




Review: materials for biocompatible tribo-piezo nanogenerators

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Received: 11 November 2022

Accepted: 20 February 2023

Published online:
16 March 2023

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ABSTRACT

The commendable growth of portable and wearable electronics has taken the energy harvesting sector to new heights. Using the idea of nanogenerators for ambient nano-energy harvesting started with the emergence of the piezoelectric energy harvester reported in 2006. Three diverse types of nano-energy harvesters have developed: piezoelectric nanogenerators, triboelectric nanogenerators for harvesting mechanical energy, and pyroelectric nanogenerators for ambient heat energy harvesting. However, the efficiency of these nano-energy harvesters is limited to converting individual energy sources into electrical energy. Since technology is improving day by day, the requirement for efficient, sustainable hybrid energy harvesters is of great interest. Hybrid nanogenerators can be developed by combining two or more nanogenerators mentioned above. In this regard, hybrid tribo-piezoelectric nanogenerators, hybrid tribo-pyroelectric nanogenerators, and hybrid tribo-piezo-pyroelectric nanogenerators exist. The hybrid nanogenerators will have the essential characteristics of the individual nanogenerator with which it is developed, and a positive synergism can enhance the efficacy. This paper reviews the recent advancements in biocompatible energy harvesting materials that are efficient enough to be used as active layers in the tribo-, piezo-, and hybrid tribo-piezoelectric energy harvesters for wearable and biomedical applications. The article focuses on the properties, synthesis, development of materials, usage, hybrid tribo-piezoelectric energy harvesters, and their efficiency, which are made up of these materials, their applications, and finally, the conclusions and future perspectives of these materials.

Handling Editor: M. Grant Norton.

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<https://doi.org/10.1007/s10853-023-08321-w>

Introduction

The concerns over the imminent energy crisis and the impact of global warming aroused interest in sustainable energy harvesting. Energy exists in nature in various ways, including light, heat, vibrations, gravitation, etc., to generate usable electrical energy. Conventional energy sources such as solar, tidal, wind, coal, hydrothermal, geothermal, and nuclear can be used to produce electricity on a large scale for industrial applications. Of course, some of these energy sources ensure sustainable energy harvesting, but their availability depends on climatic conditions. With the development of modern portable [1], flexible [2, 3], and wearable electronics [4–6], the requirement for efficient and eco-friendly means of electrical energy harvesting is seeking attention. As a result, various energy harvesters, such as electrostatic [7–9], electromagnetic [10–12], thermoelectric [13–15], piezoelectric [16–18], pyroelectric [19–21], and triboelectric [22–24], have been explored (basic schematic as in Fig. 2). Their performance is briefed in Table 1.

Materials undergo deformation on the application of mechanical stress. Certain materials can generate electricity from this deformation due to the linear electromechanical interaction of their crystalline characteristics. These materials are called piezoelectric materials [25, 26], and the effect is piezoelectricity. The piezoelectric effect can be correlated with dipole moments in the solid. This effect is a change in polarization (dipole density) countering the applied mechanical force (stress). The external stress will contribute to the bulk's reconfiguration/reorientation of the current dipoles. This unbalancing in the positive and negative dipoles of material will result in a net generation of electric charges. However, this process is reversible, and the latter is called the converse (inverse) piezoelectric effect (deformation of crystals on the application of an electrical field). The effect of piezoelectricity has been explored more in

this era as an efficient ambient energy harvesting technology to power up small-scale electronics [27, 28].

The journey of piezoelectric nanogenerators (PENG) started when Prof. Zhong Lin Wang and his team at Georgia Institute of Technology exploited the effect in 2006 for the first time in developing nanogenerators. These vertical nanowire integrated nanogenerators (VING) were made using ZnO nanowires. Later in 2010, he came up with a Lateral nanowire integrated nanogenerator (LING) having a 2D structure rather than a 3D structure of VING [29, 30]. This LING, when made using semiconductor material, can be used to tune the gate voltage in field effect transistors (FETs).

The piezoelectric energy harvesters (PEH) are diverse in their material selection, device design, circuit, energy resource utilization, area of application, optimization of design and operation, and so on [31–40]. The advanced manufacturing processes and the micro–nanoscale materials with enriched piezoelectric properties enabled the advancement of PENGs with excellent features (high electromechanical coupling factor and piezoelectric coefficient). These processes made the piezomaterial flexible enough to be integrated for specific applications [41]. PENG is a potential candidate with a long lifespan and self-powering capability and hence finds many applications in wireless sensor networks in addition to wearable and implantable devices. The speciality of PENG to capture vibrations from the environment and convert the same to valuable electrical output attracts the harvester to be an emergency vibration sensor in bridges and tall buildings. Due to the rapidly rising digital demand in human life, PENG is potent enough to replace batteries better. The current scenario is competing to explore more from PENG to develop the future portable, wearable, flexible, and implantable devices owing for a large-scale biocompatible energy production [42–49].

Table 1 Performance of energy generators

Energy harvester	Type of energy utilized	Maximum power output (mW/cm ²)
TENG, PENG, EMG	Vibrational mechanical energy	10–100
Biochemical	Biochemical energy	0.1–1
Pyroelectric nanogenerator	Thermal/heat energy	0.01–0.1

TENG Triboelectric Nanogenerator, *PENG* Piezoelectric Nanogenerator, *EMG* Electro-Magnetic generator

The technique of energy harvesting by exploiting the contact electrification phenomenon to develop a triboelectric nanogenerator (TENG) was first disclosed in 2012 by Prof. Zhong Lin Wang and his colleagues as a power backup to Internet of Things (IoT). The proposed TENG, developed using two distinct polymer layers (Kapton and Polyester (PET)), provided 3.3 V output, and a power density (PD) of $\sim 10.4 \text{ mW/cm}^3$ [50]. Contact electrification (tribocharging) coupled with electrostatic induction is TENG's basic working principle. The frequent contact of materials results in the attraction of different molecules in them, which cannot be mapped to a chemical bond among the atoms. However, there will be an exchange of electrons, leading to an electrostatic attraction. The corresponding periodic separation will disturb the attractive force and create friction among the materials. The electronic charge transfer is not reversible immediately either; as a result, a free electron will be available in one of the contacting materials, and an equivalent hole will be created on the other. This deficiency and abundance of electrons will result in a net positive and negative potential on their surfaces, which gets dissipated on separation [51, 52]. Material selection is more accessible with existing triboelectric series [53] to design TENG. The materials in the series are arranged so that the one with the minimum electron affinity (a propensity towards electron loss and thereby acquire a net positive (+ve) charge) will appear first in the list, followed by the materials with higher electron affinity (a propensity towards electron gain and thereby acquire a net negative (-ve) charge) in increasing order. One of the best examples of an energy generator that makes use of the triboelectric effect is the Van De Graaff generator which generates electricity by rubbing a rubber belt against a metallic comb. (The belt remains with a net +ve charge by losing electrons, and the comb will acquire a net -ve charge by borrowing electrons from the belt.) [54].

TENG can be accounted for future large-scale energy production as the typical large-scale energy harvesting resources are getting depleted day by day. The operating modes of TENG can be categorized into sliding and vertical contact separation (CS) modes, where in free-standing and rotating designs fall under the sliding mode. The CS model will generate electricity from ambient pressure and vibrations. This model is a prominent candidate among other designs due to its higher lifespan and negligible

friction damage compared with the other designs [55, 56]. There are various sources of energy harvesting for a TENG. These include wastewater flow energy harvesting method [57, 58], textile energy harvesting [59–61], energy harvesting from human motions (walking, running) [62–64], magnetic force, finger tapping, pressure [65–67] and so on. The efficacy of a TENG is strongly reliant upon the impact of contact forces [68, 69], temperature, pressure, humidity [70–72], surface structures and its design [73–75] as well as the arrangement of active layers within the design. The TENGs' efficacy can be further improved by advancing the surface charge density for which various processing can be employed, such as heating [76], plasma treatment [77], and laser treatment [78, 79]. There are enormous applications for TENGs due to their miniaturized structure, low cost, and ability to self-power. Applications in automobiles (braking, tires rolling), biomedical applications (human body monitoring, pulse monitoring, self-powering implantable drug delivery system, pacemaker, hearing aids, disinfection, sterilization), flexible microphone and speaker systems, and air purifying systems are a few among them [80–86] to be worth mentioning.

The operation and efficiency of various energy harvester device configurations and materials used for them can be effectively studied using finite element analysis/finite element method (FEA/FEM). COMSOL Multiphysics is a tool that can be utilized to make a comprehensive analysis of various materials and its properties, leading to material optimization, finally optimization of device features such as, geometry, dimensions, arrangement, and layers. The program supports coupled partial differential equation systems (PDEs) and standard physics-based user interfaces. The integrated development environment of COMSOL Multiphysics provides a unified workflow for electrical, mechanical, acoustic, chemical, fluids, hydrodynamics, and so on. Atomistix Tool Kit (ATK) and Technology Computer-Aided Design Software (TCAD) are promising simulation software that can be exploited enough while designing the NEG geometries and material properties. Software simulations also effectively reduce material consumption, time, and human effort and overcome limitations to have an advanced, optimal working environment to conduct real-time experiments [87–92]. The electronic composition of multibody systems, including atoms/molecules/condensed

phases, is studied using a computational quantum mechanical modelling technique called density-functional theory (DFT). DFT is one of the techniques in condensed-matter physics, computational physics/chemistry, and materials science. This technique has been recently explored to introduce novel ideas and material efficiency in gas sensing, another promising field of research. The utilization of palladium phosphide tellurium (PdPTe) monolayer for detecting SF₆ decompositions [93], dedicated gas sensors for oil transformers [94], and zealous sensors for detecting volatile organic compounds [95] are a few recent advancements in this regard.

The current research trend focuses on harvesting energy from renewable and abundant energy sources, including thermal fluctuations and mechanical vibrations. Ambient mechanical vibrations are available anywhere and everywhere; this energy source is simply a gift of nature that should be exploited enough to develop modern self-powering systems to sustainably meet the global energy crisis. Triboelectric and piezoelectric energy harvesters rely solely on these mechanical vibrations to generate electrical energy.

The performance and efficiency of an energy harvester are greatly influenced by the materials selected as the active layers for developing them and the fabrication technologies employed. The active layers' characteristics can be modified by introducing apt and adequate dopants and employing suitable fabrication technologies. This paper reviews the basis of the material selection strategy for tribo-piezo-active layers in nanogenerators, selection indicators for biocompatible materials, recent materials available, fabrication techniques employed, and applications, and emerging novel materials applied energy harvesting.

Strategies for material selection

The performance of every device depends on the material it is made of and how. Several factors should be considered while selecting the appropriate materials for designing the energy harvester, which includes various physical, chemical, and structural properties, availability, and cost, response to environmental factors (like electromagnetic interferences,

vibrations, shakes, shocks, corrosion), impact on environmental and human lives, robustness, and lifetime under given operating conditions. Also, while designing an energy harvester, it is crucial to focus on parameters such as power density, flexibility, stability, sustainability, repeatability, and efficiency. This material selection also involves different classifications of materials such as metals, ceramics, polymers, and green and inorganic materials. The essential strategies to improve the energy harvester's performance can be summarized as follows: (1) By using different types of piezoelectric materials [27], (2) Chemical doping/improvisation of piezoelectric materials [96], (3) Extemporization of microscopic morphological characteristics [97], (4) Composite thin film advancement [98], (5) External charge excitation [99], (6) Boosting stability by penta-stable configuration [100], (7) Surface engineering [101], (8) Material state interactions and interfacing [102], (9) Smart materials [103], (10) Active catalytic coatings [104], and (11) Structural modification [105, 106].

Biocompatibility indicators for material selection

Implantable energy harvesters are of prime interest to replace conventional battery-powered cardiac pacemakers and brain implants. Polymeric materials are a good choice for developing implants as they have minimal elastic modulus similar to cortical bone, excellent chemical stability, elasticity, and non-toxicity. The use of polymeric materials as high-quality biomaterials is supported by their simplicity of manufacture, which facilitates the creation of various forms (films, fibres, sheets, unique implant shapes). Polymeric biomaterials must possess good mechanical, physical, and surface qualities such as roughness, hydrophilicity/hydrophobicity, and adhesive capabilities in addition to being biocompatible and sterilizable. Biofunctionality, bioactivity, bioinertia, and biostability are a few of the numerous processes determining biocompatibility. Biocompatibility enables the human body to absorb synthetic implants without triggering an adverse immunological response, an allergic reaction, an inflammatory reaction, or a persistent illness. The biocompatible material selected should not be carcinogenic. The type of application has a significant impact on

biocompatibility. The primary elements that determine biocompatibility are as follows: (1) Environmental interactions—influence of cytotoxins, carcinogenic/mutagenic reactions, toxicological/allergic reactions, degree and quality of biodegradation, inflammatory processes, and contact with human blood, (2) Applied implant time frame, (3) Surface biocompatibility—biological, chemical, and morphological, (4) Structural and functional biocompatibility, and (5) Biodegradability [107–110].

Towards nonlinearity of TENGs

TENG is characterized by unique nonlinear electrical and capacitive properties. Energy extraction from TENG relies on the mode of interaction with external circuitry. The maximum power that can be extracted from TENG remains a mystery. However, the nonlinear behaviour of TENG can be mathematically modelled as follows. For a CS mode TENG, with an area of the electrodes A and distance between triboelectric layers x , the short circuit charge (Q_{sc}) influenced by the contact electrification can be obtained as

$$Q_{sc} = \frac{A\sigma x(t)}{d_0 + x(t)} \tag{1}$$

In the absence of a triboelectric charge, the TENG will remain as a capacitor. According to the superposition theorem, the voltage induced between the electrodes (V) will be the sum of triboelectric charges ($V_{oc}(x)$) and electrostatic induction.

$$V = \frac{-Q}{C_{TENG}(x)} + V_{oc}(x) \tag{2}$$

$$C_{TENG}(x) = \frac{A\epsilon_0}{d_0 + x} \tag{3}$$

During short circuit condition $V=0$. Then,

$$V_{oc}(x) = \frac{Q_{sc}}{C_{TENG}(x)} = \frac{\sigma x}{\epsilon_0} \tag{4}$$

During the press release operation, the distance of separation x becomes a function of time. This property makes the $C_{TENG}(x)$ and $V_{oc}(x)$ time-dependent. These nonlinear functions lead to difficulty in deriving an analytical solution for the TENG (except for resistive and capacitive loads). This unique property of TENG makes it a suitable candidate for designing switching strategies in power management systems [111].

Materials for mechanical energy harvesters

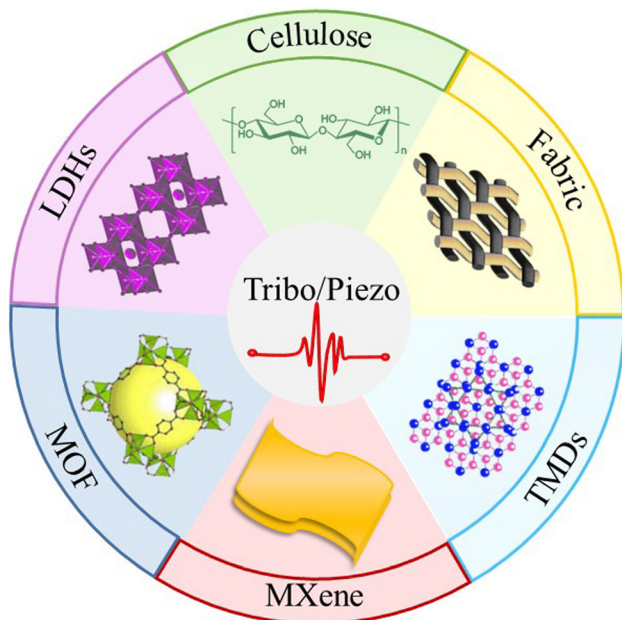
Recent research shows that triboelectric and piezoelectric energy harvesters are the frontiers among mechanical nano-energy harvesters to generate electrical energy out of mechanical vibrations. Since the initial development of nanogenerators in 2006 by Zhong Lin Wang, the field of energy harvesting has gained a great deal of focus and is used for various applications. The first nanogenerator was made up of zinc oxide (ZnO) nanowires which work according to the piezoelectric effect with 17–30% efficient power conversion; these devices are termed piezoelectric nanogenerators (PENG). The designed device generated a voltage output (V_{out}) of 8 mV corresponding to a resonant frequency of 10 MHz and an output power of 0.5 pW [112]. Piezoelectricity is the fundamental principle behind the working of PENG. The voltage generation in response to mechanical stress contributes to the piezoelectric effect, and the reverse phenomenon is termed the inverse piezoelectric effect. The materials that offer this effect are non-conductive and can be broadly classified as natural and artificial. The countless applications of portable, wearable, and flexible electronics paved the way for nano-energy harvesters based on biopolymers [113–115].

The TENG works based on induced contact electrification when two or more dissimilar materials repeatedly contact and separate. The best-known example of this effect is rubbing a glass rod with rubber, which creates an excessive -ve charge on the rubber, and a + ve charge on the glass rod, which, when separated, induces a very high voltage. Electron stealing due to quantum tunnelling is the underlying phenomenon responsible for this voltage generation. The developments in TENG started in 2012, with the first version providing an output PD of around 10.4 mW/cm³ [50]. TENGs are good candidates for the Internet of Things (IoT) because they are lightweight, have a minimum cost, and have high efficiency at low frequencies. However, conventional TENGs are limited by the high crest factor (a measure of peak to practical value in a measurement graph). This high crest factor makes it challenging to operate electronics directly, and the output is usually degraded because of air decomposition and material wear [116–118].

The best materials for piezoelectric and triboelectric energy harvesters from 2005 to 2022 (18 years) are

Table 2 Energy harvesters and performance

Material	Type of ENG	Source	Dimension	Voltage current power		
				(V)	(I)	Density
PZT-5H [226]	PENG	Wind	114 mm x 60 m, 60 × 20 × 0.6 mm ³	12.04	–	7.5 mW
PVDF [227]	PENG	Wind	41.5 × 16.3 × 0.22 mm ³	160.2	–	2.57 nW
BaTiO ₃ nanowires and PVC polymer [228]	PENG	Elbow motion	–	1.29	24 nA	10.02 nW
PMN-PT [229]	PENG	Vibration	1.7 cm × 1.7 cm	8.2	145 μA	–
Bismuth Titanate [230]	PENG	Periodic mechanical load	–	60	400 nA	18.5 mW
PMMA/PDMS, thin Au film [231]	TENG	Mechanical force 10 N	3 × 3 in × 3/32 in	1200	2 mA	313 W/m ²
PTFE/Acrylic [232]	TENG	Rotation	12 cm ²	410	90 μA	36.9 W/m ²
PDMS/Al film [233]	TENG	Human motion	2 × 2 cm	465	13.4 μA/cm ²	53.4 mW/cm ³
PTFE/Kapton/Al [234]	TENG	Human motion	3.8 × 3.8 cm × 0.95 cm	215	0.66 mA	10.24 mW/cm ³
PDMS/PS [235]	TENG	Compressive force 902 N	1 × 1 cm, 300 μm	130	0.10 mA/cm ²	–

**Figure 1** Graphical abstract.

elaborated in Table 2 concerning the device dimensions and output performance. It is clear from Table 2 that based on a material and source of input, power generated is in the range of nW to W. Some of the most efficient biocompatible materials, used as active

tribo/piezo-layers reported very recently are discussed below (Fig. 1).

Cellulose and its composites

Cellulose and fabric fall under natural materials for TENGs. Cellulose is the most efficient and abundant triboelectric material available. It is renewable, cost-effective, biocompatible, and eco-friendly. This material can be easily extracted from plants and trees and provides excellent mechanical, dielectric, and piezoelectric properties [119]. The method of developing a triboelectric nanogenerator using cellulose as the raw material enables us to replace the use of synthetic polymers (Fig. 2).

Cellulose II aerogel with a 3D open-pore structure can be synthesized from the dissolution regeneration process in lithium bromide trihydrate (an inorganic molten salt hydrate) and can be used to fabricate TENGs. The high degree of flexibility, porosity, and surface area of 221.3 m²/g are the attractive features of this material. TENGs developed from this material exhibit outstanding mechanical response, with enhanced electrical output and sensitivity making it a viable candidate for lighting light emitting diodes (LEDs), charging commercial capacitors, powering calculators and sensors, etc. [120]. The performance

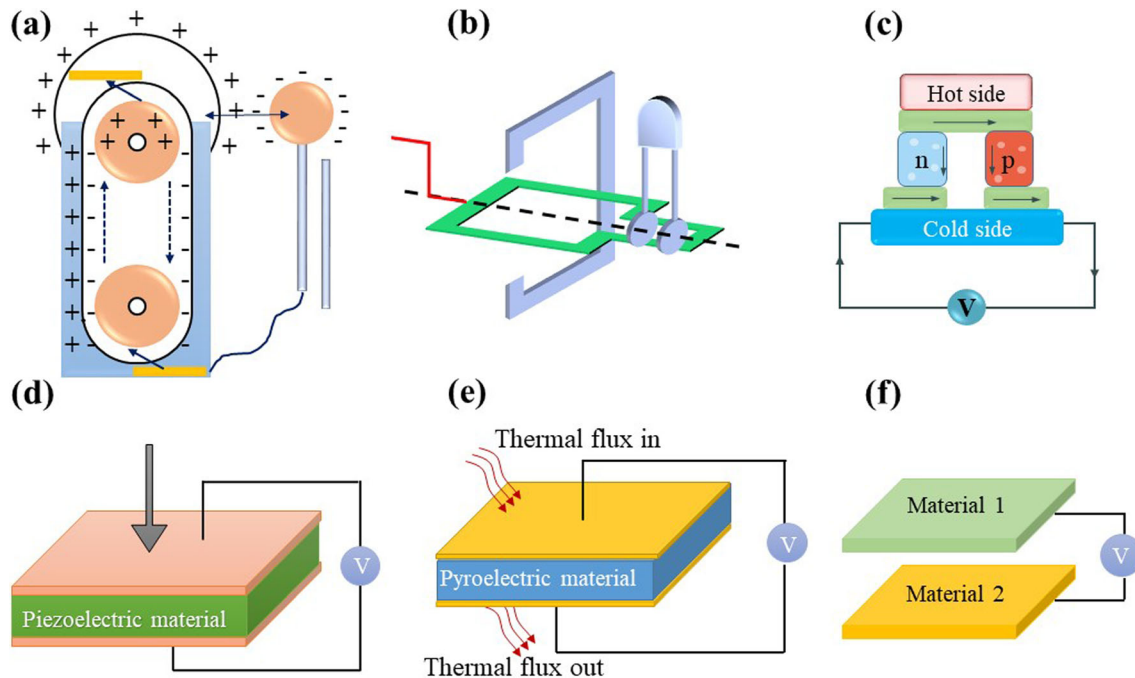


Figure 2 Various types of energy harvesters. **a** Electrostatic, **b** electromagnetic, **c** thermoelectric, **d** piezoelectric, **e** pyroelectric, **f** triboelectric.

of the cellulose-based nanogenerators might be limited because of their poor polarity, hydrophobicity, and insufficient functionalization. However, a chemical functionalization by modifying the functional groups in cellulose nanofibril (CNFs) can overcome this limitation by controlling the surface polarizability and hydrophobicity. The triboelectric charge density of the CNFs can be further improved by grafting surface-bound fluorine-bearing silane chains, which also upgrades its hydrophobicity. The proposed CNF is a suitable candidate for bio-TENGs exhibiting excellent resistance to moisture, enduring cycle stability, retaining 70% of the initial output performance for the same % of ambient humidity, and a short circuit current (I_{sc}) of 9.3 μA (Fig. 3a) [121].

The mechanical energy contained in raindrops may be effectively transformed into electrical energy. A drum-shaped TENG (DTENG) may be created by treating an elastic superhydrophobic cellulose paper with triethoxy-1H,1H,2H,2H-tridecafluoro-n-octylsilane after spray spraying it with nano-fumed silica dispersed in a solution of thermoplastic elastomer. When raindrops connect with the D-TENG in the proposed energy harvester, they separate and interact with polytetrafluoroethylene (PTFE), which in turn routinely creates electrical energy. The device

can provide a peak output voltage of 21.6 V, 16 μW per droplet for a 6 mm (about 0.24 in) water drop from a height of 2.5 m (Fig. 3b) [122]. Due to their exceptional performance, TENGs based on bacterial cellulose (BC) are widely used in self-powered wearable and implantable devices. The limitation of cellulose is that it should undergo several regenerations and solubilization techniques to form a film. These techniques seriously degrade the nanostructure and crystallinity, which may limit the efficient utilization of the TENG output. BC can be used to counter this limitation. BC nanocomposites were adhesively bonded to the conductive substrate (ITO) by drying BC hydrogel. The polarizability and surface roughness of the film was upgraded by incorporating ZnO nanoparticles at its surface. The proposed TENG exhibited an excellent open circuit voltage (V_{oc}) of 57.6 V, I_{sc} of 5.78 μA , and PD maximum of 42 mW/m^2 when connected to an external load [123]. The highest friction between layers and electrodes of a BC-TENG with a sandwich structure, created from conductive and pure BC precipitated with conducting and reinforced nanoparticles, offered the best performance with a V_{oc} of 29 V, I_{sc} of 0.6 μA , and output power at 3 μW (Fig. 3c) [124].

A piezoelectric film bionanocomposite with 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO) oxidized

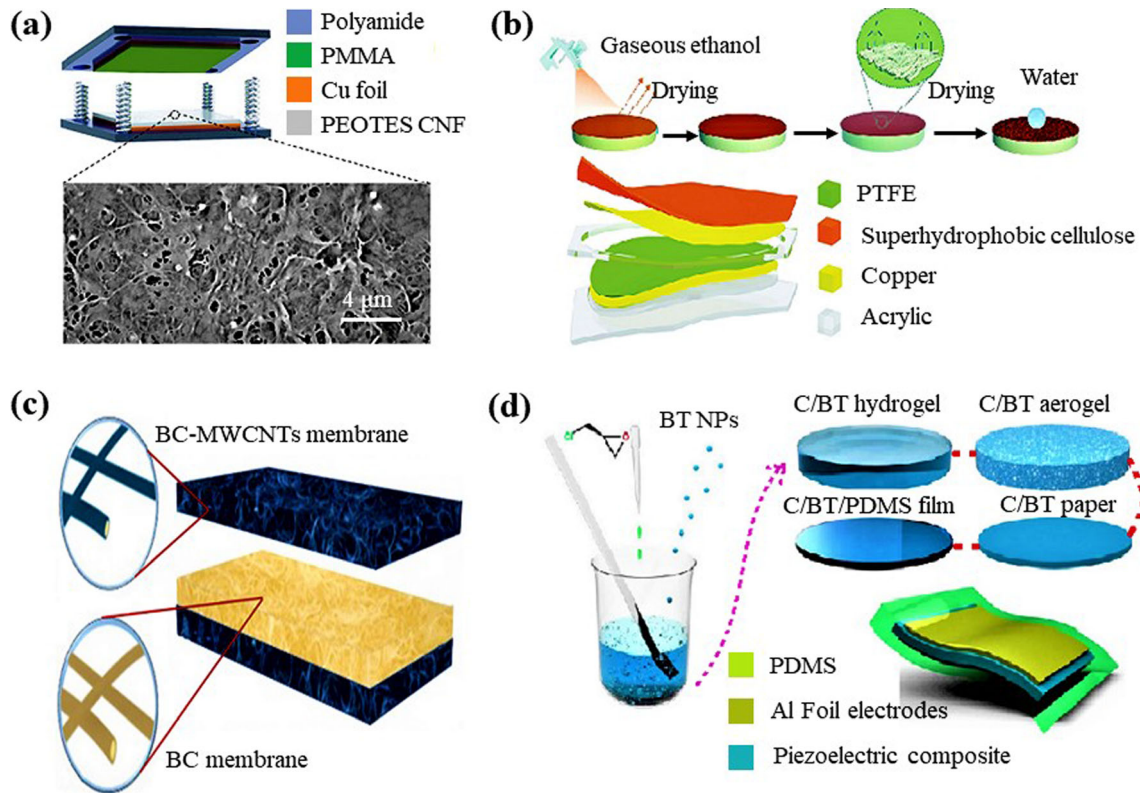


Figure 3 Cellulose-based energy harvesting an overview. **a** SEM image of the PFOTESCNF film. Reproduced with permission from [121]. **b** Procedure to prepare PFOTES silica/paper composite, Surface engineered TENG for drug loading, SEM images of Filter paper, Silica/paper composite, and PFOTES silica/paper

cellulose nanofibril (TOCN)/molybdenum disulphide (MoS_2) nanosheet was prepared by aqueous dispersion method. The proposed TOCN/ MoS_2 nanocomposite film shows more attractive features than conventional piezoelectric film based on cellulose. It showed excellent mechanical properties (Young's modulus = 8.2 and tensile strength = 307 MPa) and piezoelectric property (longitudinal piezoelectric constant (d_{33}) = 31 pC/N). The piezoelectric nanogenerator developed from this nanocomposite film could provide maximum output voltage (V_{max}) of 4.1 V and I_{sc} 0.21 μA , respectively [125]. An efficient piezoelectric nanocomposite based on cellulose was developed by incorporating gold (Au) nanoparticles (NPs) in a matrix of cellulose and PDMS. The presence of cellulose enhanced the dielectric constant of PDMS, and Au NPs limited its dielectric losses. The piezoelectric nanogenerator fabricated using the above nanocomposite exhibited V_{oc} of 6 V, I_{sc} of 700 nA, peak PD of 8.34 mW/m^2 , and high efficiency of energy conversion by 1.8%. The

composite. Reproduced with permission from [122]. **c** BC TENG's structure. Reproduced with permission from [124]. **d** Fabrication of PENG based on C/BT aerogel paper, Optical and cross-sectional SEM images of the C/BT paper and C/BT/PDMS film. Reproduced with permission from [129].

nanogenerator was able to light up two commercial blue LEDs and charged a capacitor of 10 μF to 6.3 V in 11.28 min [126]. MoS_2 nanosheets developed by mechanical exfoliation in triethanolamine were combined with regenerated cellulose (RC) to form (RC)/ MoS_2 nanocomposite. A piezoelectric nanogenerator fabricated with this nanocomposite provided a five times higher V_{out} of 2 V and 75 times higher current of 150 nA than neat RC under the same input pressure [127]. A pressure-driven piezoelectric nano-energy harvester was fabricated using electrospun nanofibre membranes of cellulose acetate/cellulose nanocrystal (CA/CNC) composite. The pervasiveness of CNC improved the piezoelectric cellulose I crystallization, and the nanofibre membranes' mechanical deformation was improved, which resulted in an enhancement of the piezoelectric performance of the composite membranes. A mass fraction of 20% CNC in the prepared nanofibre yielded a V_{out} of 1.2 V for a force of 5 N applied for a frequency of 2 Hz. This also demonstrated a linear

relationship between the output voltage and applied force, making it a potential contender for pressure-sensing applications [128]. Flexible high-performance hybrid nanogenerators are greatly interesting when designing energy harvesters for high-power applications. A flexible high-performance hybrid tribo-piezo-energy harvester was recently fabricated, PENG, using regenerated cellulose/BTO aerogel papers based on PDMS nanocomposite in combination with single-electrode mode TENG. The proposed PENG alone was able to provide a V_{\max} of 15.5 V and power of 11.8 μW while the positive coupling effect provided a V_{\max} of 48 V and 85 μW power, respectively, for the hybrid nanogenerator (Fig. 3d) [129].

Fabric and derivatives

Fabric is a natural material that can be used for developing triboelectric nanogenerators. The fabric is a desirable choice for self-powered sensors and wearable energy harvesters. Triboelectric energy harvesters based on fabric are gaining much attention due to their efficiency in utilizing wasted electrostatic energy to generate usable electrical power.

The triboelectric properties of commercial velvet textiles were upgraded by enhancing the fibre texture using amide bonds and hierarchical topologies. Chemical grafting of carbon nanotubes (CNT) and poly (ethylenimine) (PEI) via a poly-amidation process was used to enrich the fibre. The fabricated TENG showed long-term resilience and robustness with a PD of 3.2 W/m^2 on a 5 $\text{M}\Omega$ external resistor. The proposed TENG was able to power myriad electronics such as pedometers, digital watches, calculators, and digital timers (Fig. 4a) [130]. A direct current (DC) fabric (1.5 \times 3.5 cm) triboelectric nanogenerator was developed to convert the “annoying” electrostatic effect caused by the contact of two dissimilar materials into electrical energy. It can light up to almost 416 serially connected LEDs (Fig. 4b,c) [131]. The wide use of TENGs based on fabric for self-powered sensing is due to their breathability and flexibility properties. However, they are prone to fire and related hazardous issues. A textile-based flame-retardant TENG (FT-TENG) with conductive cotton fabric (CF), having flame-retardant properties in combination with CF coated with PTFE, and a spacer was developed to tackle this limitation. The device had excellent fire resistance and energy harvesting properties, with a maximum PD of around 343.19

mW/m^2 at 3 Hz. Also, this device retains its electrical output even when exposed to 220 $^{\circ}\text{C}$ (Fig. 4d) [132]. Recent developments include the introduction of a sandwich-structured F-TENG that can be used for biometric identification and biomechanical energy harvesting. F-TENG could power up a digital watch under low-frequency movement and respond to pressure changes even with leaves falling on it. A Self-Powered Wearable Keyboard (SPWK) was forged by merging large-area sensor arrays of F-TENG. It was used for tracing and recording electro-physiological signals and identifying an individual’s typing characteristics using the Haar wavelet transform [133].

A PENG capable of providing 1.45 mW/cm^2 PD was developed lately by doping silver on ZnO nanowires. This nanogenerator exhibited three times improved output than undoped ZnO-based PENG with reasonable load capacitors charging capability [134]. Even though textile-based pressure sensors are gaining significant attention, they are challenged by retaining the output performance without compromising the wearing convenience. The proposed three-layered textile-based piezoelectric pressure sensor (TEPS) was designed with top, and bottom layers made up of conductive reduced graphene oxide (rGO) polyester (PET) and a middle layer with PVDF membrane/self-oriented ZnO nanorods composite. The lower detection limit of the device was 8.51 Pa and the sensitivity of 0.62 V.kPa^{-1} [135]. The incorporation of silicon-based quantum dots (SiDs) enhanced the piezoelectric response of electrospun polymer fabrics. The proposed hybrid fabric exhibited an output voltage and current 11 and 19 times improved, respectively, than the pristine fabric [136].

Using electrospinning and 2D braiding technology, a PENG was fabricated using PVDF/conductive nylon core-sheath structured piezoelectric yarns. Thus, the fabricated PENG provided an output of 120 mV with a yarn dimension of 10 cm \times 600 μm (length \times fineness) under various human movements (like walking, bending, etc.). This device provided an output stable for a fatigue test conducted at 4 Hz even after 800 s, thus maintaining the cycle stability for more than 3200 cycles [137]. A PENG was developed recently based on non-woven piezoelectric textiles inspired by muscle fibres for physiological monitoring, such as human motion, voice recognition, and pulse wave monitoring. The muscle fibres were mimicked by the dispersion of polydopamine

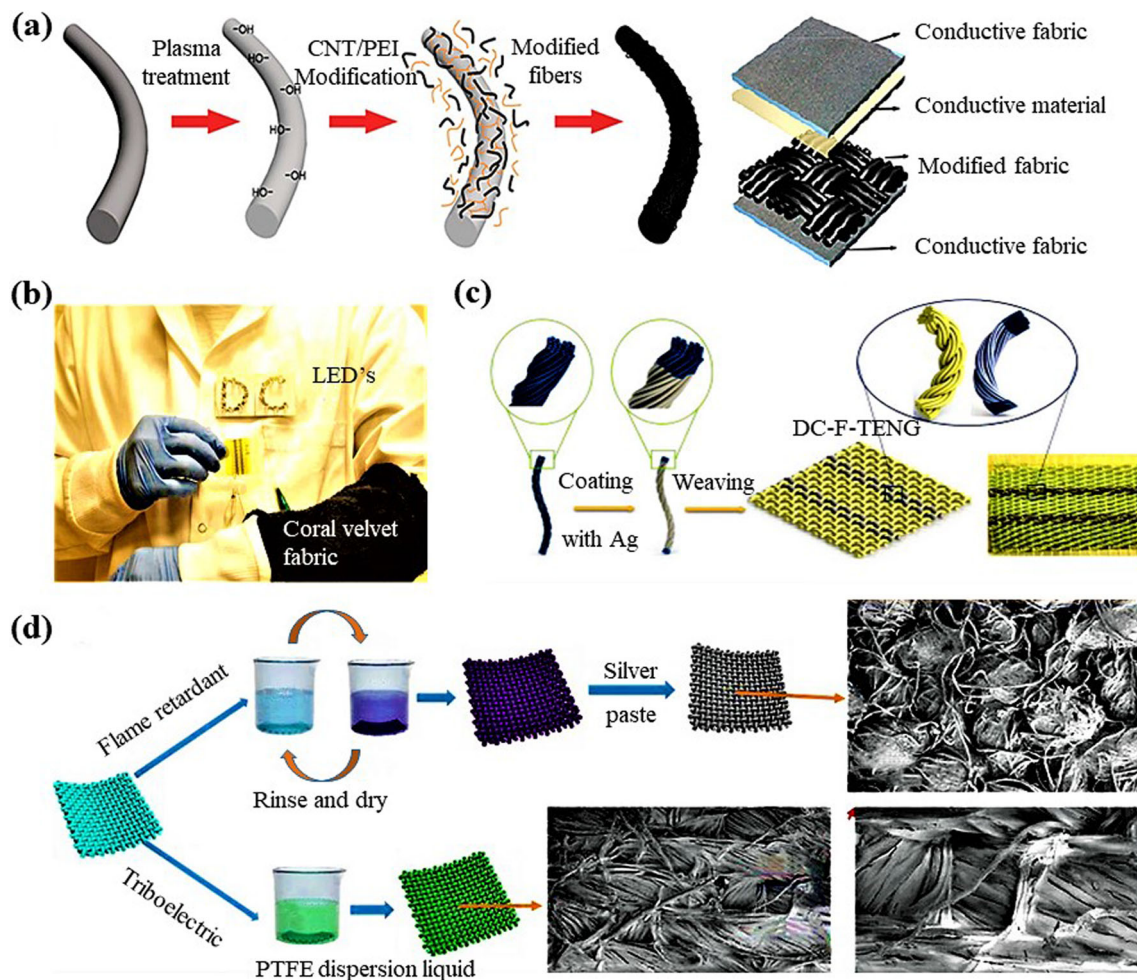


Figure 4 A look back to energy harvesting based on fabrics. (a) Modification of velvet fabric using CNT and PEI. Reproduced with permission from [130]. (b) Demonstration of LED lighting with the sliding DC-TENG, (c) Fabrication, and working

mechanism of DC-TENG. Reproduced with permission from [131]. (d) EM images of flame-retardant conductive fabric (CF), Flame retardant CF. Reproduced with permission from [132].

(PDA) into electrospun BTO/PVDF nanofibres (PDA@BTO/PVDF), which enhanced the interfacial bonding, mechanical force, and piezoelectric properties. The proposed device demonstrated excellent sensitivity and long-term stability of 3.95 VN⁻¹ and < 3% decline after 7,400 cycles, respectively [138]. A simple, cost-effective, flexible BiFeO₃ (BFO) film-based hybrid piezo-triboelectric nanogenerator (HyTPENG) was implemented to obtain a V_{oc} of 110 V and I_{sc} of 3.67 $\mu\text{A}/\text{cm}^2$. The proposed device furnished a maximum output PD of 151.42 $\mu\text{A}/\text{cm}^2$ for a load resistance of 250 M Ω [139]. A HyTPENG is introduced for energy generation and monitoring that employs magnetic force to provide the opposing force between the Cu/Al layers and Kapton in the sliding mode for the triboelectric component and

PVDF strips. The peak power for the triboelectric portion with Cu set up in HyTPENG mode two is around 12 μW , whereas the piezoelectric part delivered a PD of 70 μW . The suggested HyTPENG is used as part of a self-powered walking sensor system to examine how people walk on a treadmill [140].

Transition metal dichalcogenides (TMD or TMDC)

The TMD/TMDC monolayers are thin atomic semiconductors of MX_2 , with M being a transition metal atom (yttrium (Y), zirconium (Zr), niobium (Nb), molybdenum (Mo)), and X is a chalcogen atom (Oxygen (O), sulphur (S), tellurium (Te), selenium (Se), polonium (Po)). The configuration of MX_2

consists of a single layer of M atoms sandwiched between 2 layers of X atoms. TMD developed with WTe_2 exhibits incredible magneto-resistance and superconductivity. TMD monolayers made of MoS_2 , WS_2 , $MoSe_2$, WSe_2 , and $MoTe_2$ have direct bandwidth. This property makes them find applications in transistors in electronics and emitters/detectors in optics [141–148]. TMD monolayers can be fabricated using techniques such as exfoliation [149–152], chemical vapour deposition (CVD) [153–156], and molecular beam epitaxy (MBE) [157–159]. While designing patchable and implantable type nanogenerators, diverse technical aspects such as efficiency, high capacity, compact battery technology, minimal power, component miniaturization, and flexible technology should be addressed. 2D materials are ideal for self-powering implantable and patchable nanodevices due to their distinctive qualities, including flexibility, transparency, mechanical stability, and non-toxicity (Fig. 5a) [160].

An innovative strategy was recently used to tailor the texture of TMDs in conjunction with thiolated ligands of various alkyne chain statures to build TMD-based TENG devices with higher output performance. The TENGs showed their excellence in performance by providing a V_{oc} of 12.2 V and a PD of 138 mW/m^2 under vertical contact-separation mode (Fig. 5b) [161]. A simple technology has been disclosed to exhibit self-polled, flexible, and high-performance PENGs using chemically exfoliated MoS_2 nanosheets entrenched in PVDF polymers. These nanogenerators produced V_{oc} of 22 V and an unprecedented piezoelectric sensitivity of 2.07 V/kPa [162]. The larger piezoelectricity of the $MoSe_2$ monolayer than the other groups of VI B TMDs (including MoS_2) has only been explored theoretically. However, in-plane piezoelectricity has been demonstrated experimentally in $MoSe_2$. The monolayer single-crystalline $MoSe_2$ flakes derived from CVD exhibited superior properties to MoS_2 monolayers (60 mV over a 0.6% strain) (Fig. 5c) [163]. The

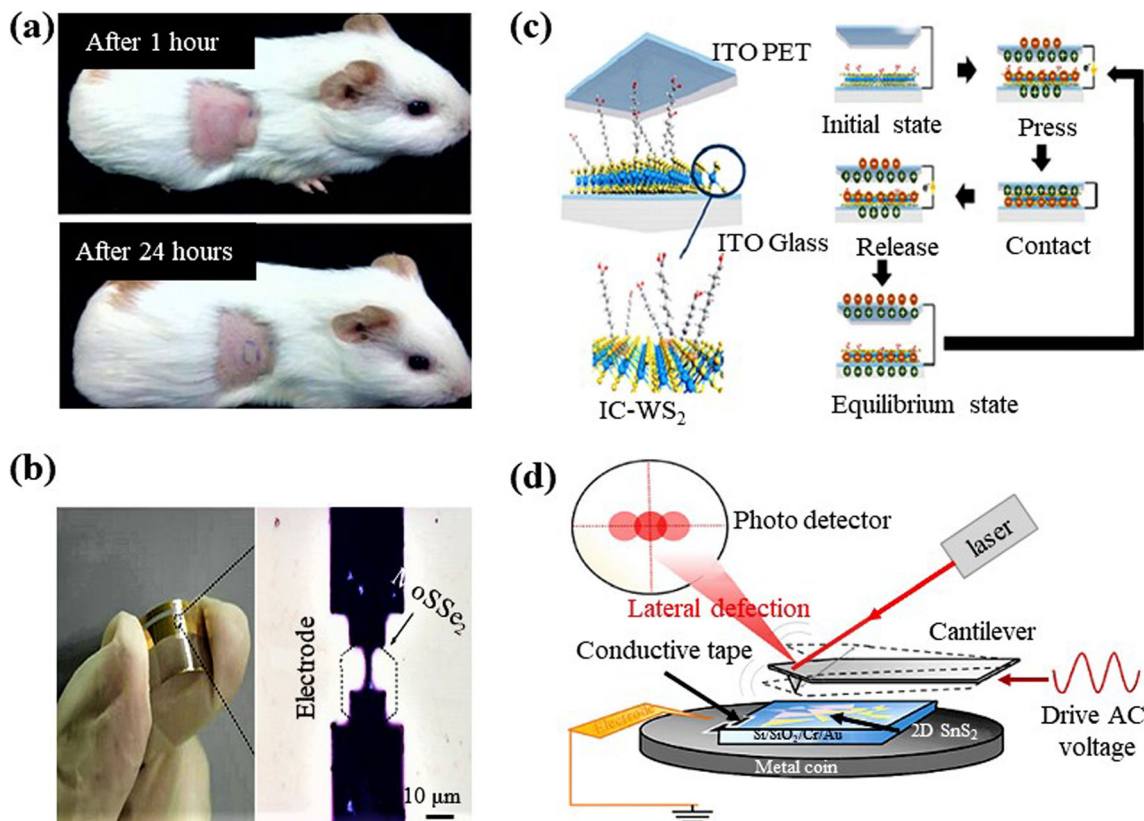


Figure 5 Energy harvesters using Transition metal dichalcogenides. (a) Patchable MoS_2 thin film attached to the skin of guinea pigs after 1 h. and 24 h. Reproduced with permission from [160]. (b) Device structure of TENG based on

ligand conjugated WS_2 . Reproduced with permission from [161]. (c) MoS_2 pH sensor. Reproduced with permission from [163]. (d) Piezoelectric behaviour of 2D SnS_2 nanosheets. Reproduced with permission from [165].

synergistic effect of chalcogen atoms on TMDCs and PVDF-based compounds was investigated in the piezoelectric performance of a fabricated piezoelectric nanogenerator. The PVDF/MoSSe-based nanogenerator showed a maximum V_{oc} of 31.2 V and an I_{sc} of 1.26 μ A. This improvement was achieved with the addition of TMDCs without further treatment [164].

Recent research focuses on another category of 2D TMDs, like post-transition metal dichalcogenides (PTMDs). An example of such a TMD is 2D tin disulphide (SnS_2) nanosheets whose piezo-response concerning change in thickness was studied. The results reveal that 4-nm-thick 2D SnS_2 nanosheets exhibited a 2 ± 0.22 pm/V out-of-plane piezoelectric response. The dependence of piezoelectric property on resonance frequency with thickness suggests that the piezoelectric coefficient falls as the thickness of 2D SnS_2 nanosheets increases. The effectual lateral piezoelectric coefficients evaluated at distinct voltages vary from 0.61 to 1.55 pm/V, with a moderate value of 1 pm/V, using the periodically polished lithium niobate piezoelectric crystal (Fig. 5d) [165].

A few layers of MoS_2 were electrodeposited on a Cu foil, followed by the electrodeposition of ZnO and spin coating of PVDF to add piezoelectricity. During piezo-tribo-impacting, the PVDF/ MoS_2 @ZnO-based HyTPENG exhibits great values of around 140 V and 4.6 μ A with a PD of 256 W/cm². The suggested HyTPENG can draw power from various biomechanical actions, including walking, pressing heels, bending elbows, and machine vibration. It can run electrical gadgets like a wristwatch, calculator, and 33 LEDs connected in series. The PENG proposed is sensitive to physiological signal monitoring and biocompatible, both advantageous for future biomedical applications [166].

MXenes

MXenes are material that comes under the classification of 2D inorganic compounds. The material comprises a few atoms' thick layers of transition metals such as carbon nitrides or carbonitrides. This material was first discovered in 2011 by Yury Gogotsi and Michel Barsoum [167, 168]. MXenes are synthesized via the hydrofluoric acid (HF) etching technique [169]. MXenes are excellent antibacterial agents when compared with graphene oxide. However, the high electron density at the Fermi level makes MXene monolayers to be metallic [170, 171]. This conductive

layered material with tuneable surface termination makes it a promising candidate for energy storage, photocatalysis, purification and sensing applications [172, 173]. MXenes can also behave as meta-material (composites engineered to exhibit unique electromagnetic properties) and are used for applications such as photonic diodes, electrochromic devices, and nanogenerators [174, 175].

Electrospun nanofibre TENG (EN-TENG) using (PVDF-TrFE)/MXene nanocomposite shows excellent surface charge density and dielectric constant. The device demonstrated a maximum PD of 4.02 W/m² for a 4 M Ω external load resistance. It was used to power a stopwatch and hygrometer by gathering energy from finger tapping (Fig. 6a) [176]. A self-powered multifunctional system for the detection of gases was devised by using TENG based on PVA/Ag (poly (vinyl alcohol) /silver) nanofibre and NO₂ gas sensor developed from Titanium carbide MXene/Tungsten oxide nanofibres ($TiC_3C_2T_x$ -MXene/ WO_3). The proffered TENG yielded a peak-to-peak V_{oc} and PD of 530 V and 359 mW/m², respectively. This sensor exhibited an excellent response of around 15 times the resistive MXene/ WO_3 sensor at room temperature (Fig. 6b) [177]. With optimal triboelectric capability, a TENG was designed and fabricated using alternative-layered MXene Niobium Carbide (Nb_2CT_x)/ $TiC_3C_2T_x$ composite nanosheet films with multiple-F (fluorine) groups, stacked layer by layer. When the volume of Nb_2CT_x nanosheets was increased by 15 wt%, this TENG yielded a voltage of 34.63 V and I_{sc} of 8.06 A/cm², which was 3.5 times greater than pure $TiC_3C_2T_x$ films [178]. A liquid electrode MXene-based flexible and shape adaptive TENG can provide a V_{oc} of 300 V. By extracting mechanical energy from hand tapping, the capacitors can be charged to drive the wearable electronics in the self-charging system by using this TENG (Fig. 6c) [179]. A PTFE electret film enhanced by MXene, having excellent mechanical properties and surface charge density, was recently used as the triboelectric active layer. During the friction test, the composite film was made by spraying and annealing treatments. The composite has tunable crystallinity, 450% enhanced tensile property, and 80% reduced wear volume. The film proved its excellence by providing a V_{oc} of 397 V, I_{sc} of 21 μ A, and a transfer charge of 232 nC that was roughly four, six, and six times higher than the TENG based on pristine PTFE film [180].

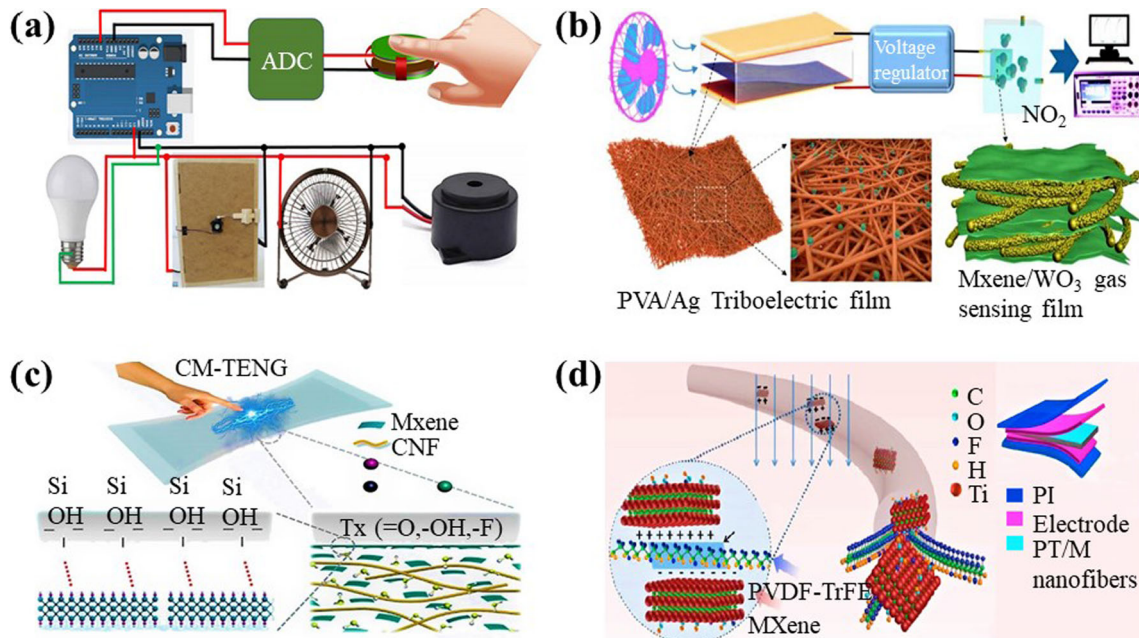


Figure 6 MXenes to power up portable electronics. (a) EN-TENG as a self-powered switch to operate an electric fan system diagram. Reproduced with permission from [176]. (b) TENG for self-powered NO₂ sensor. Reproduced with permission from [177].

(c) CNFs/MXene liquid electrode-based shape adaptive TENG. Reproduced with permission from [179]. (d) Piezoelectric sensor based on PVDF-TrFE/MXene. Reproduced with permission from [181].

In piezoelectric polymer films, obtaining a large output voltage in response to pressure is difficult. Electrospun PVDF-TrFE/MXene nanofibre mats were used to create a piezoelectric pressure sensor to overcome this issue. Under 20 N pressure and a frequency of 1 Hz, the hybrid film with 2.0 wt% MXene produced an immediate output PD of 3.64 mW/m², increasing its potential uses in multifunctional electronic skins (Fig. 6d) [181]. Titanium carbide (Ti₃C₂) seems to be an efficient candidate among the 2D low-dimensional materials in the MXene series. The first experimental study on the piezoelectric response of Ti₃C₂T_x MXene monolayer has been reported by Song et al. In this paper, the inversion symmetry of lattice configurations was disturbed by the functional groups on the MXene's surface to exhibit enriched piezoelectric properties. The armchair direction of the Ti₃C₂T_x MXene sheet exhibited an inherent output current of 0.3 nA under 1.08% tensile stress, correspondingly 6.5 mW/m² output PD, and 11.15% of conversion efficiency [182]. 0.03 g/L Ti₃C₂ into the piezoelectric polymer PVDF induced a V_{out} of 6 V and a PD of 14 μW/cm² across a load resistor of 10.8 MΩ which is high in comparison with other 2D MXene-based piezoelectric nanogenerators [183]. A PVDF/MXene-based piezoelectric microdevice

was forged through microinjection moulding. The stacking in MXene was a propelling force for the regular arrangement of dipoles to provide a self-polarization effect. Without more outlying polarization, the microdevices exhibited a V_{oc} of 15.2 V and I_{sc} of 497.3 nA for one wt% MXene in the polymer [184].

A triboelectric and piezoelectric hybrid generator was created by incorporating MXene and BaTiO₃ ceramic filler into the PDMS matrix (HG-MBP). The introduction of fluorine (F) atoms as concluded groups into MXene substantially influenced the electrical output of the HG-MBP by providing maximum V_{out} of 80 V, current of 14 μA, and PD of 13.5 W/m². The hybrid device successfully operated a 3D-printed robot hand model by scavenging electricity out of finger joint motions of a natural hand. A high-accuracy (93.33%) object detection system is developed by the HG-MBP utilizing the K-mean clustering technique. The proposed hybrid energy harvester is suitable for material sensors and human gesture manipulators, which can be used enough to develop a future e-skin in the human-machine interface [185]. An HyTPENG was reported recently using PVDF and poly(l-lactic acid) (PLLA) as triboelectric layers. 2D-MXene and 1D multiwalled carbon nanotubes (MWCNTs-COOH) were employed as conductive

nanofillers in the electrospun nanofibre membranes. The synergism of MXenes and MWCNTs-COOH resulted in a 132-fold boost in the PD (18.08 W/m^2) of a PVDF nanofibre-based PENG. This combination can be the future of portable and wearable electronics [186]. The design of an ammonia (NH_3) monitoring system was proposed, which was used for real-time breath analysers and ambient gas sensors. An edge-site-enriched MXene/ MoS_2 nanosheet heterostructure is extensively examined and employed as a sensing element and an active layer of an HyTPENG using first principles DFT simulations. The HyTPENG combined gas detection and energy harvesting capabilities into a single device, benefiting from a self-powered NH_3 monitor. DFT tests revealed that the constructed sensor has excellent sensitivity, reversibility, and sensitivity ($47\% @10 \text{ ppm}$) to NH_3 gas due to enhanced adsorbent surface and upgraded charge transport at the edge sites. The HyTPENG's motions (tapping, bending) successfully activate instantaneous triboelectric and piezoelectric PDs of 1604.44 and 15.62 mW/cm^2 , respectively. The HyTPENG developed using electrospinning offers remarkable flexibility and conformability that could be harnessed to develop wearable monitors for healthcare [187].

Metal organic frameworks (MOFs)

MOFs are hybrid organic/inorganic porous crystalline materials discovered recently. They comprise regularly arranged metal ions with +ve charge shrouded by organic 'linker' molecules. These metal ions cluster together to form nodes connected to the links' arms to form a hive-like configuration. MOFs have an extraordinarily high interior surface area due to their hollow structure, making them attractive for various applications. This material can be synthesized using a hydrothermal or solvothermal method, in which crystals are gradually grown from a hot solution. Gas storage, filtration, separation, catalysis, and sensing are all applications for this material. Efficient nanoparticles can be incorporated into the MOFs to form composites with both materials' properties. Hence, these materials find applications in developing TENGs and PENGs [188–192].

A MOF-based TENG intended to operate in the CS model was fabricated using zeolitic imidazole framework-8 (ZIF-8) (positive material) and Kapton film (Negative material). The TENG provided a

sustainable voltage and current output of 164 V and $7 \mu\text{A}$ [193]. Cyclodextrin (CD) is a green multifunctional material that is abundantly available and has non-toxic, biocompatible, and edible properties. The TENG developed from CD-MOF, sodium metal ions, and cyclodextrin ligand (α , β , and γ) exhibits a positive potential while scanning Kelvin Probe Microscopy (SKPM) can be added as a viable material in triboelectric series. The TENG developed from the various CDs are named A TENG, B TENG, and G TENG for α , β , and γ , respectively. The electrical outputs produced by various TENGs are in the order of A-TENG (152 V , $1.2 \mu\text{A}$) > G-TENG (116 V , $0.7 \mu\text{A}$) > B-TENG (90 V , $0.52 \mu\text{A}$) for various forces. The fabricated multiunit Z-shaped TENG device is employed for powering numerous low-power electronics by attaching them to the rear of the school bag and the shoe's heel (Fig. 7a) [194].

A TENG fabricated from latex/PTFE effectively powered an MXene/MOF derived copper oxide (CuO) gas sensor to detect ammonia (NH_3). Latex and PTFE have been used as +ve and -ve triboelectric materials with copper back electrodes. The value of V_{oc} and I_{sc} generated by TENG reached up to 810 V and $34 \mu\text{A}$, respectively, along with the highest PD of 10.84 W/m^2 , and was capable of lighting up 480 LEDs. The proposed sensor had an excellent response at room temperature extending its potential applications in pork quality monitoring [195]. MIL-88A (MOFs based on iron) synthesized by reacting iron chloride (FeCl_3) and fumaric acid in water is a good choice of biocompatible material for the TENGs (MIL-TENG). Various materials such as ethyl cellulose (EC), fluorinated ethylene propylene (FEP), and Kapton were used as an opposite layer for the CS mode TENG. MIL-88A and FEP provided a V_{max} of 80 V and a current of $2.2 \mu\text{A}$, proving they can be used for biomechanical gesture energy harvesting (Fig. 7b) [196].

TENGs based on the ZIF (Zeolitic imidazole framework) sub-family constituents (ZIF-7, ZIF-9, ZIF-11, and ZIF-12) are the latest trend in TENG material. The TENG was forged with ZIFs and Kapton as triboelectric layers. The vertical mode TENG with ZIF-7 sub-family exhibited a V_{max} of 60 V and I_{sc} of $1.1 \mu\text{A}$ and can drive low-power electronic devices (Fig. 7c) [197]. PENG based on a 2D MOF (with naphthylamine bridging)-reinforced PVDF composite nanofibres mat was able to deliver a V_{oc} and PD of

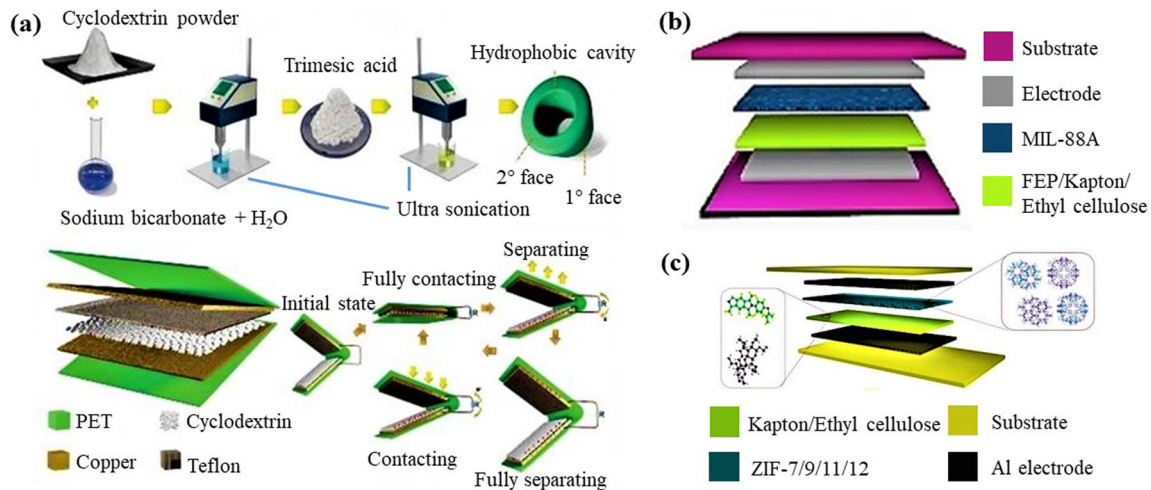


Figure 7 Metal organic frame work-based energy harvesters. (a) Synthesis, fabrication, and working mechanism of CD-MOF-based contact suppression mode TENG. Reproduced with permission from [194]. (b) Biomechanical energy harvesting to

run low power electronics. Reproduced with permission from [196]. (c) ZIF-TENG 3D layer view. Reproduced with permission from [197].

22 V and 24 $\mu\text{W}/\text{cm}^2$. The device also exhibited a PD of 6.25 μW and a sensitivity of 0.95 V/Pa against acoustic vibrations [198]. $\text{Co}_3[\text{Co}(\text{CN})_6]_2$ -based MOF was introduced as reinforcing fillers to enrich the piezoelectric response of PVDF-based nanocomposites. The unique open framework nanostructure and vast surface area of $\text{Co}_3[\text{Co}(\text{CN})_6]_2$ induced an interfacial coupling effect, resulting in enhanced piezoelectricity. When 0.6 wt% of $\text{Co}_3[\text{Co}(\text{CN})_6]_2$ was introduced into PVDF, the piezoelectric response of composites was improved by twofold ($d_{33} = 37 \text{ pC}/\text{N}$ and $d_{31} = 33 \text{ pC}/\text{N}$). The electromechanical coupling factor also showed a significant improvement $k_{31} = 0.135$, whereas pristine PVDF only had a value of 0.078 [199].

PVDF nanofibre membranes incorporating zirconium-based MOFs are reported as an excellent candidate for a wearable piezoelectric sensor for monitoring arterial pulse. The addition of 5 wt % of MOF improved the piezoelectric constant by 3 to fourfold without losing the flexibility of the fibrous mat. The composite also provided a V_{max} of 600 mV for a devoted force of 5 N. The sensor exhibited an incredible sensitivity of 0.118 V/N [200]. A twostep procedure synthesized an ultra-thin 2D ferroelectric nanoplatelet based on Ag–Ni MOF. The addition of 10 wt % of hybrid nanoplatelets in PVDF provided a maximum discharge density of 6.987 J/cm^3 ($\sim 2300 \text{ kV}/\text{cm}$) [201] and can be used for future PENGs material.

Layered double hydroxides (LDHs)

LDHs are ionic lamellar mixtures, a material classification with good physical and biological properties. They have many uses in drug delivery, energy storage, and catalyzing. Because of their excellent optical properties, they are also used for luminescence applications. Various techniques that can be employed for the synthesis of LDHs are co-precipitation, urea hydrolysis, and sol-gel synthesis. LDH polymer nanocomposites can be formed by various techniques, including the intercalation of monomer between LDH layers, subsequently in situ polymerization, direct exchange, and restacking of the exfoliated layers over the polymer [202–210].

A ZnAl-LDH-PVDF-based TENG, an eco-friendly triboelectric dielectric material with high performance, flexibility, and transparency, was devised. For a 20 wt% of ZnAl-LDH-PVDF, the TENG was able to deliver a V_{max} , current density, and PD of 230.6 V, 5.6 $\mu\text{A}/\text{cm}^2$, and 0.43 mW/cm^2 , respectively. A pressure sensor powered using this TENG exhibited an extraordinary pressure sensitiveness of 13.07 V k/Pa, whereas a humidity sensor responded to 259.4% in voltage detection mode (Fig. 8a) [211].

Although the concept of wound healing was put forward long ago, the subject is still well-researched. TENGs wounds, which function as tiny electrical stimulation (ES) instruments at the wound site, are good candidates for wound healing [212, 213]. A

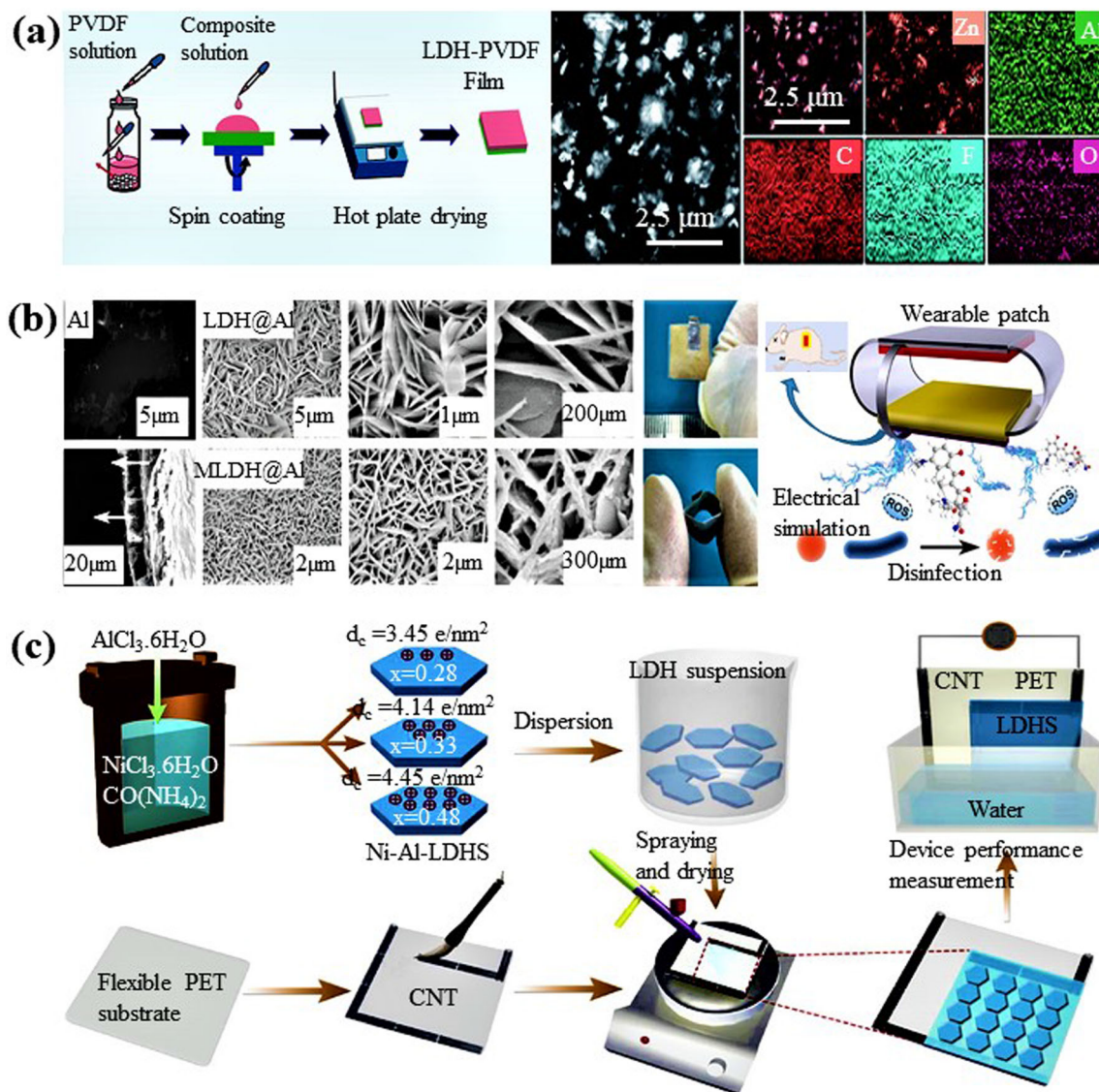


Figure 8 An overview of the LDH based energy harvesting and applications. **(a)** Fabrication of ZnAl-LDH-PVDF composite film, SEM/EDS images of ZnAl-LDH-PVDF composite film. Reproduced with permission from [211]. **(b)** SEM images of Al foil, LDH@Al, and LDH@Al film with minocycline MLDH@Al),

Surface engineered TENG for drug loading, Mouse wearing MSE-TENG for healing wounds. Reproduced with permission from [214]. **(c)** Fabrication and measurement of self-driven NWEs based on Ni-Al LDH films. Reproduced with permission from [215].

flexible TENG patch with magnesium–aluminium (Mg–Al) LDHs functioning as an intelligent medication receptacle and friction layer to speed infected wound healing was recently developed. The proposed TENG demonstrated excellent properties and was also remarkably efficacious in delivering minocycline (tetracycline antibiotics that prevent the growth and spread of bacteria to treat the infection). From the *in vitro* experiments, it was found that such patches were able to kill around 100% of *E. coli* bacteria (Fig. 8b) [214].

Energy generators driven by natural water evaporation (NWE) are comparatively innovative and cost-effective methods to generate electric energy. NWEs were fabricated by assembling Ni–Al LDH films (surface charge density of 2.52 to 4.59 e/nm^2), demonstrating an excellent positive correlation between the surface charge density, V_{oc} , and I_{sc} . The fabricated device displayed to V_{oc} of 0.6 V and I_{sc} of 0.3 μA along with an output PD of 15 $\mu\text{W/cm}^3$ for a $4 \times 1.5 \text{ cm}$ NWE. Powering a simple electronic watch was demonstrated with eight such assembled

NWEGs (Fig. 8c) [215]. High-performance flexible PENG based on PVDF and Zn:Al nanosheets have been recently reported. The bottom electrode was made by a sputtered aluminium-doped ZnO (AZO)/Ag/AZO multilayer, which also helped grow Zn:Al LDH nanosheets. The proposed energy harvester provided a V_{out} of 6.24 V and a current density of $0.655 \mu\text{A}/\text{cm}^2$. In contrast, the pristine nanogenerator provided a V_{out} and current density of 1.71 V and $0.19 \mu\text{A}/\text{cm}^2$, respectively [216]. Organically modified Ni-Co LDH (OLDH) nanosheets were used as fillers in electrospun PVDF nanofabrics to modify the morphological, crystalline, dielectric, and piezoelectric properties of the fabric. Adding OLDH into PVDF nanofabrics improved the polar β phase and dielectric constant while reducing the dielectric loss of PVDF. The proposed device was enough to power flexible and portable electronic gadgets providing a V_{out} of 6.9 V and PD of $0.92 \mu\text{W}/\text{cm}^2$ under human finger tapping for three wt% of OLDH [217].

LDHs find a broad spectrum of applications other than energy harvesting. These include catalysts for various chemical and biological reactions, flexible batteries, multifunctional materials for environmental remediation, supercapacitors, coatings, absorption, photo-thermal reaction, mineralization for agricultural soil remediation, and so on. Research is open for triboelectric, piezoelectric, and hybrid energy harvesting and storage using this material. The distinct types of energy harvester materials used recently and discussed so far are summarized in Table 3.

Emerging novel nanomaterials and energy harvesting

This section intends to discuss recent works published for a few emerging novel nanomaterials and their applications in the area of nano-energy harvesting. The invention and development of energy harvesting materials with advanced properties is an indispensable factor in the design and fabrication of nano-energy generators (NEGs). Numerous researches are focusing on engineering material, and it will be beyond the scope of this paper to describe them all, so a few recently developed novel materials as a counterpart for those existing are discussed below.

Lead-free Metal halide((BTMA)₂CoBr₄ (BTMA = benzyl trimethylammonium))

Due to their straightforward synthesis, mechanical flexibility, and designability, hybrid organic–inorganic piezoelectrics have gained interest. These materials have tremendous application potential in flexible sensors and self-powered energy harvesting systems. Despite discovering several hybrid piezoelectrics, most of these materials' architectures remain perovskite-type. Recently, the synthesis/structure/piezoelectric properties of a novel hybrid lead-free metal halide (BTMA)₂CoBr₄ were reported. The material, [CoBr₄]₂-tetrahedra and BTMA + cations, displayed notable piezoelectricity ($d_{22} = 5.14$, and $d_{25} = 12.40 \text{ pC}/\text{N}$), low Young's modulus (4.11–17.56 GPa) and low shear modulus (1.86–7.91 GPa). Thin films of the (BTMA)₂CoBr₄/PDMS composite were made and used in energy harvesting. The 10% (BTMA)₂CoBr₄/PDMS-based flexible devices outperform piezo-ceramic composites in energy harvesting with a V_{oc} of 19.70 V, I_{sc} of 4.24 A, and a PD of $11.72 \text{ W}/\text{cm}^2$. The device also demonstrated exceptional accuracy in detecting human body actions like finger bending and tapping. So, the potential of (BTMA)₂CoBr₄ and similar piezoelectric lead-free halides can be exploited as molecular materials for cutting-edge energy and sensing applications [218].

NiO-Mg magnetic nanocomposite

NiO–Ni–MgO ferromagnetic nanocomposite was developed from NiO–Mg by the mechano-chemical reduction process. The nanocomposite developed was employed as a positive triboelectric layer in the proposed TENG for biomechanical energy harvesting. The TENG demonstrated a V_{oc} of 35 V, I_{sc} of 130 nA, and a PD of $0.72 \mu\text{W}/\text{cm}^2$ across a load resistance of 100 M Ω for a device area of $2 \times 2 \text{ cm}^2$ [219].

Lead-Free Perovskite/Polymer nanofibre composite

Due to distinctive ferroelectricity and dielectricity, halide-perovskite-based mechanical energy harvesters produce an exceptional electrical output. Nevertheless, their elevated toxicity and moisture perceptiveness limit practical usage. A stretchy, breathable, and durable nanofibre composite was

Table 3 From energy harvesting to Applications

Material	Type of ENG	Source	Applications
Cellulose paper with PTFE [122]	TENG	Water droplets	Rain water energy harvesting
Bacterial cellulose [123]	TENG	Human motion	Self-powered wearable and implantable devices
Sandwich structured BC [124]	TENG	Interactive interface	Powering commercial electronics
AuNPs/PDMS [126]	PENG	Mechanical energy	To charge dielectric capacitor and to light up commercial LEDs
C/BT [129]	PENG	Mechanical input	Eco friendly energy harvesting
Direct current DC fabric [131]	TENG	Mechanical movements	To light up commercial LEDs
CNT/PEI [130]	TENG	Human motion, compressive force	Powering digital watch, calculator, Pedometer
PTFE/FT [132]	TENG	Human motion	Fire resistant
BFO [139]	Hybrid	Mechanical input	Efficient energy harvesting
PVA/Ag Nanowires [177]	TENG	Mechanical input	NO ₂ gas sensor
Nb ₂ CT _x /Ti ₃ C ₂ Tx [178]	TENG	Mechanical input	Improving TENG capability
CNFs/MXene [179]	TENG	Mechanical input	Self-powered wearable electronics
PVDF-TrFE/MXene Nanofibre mats [181]	PENG	Mechanical pressure	Pressure sensor, multifunctional electronic skin
Pvdf/MXene [184]	PENG	Mechanical input	Microdevice
Cyclodextrin [194]	TENG	Pressure	Shoe heels and school bags
Latex/PTFE [195]	TENG	Mechanical input	Ammonia sensor
MIL-[196]	TENG	Mechanical input	Biomechanical motion energy harvester
ZIF-TENG [197]	TENG	Mechanical input	To drive low power electronic devices
PVDF/Zirconium [200]	PENG	5 N	Arterial pulse monitoring
ZnAl-LDH-PVDF [211]	TENG	Mechanical input	Pressure sensor

developed by electrospinning lead-free perovskite/PVDF-co-hexafluoropropylene (PVDF-HFP) and styrene-ethylene-butylene-styrene (SEBS) simply LPPSNFC. The Cs₃Bi₂Br₉ perovskites act as proficient electron receivers and provincial nucleating vendors for polymer chain crystallization, improving the electron-trapping capability and polar crystalline phase in LPPS-NFC. The exceptional energy level matching between Cs₃Bi₂Br₉ and PVDF-HFP improves the electron transfer efficiency and lowers charge loss, enhancing the process of electron-trapping. As a result, the energy harvester devised of LPPS-NFC demonstrated a high electrical output (400 V, 1.63 A/cm², and 2.34 W/m²). Water resistance, stretchability, breathability, and robustness to extreme mechanical deformations are attractive features of this proposed material [220].

Antimony seleniodide (SbSeI)

A hybrid energy harvester exploiting the piezoelectric/triboelectric effects to harvest electrical energy

from mechanical energy using compressed (SbSe) was reported for the first time in 2022 by Krystian Mistewicz and his team. SbSe nanowires were fabricated by employing a sonochemical method. The nanowires were then compressed to a high pressure of 120 MPa to develop one of the electrodes, and the other electrode used was Kapton. The attractive feature of the proposed material is that the electrical loss is too small or negligible and hence can be accounted for future nanogenerator applications. The authors experimented with a triangular and rectangular excitation signal to drive the shaker. On applying a 50-gm load over the top electrode, the triangular shaker tip motion provided a V_{pp} of 0.38 to 1 V at a frequency of 90 Hz and 1.05 to 2.71 V at 80 Hz for the rectangular shaker tip motion. The proposed HNG powered up an LCD directly and an LED using a Graetz circuit [221].

NiO-Ti nanocomposites

NiO–Ni–TiO₂ nanocomposites with excellent structural, magnetic, and spectroscopic properties were synthesized by the NiO-Ti reduction technique. The enhanced magnetic and charge proliferation properties were investigated using density functional theory (DFT). The proposed nanocomposite had a dielectric permittivity of 298 and negligible loss factor of 0.098 at 1 MHz, and a surface polarity of 769 mV. The energy harvester was designed and fabricated to have a 3D-printed eye shape that works in CS mode. The proposed TENG demonstrated an output of 60 V and 600 nA [222].

Methylammonium lead tribromide (MAPbBr₃)

An HNG was recently introduced, fabricated from methylammonium lead tribromide (MAPbBr₃) single crystals (SCs)/PVDF nanocomposite. The sol-gel synthesis method was employed to develop and fabricate the nanocomposite layer. The MAPbBr₃-SCs-PVDF nanocomposite was used as a dielectric layer to design the TENG, and a similar layer was utilized to design a PENG. While comparing the result obtained for the single PENG, it was found that the output performance of the HNG was enhanced by 3.87 times (256 V), and the power efficiency was enhanced up to 200 times (16.17 mA/cm²) [223].

ZnFe₂O₄ nanocomposite films

A hybrid multimodal nanogenerator (HNG) comprising TENG, PENG, and EMG is designed and implemented using nanocomposite films of cuboctahedron zinc ferrite nanoparticles (CZF NPs) in polymer matrixes of PDMS/PVDF. The proposed device has a multispiral coil structure. CZF NPs @ PDMS nanocomposite film utilized the energy harvesting technologies of EMG and TENG, whereas the ferroelectric CZF NPs @ PVDF contributed to PENG. The HNG can support three different operating modes, viz. contact-separation (output voltage = 13 V), non-contact (output voltage = 0.65 V), and non-separation (output voltage = 1.2 V) mode, which is much more promising for future flexible hybrid nanogenerators [224].

Zirconium metal-organic framework and hybridized Co-NPC@MXene nanocomposite

Multilayered TENGs (M-TENG) finds great interest in developing enhanced performance energy harvesters, especially for self-powered biomedical applications. An M-TENG was developed out of two different layers; zirconium MOF (MOF-525) @ Ecoflex nanocomposite and Ecoflex @ cobalt nanoporous carbon (Co-NPC) @ MXene. The presence of MOF-525 in the first layer improved the proposed M-TENG's output about four times because of its highly homogeneous porous structure and charge accumulation. In contrast, the presence of Co-NPC in the second layer contributed to advanced charge trapping, and the MXene contributed to acting as microcapacitors. It should be noted that the M-TENG performed 13 times better when the second layer was used as an intermediate layer due to its high charge-capturing nature. The device proved to be a good choice for the self-powering (PD = 25.7 W/m²) biomotion/tactile sensor (sensitiveness = 149 V/KPa), with notable stretchability (245%) and humidity resistance. The device comprises low-frequency operation, wearable biomotion sensing, energy harvesting, real-time sensing, and human-machine interfacing [225].

Conclusions and future perspectives

The current review paper deals with the recent advancements in biocompatible materials that are suitable candidates for fabricating triboelectric, piezoelectric, and as well as for hybrid tribo-piezoelectric energy harvesters. The manuscript briefs the strategies for selecting materials for active layers of harvesters and common biocompatibility indicators to be checked while designing such harvesters. The article focuses on six critical materials as Cellulose, Fabric, Transition Metal Dichalcogenides (TMD), MXenes, Metal Organic Frameworks (MOF), and Layered Double Hydroxides (LDHs). These materials can power many electronic devices by generating energy from subtle movements. The limited performance of cellulose-based nanogenerators can be enhanced by using open-pore 3D structures such as aerogels, hydrogels, chemical modification by functionalization, doping using semiconductor materials

(ZnO, BTO) as well as TMDs, and electrospun nanofibres. The PD and voltage generated by these devices were increased with the processes mentioned above and hybridizing the device for both piezo/triboelectric effects. Hence, enriched cellulose is an eco-friendly substitute for synthetic polymers as piezo/tribo-active layers. One of the most significant problems with fabric is its flammability. However, by adding other energy-tolerant materials to the fabric, changing its characteristics, turning it into non-flammable clothing, and at the same time generating energy from subtle human movements, the benefits brought by the fabric to the field of wearable energy harvesters are indispensable. Fibre surface modifications by chemical grafting, doping with piezoelectric and triboelectric nanomaterials, electrospinning and 2D braiding technology with the nanocomposite materials are used to enhance the performance of these devices. The thus-made harvesters are used as self-powered wearable sensors for acquiring electrophysiological signals. These fabric products, Smart/e-textiles, with the ability to produce and extract high electrical output from subtle human movements in a way that does not cause harm to humans, may be the pillar of future research.

2-D TMD with more surface area is an asset for future power generation and energy conversion. Graphene and hBN, which have high energy efficiency, all fall into this category. The unique properties of high and controllable electrical and optical properties, flexibility, and stretchability make 2D TMD materials a leader in wearable energy harvesting. Designing self-powered implantable patchable nanodevices using these materials is an innovative strategy. Tailoring the surface of TMDs using ligands and adding these into piezoelectric polymers, enhancing nucleation sites, thickness modulation of nanosheets, electrodeposition for multiple active layers stacking, etc., are used for improving the overall capability of nanogenerators. These implantable patches find use in physiological signal monitoring and are an asset for future biocompatible biomedical applications. MXenes have excellent mechanical, electrical, thermal, optical, and chemical properties. Electrospun nanofibre of piezoelectric polymer with MXene, nanocomposite sheet stacking, etc., are utilized to improve the surface charge density, dielectric properties, gas sensing capabilities with self-powering, tensile properties and self-polarization effect. Improving flexibility, conformability

and self-powering properties, the energy harvesters can be used as wearable healthcare monitors in disease detection and tracking. Although MOF materials, commonly known as coordination polymers, have been discovered and exploited for nearly 20 years, this new type of material continues to be a source of research due to its unique properties and structure. Gas storage (hydrogen, methane), catalysis, solar energy conversion, electrical energy conversion and storage, and water electrolysis are essential milestones in this material research. Increasing their conductivity in photo-electrochemical applications, design strategies, preparation methods of composites and derivatives, application environment (high temperature, high pressure), finding and utilizing efficient materials, and fabricating high-quality devices become a challenge for the further use of this material. With functional nanomaterials, multifunctional LDHs have attracted more attention worldwide. Due to its structure, synthesis, and property relationship, this material possesses many applications and unique potential not found in other nanomaterials. LDH has been combined with other advanced materials in the energy field to produce energy harvesting, transport, and storage devices. These materials can be engineered to form emerging nanomaterials that can be utilized for developing hybrid nano-energy harvesters involving piezo/tribo-energy harvesting technologies. Therefore, LDH-based materials have a bright future, and there is much potential for focused research in this area.

Thus the article provides information of various recently developed biocompatible materials that apply to hybrid energy harvesting techniques, biomedical applications, biosensing, powering up various portable and wearable electronic devices and systems, developing flexible electronics technology, and so on. Even if many studies and research have already been done on these materials, their properties, new materials with unusual characteristics that can be made using them, and the new technologies that are suitable for them, the possibilities, development, and future of the materials are essential and are still open for research.

Acknowledgements

The authors would like to express their deepest gratitude to Mata Amritanandamayi Devi for being

there as our guiding force and inspiration. Thanks go to Amrita Vishwa Vidyapeetham, Amritapuri Campus, for providing the necessary facilities and an ideal environment to carry out this work.

Authors Contribution

SPR conceived the idea and guided the review. GMG contributed to writing the manuscript and figure design.

Declarations

Conflict of interest The authors declare no conflict of interest.

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