 107343, https://doi.org/10.1016/j.nanoen.2022.107343.
- [156] S. Cheon, H. Kang, H. Kim, Y. Son, J.Y. Lee, H.J. Shin, S.W. Kim, J.H. Cho, Highperformance triboelectric nanogenerators based on electrospun polyvinylidene fluoride-silver nanowire composite nanofibers, Adv. Funct. Mater. 28 (2018) 1–7, https://doi.org/10.1002/adfm.201703778.
- [157] X. Pu, J.W. Zha, C.L. Zhao, S.B. Gong, J.F. Gao, R.K.Y. Li, Flexible PVDF/nylon-11 electrospun fibrous membranes with aligned ZnO nanowires as potential triboelectric nanogenerators, Chem. Eng. J. 398 (2020), 125526, https://doi.org/ 10.1016/j.cej.2020.125526.
- [158] S. Kalani, R. Kohandani, R. Bagherzadeh, Flexible electrospun PVDF-BaTiO3hybrid structure pressure sensor with enhanced efficiency, RSC Adv. 10 (2020) 35090–35098, https://doi.org/10.1039/d0ra05675h.
- [159] J.H. Zhang, Z. Zhou, J. Li, B. Shen, T. Zhu, X. Gao, R. Tao, X. Guo, X. Hu, Y. Shi, L. Pan, Coupling enhanced performance of triboelectric-piezoelectric hybrid nanogenerator based on nanoporous film of poly(vinylidene fluoride)/ BaTiO3composite electrospun fibers, ACS, Mater. Lett. 4 (2022) 847–852, https://doi.org/10.1021/acsmaterialslett.1c00819.
- [160] T. Huang, M. Lu, H. Yu, Q. Zhang, H. Wang, M. Zhu, Enhanced power output of a triboelectric nanogenerator composed of electrospun nanofiber mats doped with graphene oxide, Sci. Rep. 5 (2015) 1–8, https://doi.org/10.1038/srep13942.
- [161] T. Bhatta, P. Maharjan, H. Cho, C. Park, S.H. Yoon, S. Sharma, M. Salauddin, M. T. Rahman, S.S. Rana, J.Y. Park, High-performance triboelectric nanogenerator based on MXene functionalized polyvinylidene fluoride composite nanofibers, Nano Energy 81 (2021), 105670, https://doi.org/10.1016/j. nanoen.2020.105670.
- [162] S.M.S. Rana, M.T. Rahman, M. Salauddin, S. Sharma, P. Maharjan, T. Bhatta, H. Cho, C. Park, J.Y. Park, Electrospun PVDF-TrFE/MXene nanofiber mat-based triboelectric nanogenerator for smart home appliances, ACS Appl. Mater. Interfaces 13 (2021) 4955–4967, https://doi.org/10.1021/acsami.0c17512.
- [163] Y. Zheng, L. Cheng, M. Yuan, Z. Wang, L. Zhang, Y. Qin, T. Jing, An electrospun nanowire-based triboelectric nanogenerator and its application in a fully selfpowered UV detector, Nanoscale 6 (2014) 7842–7846, https://doi.org/10.1039/ c4nr01934b.
- [164] T. Huang, H. Yu, H. Wang, Q. Zhang, M. Zhu, Hydrophobic SiO2 electret enhances the performance of poly(vinylidene fluoride) nanofiber-based triboelectric nanogenerator, J. Phys. Chem. C. 120 (2016) 26600–26608, https://doi.org/ 10.1021/acs.jpcc.6b07382.
- [165] M. Tayyab, J. Wang, J. Wang, M. Maksutoglu, H. Yu, G. Sun, F. Yildiz, M. Eginligil, W. Huang, Enhanced output in polyvinylidene fluoride nanofibers based triboelectric nanogenerator by using printer ink as nano-fillers, Nano Energy 77 (2020), 105178, https://doi.org/10.1016/j.nanoen.2020.105178.

- [166] L. Shi, H. Jin, S. Dong, S. Huang, H. Kuang, H. Xu, J. Chen, W. Xuan, S. Zhang, S. Li, X. Wang, J. Luo, High-performance triboelectric nanogenerator based on electrospun PVDF-graphene nanosheet composite nanofibers for energy harvesting, Nano Energy 80 (2021), 105599, https://doi.org/10.1016/j. nanoen.2020.105599.
- [167] G. Min, A. Pullanchiyodan, A.S. Dahiya, E.S. Hosseini, Y. Xu, D.M. Mulvihill, R. Dahiya, Ferroelectric-assisted high-performance triboelectric nanogenerators based on electrospun P(VDF-TrFE) composite nanofibers with barium titanate nanofillers, Nano Energy 90 (2021), 106600, https://doi.org/10.1016/j. nanoen.2021.106600.
- [168] C. Sun, G. Zu, Y. Wei, X. Song, X. Yang, Flexible triboelectric nanogenerators based on electrospun poly(vinylidene fluoride) with MoS2/carbon nanotube composite nanofibers, Langmuir 38 (2022) 1479–1487, https://doi.org/10.1021/ acs.langmuir.1c02785.
- [169] B. Fatma, R. Bhunia, S. Gupta, A. Verma, V. Verma, A. Garg, Maghemite/ polyvinylidene fluoride nanocomposite for transparent, flexible triboelectric nanogenerator and noncontact magneto-triboelectric nanogenerator, ACS Sustain. Chem. Eng. 7 (2019) 14856–14866, https://doi.org/10.1021/ acssuschemeng.9b02953.
- [170] M. Tayyab, J. Wang, J. Wang, M. Maksutoglu, H. Yu, G. Sun, F. Yildiz, M. Eginligil, W. Huang, Enhanced output in polyvinylidene fluoride nanofibers based triboelectric nanogenerator by using printer ink as nano-fillers, Nano Energy 77 (2020), 105178, https://doi.org/10.1016/j.nanoen.2020.105178.
- [171] Q. Ye, Y. Wu, Y. Qi, L. Shi, S. Huang, L. Zhang, M. Li, W. Li, X. Zeng, H. Wo, X. Wang, S. Dong, S. Ramakrishna, J. Luo, Effects of liquid metal particles on performance of triboelectric nanogenerator with electrospun polyacrylonitrile fiber films, Nano Energy 61 (2019) 381–388, https://doi.org/10.1016/j. nanoen.2019.04.075.
- [172] A. Yar, Enhanced output performance of tetraethyl orthosilicate and graphene nanoplates-decorated nanofiber-based triboelectric nanogenerators, Colloids Surf. A Physicochem. Eng. Asp. 631 (2021), 127670.
- [173] S. Parandeh, M. Kharaziha, F. Karimzadeh, An eco-friendly triboelectric hybrid nanogenerators based on graphene oxide incorporated polycaprolactone fibers and cellulose paper, Nano Energy 59 (2019) 412–421, https://doi.org/10.1016/j. nanoen.2019.02.058.
- [174] S. Parandeh, M. Kharaziha, F. Karimzadeh, F. Hosseinabadi, Triboelectric nanogenerators based on graphene oxide coated nanocomposite fibers for biomedical applications, Nanotechnology 31 (2020), https://doi.org/10.1088/ 1361-6528/ab9972.
- [175] N. Sun, G.G. Wang, H.X. Zhao, Y.W. Cai, J.Z. Li, G.Z. Li, X.N. Zhang, B.L. Wang, J. C. Han, Y. Wang, Y. Yang, Waterproof, breathable and washable triboelectric nanogenerator based on electrospun nanofiber films for wearable electronics, Nano Energy 90 (2021), 106639, https://doi.org/10.1016/j. nanoen.2021.106639.
- [176] Z. Sha, C. Boyer, G. Li, Y. Yu, F.M. Allioux, K. Kalantar-Zadeh, C.H. Wang, J. Zhang, Electrospun liquid metal/PVDF-HFP nanofiber membranes with exceptional triboelectric performance, Nano Energy 92 (2022), 106713, https:// doi.org/10.1016/j.nanoen.2021.106713.
- [177] G. Min, A. Pullanchiyodan, A.S. Dahiya, E.S. Hosseini, Y. Xu, D.M. Mulvihill, R. Dahiya, Ferroelectric-assisted high-performance triboelectric nanogenerators based on electrospun P(VDF-TrFE) composite nanofibers with barium titanate nanofillers, Nano Energy 90 (2021), 106600, https://doi.org/10.1016/j. nanoen.2021.106600.
- [178] J. Muller, C. Kumar, A.K. Ghosh, V. Gupta, M. Tschopp, V. Le Houérou, A. Fery, G. Decher, M. Pauly, O. Felix, Spray-deposited anisotropic assemblies of plasmonic nanowires for direction-sensitive strain measurement, ACS Appl. Mater. Interfaces 14 (2022) 54073–54080, https://doi.org/10.1021/ acsami.2c14526.
- [179] W. Liu, Z. Wang, G. Wang, G. Liu, J. Chen, X. Pu, Y. Xi, X. Wang, H. Guo, C. Hu, Z. L. Wang, Integrated charge excitation triboelectric nanogenerator, Nat. Commun. 10 (2019) 1–9, https://doi.org/10.1038/s41467-019-09464-8.
- [180] J. Zhong, Y. Zhang, Q. Zhong, Q. Hu, B. Hu, Z.L. Wang, J. Zhou, Fiber-based generator for wearable electronics and mobile medication, ACS Nano 8 (2014) 6273–6280, https://doi.org/10.1021/nn501732z.



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