Nano-Structures & Nano-Objects 35 (2023) 100990

Contents lists available at ScienceDirect

Nano-Structures & Nano-Objects

journal homepage: www.elsevier.com/locate/nanoso

# Recent advances of triboelectric, piezoelectric and pyroelectric nanogenerators

Xiaoxiao Dong [a], Zhen Yang [b], Jiayi Li [c], Wenquan Jiang [a], Jianmin Ren [a], Yanting Xu [d], Travis Shihao Hu [d], Ming Li [e,*]

[a] College of Mechanical Engineering, Liaoning Petrochemical University, Fushun 113001, Liaoning, China
[b] School of Mechanical Engineering and Automation, Northeastern University, Shenyang 110819, Liaoning, China
[c] School of Information and Control Engineering, Liaoning Petrochemical University, Fushun 113001, Liaoning, China
[d] Department of Mechanical Engineering, California State University, Los Angeles, CA 90032, USA
[e] Centre for Advanced Structural Ceramics, Department of Materials, Imperial College London, London, SW7 2AZ, UK

ARTICLE INFO

*Article history:*
Received 26 February 2023
Received in revised form 28 April 2023
Accepted 31 May 2023

*Keywords:*
TENG
PENG
PyNG
Artificial intelligence
Wearable electronics

ABSTRACT

Nanogenerators are widely used in harvesting and converting mechanical or thermal energy into electric energy. It is a promising way to capture energy from the surrounding environment as portable power supplies and self-powered systems, offering great versatility and feasibility. Current studies focus on designing nanogenerator structures and materials with high power outputs, multifunctionality and low-cost characteristics. In this review, we will introduce the technological advances of three common nanogenerators, namely, triboelectric, piezoelectric and pyroelectric nanogenerators. The fundamental components, including the working mechanism, structure design, materials selection and main applications, are compared and systematically discussed. Current challenges and future perspectives are highlighted with a focus on offering new insights into developing the next generation nanogenerators that are highly integrated, multifunctional and of upgraded performance.


## 1. Introduction

Climate change and energy depletion are both significant international crises facing the world today, so the rational use and distribution of energy is of paramount importance. Reducing existing energy consumption, finding new energy sources and collecting different types of energy in our daily life for energy conversion are effective solutions. Of these, finding new energy source and methods of energy harvesting is more practical in the long run [1–3]. Capturing as much energy as possible from the surrounding environment is one of the prevalent research topics today. Among the various energy harvesting systems, the nanogenerator (NG) is widely acknowledged by scientists because of its astonishing working mechanism and high energy conversion efficiency with a minimal amount of waste [4–7].

Unlike the ordinary generators, which need a certain speed to generate voltage, NGs can generate electricity by collecting the small scale of mechanical or thermal energy in the ambient environment to change the micro physical structure of nanomaterials, which generates static electricity and the flow of electrons between two materials [8–10]. Common NGs can be roughly divided into three types according to the different generation mechanisms, namely, triboelectric nanogenerators (TENG), piezoelectric nanogenerators (PENG) and pyroelectric nanogenerators (PyNG). Among them, TENG and PENG convert external mechanical energy into electrical energy, while PyNGs convert heat energy into electrical energy through temperature changes over time [11]. There are some reviews on nanogenerators which are based on their different materials. The materials include various types, such as aerogel [12], $BaTiO_3$ [13], cellulose [14], carbon [15], lateral nanowire or nanobelt [16], etc. These reviews help readers to understand the nanogenerators from material aspects, and we will not explain thiS ASPECT in this review any further.

Besides advancement/improving energy generation technologies, NGs are gaining popularity and developing rapidly due to their many advantages, such as being self-powered, sustainably harvesting energy from the environment, together with their light weight, low cost and environmental friendliness. This kind of self-powered system has broad application prospects (intelligent humanoid robot, motion tracking, human-interactive interfaces, physiological signal monitoring etc.). For example, NGs could be used to monitor things like pulse signals in the medical field, and in the textile field, garments prepared by the combination of fibers and nanogenerators can realize signals to sense for energy harvesting or signal monitoring [17–21]. However, the NG system has a limited electrical power output, multifunctional capability,

* Corresponding author.
*E-mail addresses:* jmren661216@163.com (J. Ren), m.li19@imperial.ac.uk (M. Li).

https://doi.org/10.1016/j.nanoso.2023.100990
2352-507X/

stability and service life [22–26]. On the basis of understanding the working mechanism of different kinds of NGs, the core of improving the performance of NGs is through ingenious structural design and material selection [27,28]. Although developments and advances are becoming available, there remains a large gap between the best performing NGs and real-world applications. Designing rational structures and materials for NGs are urgent issues, which need addressing for the process of commercialization and industrialization in the future.

In this review, we provide a comprehensive overview of three standard NGs, namely TENG, PENG and PyNG, including the working mechanism of three common nanogenerators (Section 2), compositional structures (Section 3), materials (Section 4) and the emerging applications (i.e., sensors, wearable electronics and artificial intelligence) (Section 5). The challenges and perspectives of NGs are also discussed at the end of the review. Although this article describes the basic situation of three common nanogenerators, some details can be further expanded. We hope that this paper can provide a comprehensive framework for readers who want to understand the field of nanogenerators.

## 2. Nanogenerators working mechanism

Understanding the working mechanism of various NGs helps to design structures and select material, to achieve the desired performance by optimizing the relevant parameters. This chapter systematically introduced the working mechanism of three common NGs, TENG, PENG and PyNG, and briefly discussed the advantages and disadvantages of these kinds of NGs.

### *2.1. Triboelectric nanogenerator (TENG)*

Triboelectric nanogenerators (TENGs) generate static electricity by contact electrification, which is where two materials come in contact, electricity charges swap between them which leads to one material being positively charged while the other one is negatively charged [29–31]. The electric cloud-potential well mode explains the foundation mechanism of a TENG. The combination of the electrification effect and electrostatic induction converted various kinds of random and low-frequency mechanical energy existing in the environment into electric energy. When exposed to external stimuli, the displacement between the layers of the TENG changes, causing a change in the number of positive and negative charges on dielectric fabric and conductive fabric. The accumulated potential difference between the two layers of the TENG promotes the flow of electrons to generate a current (Fig. 1(a)) [32–35]. Relevant studies also show that temperature and curvature play an essential role in charge transfer during tribological generation. The hotter sides are prone to be positively charged, while the cooler sides are inclined to be negatively charged. Due to the thermionic emission effect, electrons are transferred from a hotter part to a cooler one. Besides that, irregular surfaces also affect the electric charges. Convex sides tend to be negatively charged, while concave sides prefer to be positively charged. These basic theoretical studies provide essential guidance and reference for the design and preparation of TENGs.

The mode of TENGs can be classified into four types: contact-separation (CS) mode, lateral sliding (LS) mode, single-electrode (SE) mode and freestanding (FS) mode. In the next chapter, the four modes are discussed in detail. Overall, TENGs offer low costs, high output performance, stability, short response time and high sensitivity. The future development trend of TENGs is closer to integration and miniaturization [36].

### *2.2. Piezoelectric nanogenerator (PENG)*

PENGs generate electric power through external mechanical stress and material deformation, known as the piezoelectric effect [37,39,40]. The piezoelectric effect is introduced when a piezoelectric material is subjected to an external force, the electrical charges are exchanged between the materials and the accumulation of charges at both ends of the material forms an electric dipole. The electric dipole moment is generated by the deformation of the orientated non-centrosymmetric crystal structure or by the presence of a porous electret with a persistent charge in the pore (Fig. 1(b)) [41–43]. In general, the main reason for the formation of piezoelectric potential is due to the relative displacement between the cations with respect to the anions. Precisely, due to the charges of the cations and anions coinciding with one another, the piezoelectric materials do not have polarization inside at the beginning. When the material is subjected to external pressure, it deforms and generates the negative strain and the volume decreases. The charging center separates to form an electric dipole, and the electric dipole distance also changes, thus generating a voltage potential. This is the conversion process from mechanical energy to electricity [44–47]. Due to the two conducting electrodes being in full contact, the maximized pressed state is achieved with the highest polarization density. When the external force is released, the electron reflux rebalances the charge caused by the strain release under the short circuit condition. When the measuring system is connected in reverse, the signal of output voltage and current will be reversed. The piezoelectric coefficient of piezoelectric materials measures the conversion efficiency from mechanical stress to electric energy, the higher the piezoelectric coefficient, the higher the piezoelectric material's energy conversion efficiency [48–51].

On the whole, PENGs have high sensitivity, fast response, low energy consumption and can be effectively used for the detection of dynamic pressure signals, however, due to the working mechanism, PENGs cannot detect static pressure, so the application scope of PENGs has limitations [52,53].

### *2.3. Pyroelectric nanogenerator (PyNG)*

PyNGs convert free heat in the environment into electrical energy based on the Seebeck and pyroelectric effects [54–58]. The pyroelectric effect depicts temporary voltage generation by the thermoelectric materials under transient conditions [59]. Studies show that the higher the temperature, the more kinetic energy the charge carrier has, and the kinetic energy of a hot charge carrier is higher than that of a cold one, which makes the hot carrier move to the cold side. Therefore, the open circuit voltage obtained between the two ends of a material is subject to a temperature gradient. When the charge carrier concentration reaches equilibrium, an electromotive force will be generated at both ends of the conductor and signal sensing will be realized according to the Seebeck effect of the material (Fig. 1(c)) [38,60–62]. To ensure efficient generation of thermoelectric power, since the Seebeck effect greatly dependent on the temperature gradient and the electricity flow, the thermoelectric materials are typically semiconductors or semi-metals with a combination of reasonable electrical conductivity and Seebeck coefficient.

PyNGs do not have to undergo mechanical deformation like TENGs and PENGs, which make PyNGs have great potential for a powering wearable electronic system. For example, in cold conditions outside the winter, to make full use of the temperature fluctuations between the environment and the human body, through the polarization changes between the electrodes to convert heat energy into electricity [63].

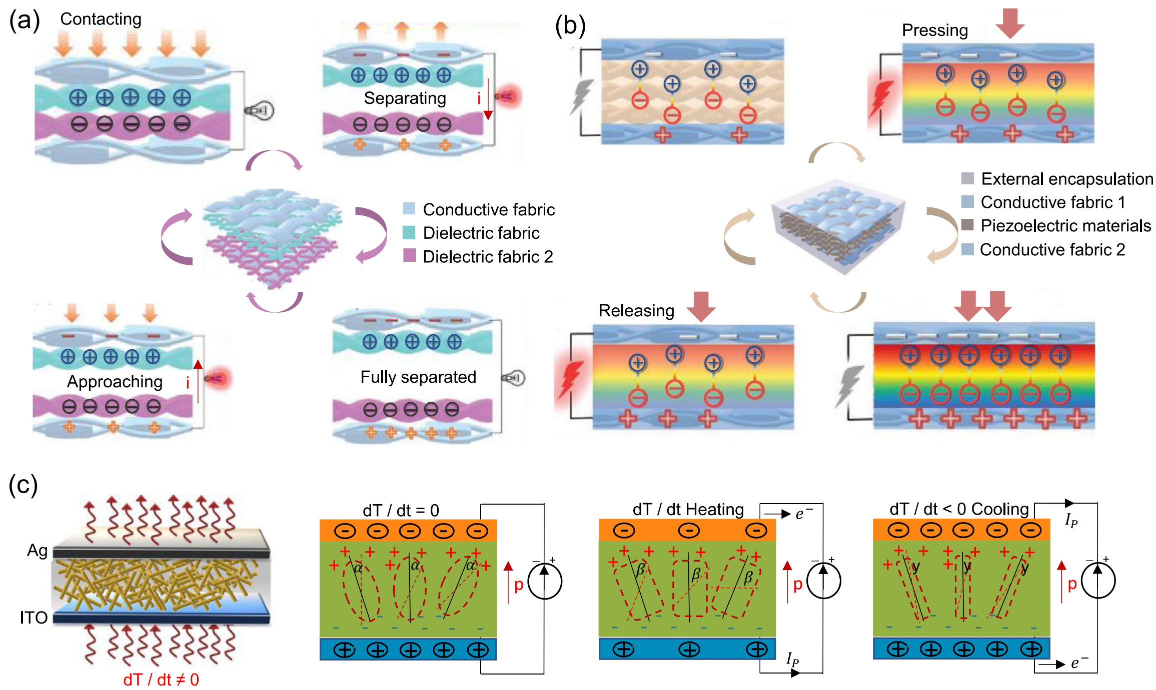


**Fig. 1.** The working mechanism of different NGs, (a) TENG. (b) PENG [37]. Copyright 2019, Wiley-VCH. (c) PyNG [38]. Copyright 2012, Elsevier B.V.

## 3. Compositional structures

The structure has a critical effect on the performance output of nanogenerators. Among various usage scenarios and functional characteristics, the structure of TENGs and PENG can be classified as a single structure, forming structure and multilayer stacking structure. In addition to that, the forming structure can be further categorized into 2D and 3D. Specifically, PyNGs have combined other structural NGs together to reach a high energy conversion efficiency in recent years.

### 3.1. Triboelectric nanogenerator (TENG)

As mentioned in chapter 2, TENGs have four modes. Although the four TENG types have similar working mechanisms, their structures are quite different [36]. The contact-separation (CS) mode of a TENG is composed of two different triboelectric polarity materials and electrodes, which requires a large gap. Under the action of an external force, the two materials produce vertical movement and generate charge on the surface of the contact (Fig. 2a (i)) [64–66]. The CS is more convenient for sensing pressure changes and producing high output voltage, however, the downside is the pulse output [67,68].

The lateral sliding (LS) mode uses two materials to form triboelectric through external forces in the horizontal/ rotational direction, and this mode almost does not require a gap (Fig. 2a (ii)) [69]. The advantages of the LS mode are high frequency, continuous and high electricity output. However, this mode is prone to damage the material surface and has poor long-term stability [70].

Unlike the CS and LS structures, the single-electrode (SE) mode has only one electrode and one friction layer (Fig. 2a (iii)). The TENG in this mode uses the external object as another layer of friction material and triboelectrification through the interaction of the external environment with itself makes it easy to integrate with other components/devices or to carry along. It is also beneficial for harvesting energy, while the structure of this mode is relatively simple. However, the disadvantages of this mode are low output and signal instability [71].

The structure of the freestanding (FS) mode consists of two fixed symmetric electrodes and external objects move freely between the two electrodes. This type of TENG is suitable for detecting multiple forms of the moving objects to get a signal for change (Fig. 2a (iv)). The drawback is that this mode is inconvenient to move due to fixed electrodes and complex integration [72]. In addition to the four basic modes of TENGs, the specific structural design of a TENG can be divided into a single structure, forming structure and multilayer stacking structure.

The single structure is made by attaching a dielectric polymer to the electrode material by winding and wrapping, having a similar structure to a spinning thread. The advantages of this structure are simple, low cost, good performance and suitable for large-scale preparation [73–75]. However, due to the limitation of the electrode material's properties, its overall scalability needs to be improved. To solve this problem, the electrode material can be wound on a stretchable fiber surface, or a conductive material can be prepared into a helical structure which is built into the dielectric polymer [76–80]. Usually, this single structure is suitable for the CS and SE modes, where the CS mode has higher electrical output and better stretchability than the SE mode. In most cases, the CS mode of this structure is designed with a core–shell structure to avoid abrasion, in which a gap exists between the inner core column and the outer shell tube. In this situation, the structure of the CS mode can be classified into four types: the dielectric layer wrapped around an inner electrode as the core with the outer electrode as the shell is type I [81–84], the outer electrode with the same configuration as the core is type II [85,86], the inner electrode wound on a dielectric fiber as the core with the same configuration as the shell is type III [87–89], while the outer surface of type III further covered by a dielectric or encapsulation layer is type IV [90,91]. Type I is the simplest structure. Types II and III have gradually evolved to prevent charge leakage and to expand the application scenarios. Finally, type IV was developed to improve stretchability. One of the most important design principles is that materials with different triboelectric polarities are chosen for the outer side of the inner core column and the inner side of the outer shell tube [92].

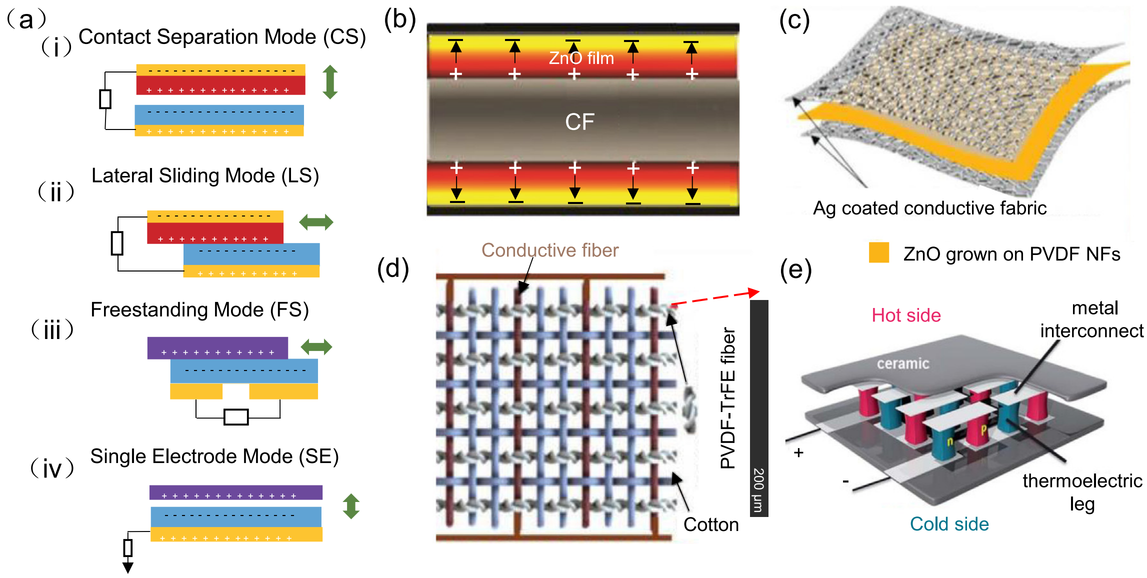


**Fig. 2.** (a) CS, LS, FS and SE four basic modes of TENGs [36]. Copyright 2019, Elsevier Ltd. (b) The single structure of a PENG [97], Copyright 2011, Wiley-VCH. (c) The forming structure of a PENG [98], Copyright 2018, The Authors, Published by MDPI. (d) The stacking structure of a PENG [99], Copyright 2015, American Chemical Society. (e) The structure of a PyNG [100]. Copyright 2012, the Royal Society of Chemistry.

To improve the electrical output and expand the width of applications, the fibers or straps can be woven in 2D/ 3D forming structures. The cooperation of various materials and weaving methods can improve the electrical output performance of TENGs more efficiently [93]. In addition, to facilitate the preparation of a high-performance TENG, for example by protecting electrodes and preventing charge leakage, the TENG can be designed and prepared with a multilayer stacking structure [94,95]. For example, Seung et al. grew ZnO nanorods on an Ag-coated common fabric, coated them with a layer of polydimethylsiloxane (PDMS), then coated the top side with a layer of silver-coated fabric. This multilayer stacking structure can efficiently improve the overall TENG power output performance and mechanical robustness [96].

### 3.2. Piezoelectric nanogenerator (PENG)

The PENG structure is divided into three parts consisting of a metal–insulator–metal sandwiched structure, which has two metal electrodes with an insulating piezoelectric layer between them [101].

Preparing a PENG with working stability, mechanical endurance, piezoelectric output performance and other characteristics is the direction that researchers have been working hard on. To refine and enrich the functional features, the structures of PENGs are continuously improving. In the beginning, a simple single structure appeared, wrapping flexible piezoelectric materials on high strength and high modulus fibers. This method relies on the relative motion of fibers to generate power, so it is difficult to be applied in daily wearable usage (Fig. 2b) [97,102]. Subsequently, by spirally winding fiber electrodes on the outer layer of the selected piezoelectric fiber, a PENG prepared in this way can effectively collect human motion energies. The outer electrode can be directly deposited on the surface of a piezoelectric fiber by the vacuum deposition method, which makes it have great application prospects in human body wearing [103,104]. The studies show that the hybrid PENG structure composed of a piezoelectric polymer coated on the surface of nano-structured piezoelectric materials can improve the mechanical properties and polarization output characteristics [105–107]. Moreover, preparing a PENG with a core–shell nanostructure can provide higher polarization possibility between the inner and outer electrodes [108]. In addition, the PENG can be designed with a spiral multilayer cladding structure, which helps increase the piezoelectric composite's active area, thus improving the power generation efficiency
[109–111].

To further improve the piezoelectric outputs of PENGs, similar to the structure of TENGs, one strategy is to combine multiple piezoelectric fibers into fabrics by using textile forming techniques, such as woven, double-arc shaped, knitted, microporous and looping structures (Fig. 2c) [98]. Zhou et al. used poly(3,4-ethylenedioxythiphene) (PEDOT)-coated poly (vinylidene fluoride) (PVDF) electrospun yarns to weave high pressure-sensitive 2D fabric-based PENGs [112]. The single fiber or strap weave sometimes results in lower power outputs. To solve this problem, various fibers or straps can be used for 2D fabrics and the short circuit phenomenon can be well avoided by changing the material of the fibers or straps used [113–116]. To integrate more piezoelectric components into PENGs to enhance the energy harvesting capability of the PENGs, researchers have woven fibers or straps into 3D structures [117,118], and have even used multilayer stacking structures of multiple materials. Choosing functional materials for the integrated design of a multilayer network structure can give the prepared multilayer stacked structure better piezoelectric outputs (Fig. 2d) [99,119–123]. For example, Maity et al. designed an all-organic PENG based on multilayer assembled electrospun PVDF nanofiber mats. They used PVDF NFs as the active piezoelectric component and placed PEDTO-coated PVDF NFs as the electrodes through the vapor phase. This multilayer network structure integrated with compatible electrode enhances the output voltage and current (Voc $\sim$ 48 V, Isc $\sim$ 6 μA) and shows ultra-sensitivity towards human movements [124].

### 3.3. Pyroelectric nanogenerator (PyNG)

PyNGs are composed of many thermocouples connected electrically in series and thermally in parallel in order to achieve a thermo-induced potential of a few volts to power small electronic devices, motors or to charge a battery. PyNGs usually consist of three parts, which are an upper metal layer, a middle-polarized layer and a lower metal layer. The upper metal layer is designed for the sufficient collection of heat, the middle layer converts thermal energy into electricity through internal polarization and the lower metal layer acts as the lower electrode [125]. The device is sandwiched between ceramic layers to avoid short-circuiting between the metal interconnects and to ensure an excellent thermal exchange with the surroundings. The additive effect of the thermo-voltage created by each thermocouple is only possible if their legs are connected. The n-type leg has a negative Seebeck coefficient, i.e. the electrons thermally diffuse to the cold side, while in the p-type leg a temperature gradient causes the propagation of holes towards the lower energy region. An electromotive force is induced in the circuit as a force acting against further charge carrier migration (Fig. 2e) [100]. In addition, on researching for pyroelectric energy conversion, the Olsen cycle (also called Ericsson cycle) can also be adopted, where a pyroelectric material is sandwiched between two electrodes in contact with a cold ($T_C$) and a hot source ($T_H$), while applying low ($E_L$) or high electric fields ($E_H$) to perform the Olsen cycle [126–131].

PyNGs are often combined with other kinds of nanogenerator to design and produce multifunctional nanogenerators. Wang et al. prepared a coupling nanogenerator based on a single structure and realized the coupling effect of friction-piezoelectric-thermal by selecting and modifying the materials. Since a single friction-piezoelectric unit has the defect of a low voltage and a single pyroelectric unit has the problem of a low current, neither of them alone can provide power for two in-series light bulbs. However, the synergies between the two units, which gets the most energy from the surroundings, combine the nanogenerators to light the series bulbs [132]. Zi et al. also reported a friction-piezoelectric-thermal hybrid NG, where mechanical energy is collected by a sliding TENG and a pressing PENG. The sliding motion causes a change in the surface charge and the pressure causes a change in the internal polarization of the material, creating an electric current that flows through the external circuit. In addition, heat generated by the friction of the selected material causes a change in polarization within the device to form an electric current. This coupled configuration of the structured device successfully realized the temperature and normal force of the self-power supply [133].

## 4. Materials of nanogenerators

The materials of a nanogenerator directly affect the performance and efficiency of the nanogenerator. Materials are undoubtedly one of the most important elements in the preparation of NGs. In this chapter, we mainly introduce various NG (TENG, PENG and PyNG) materials, including the electric materials and electrode materials.

### 4.1. Electric materials

Electric materials are mainly used to convert external energy into electricity, which is the most important part of the NGs' materials. The design of such materials is key to improving the performance of NGs. Due to the different working mechanisms of TENGs, PENGs and PyNGs, there are also great differences in the selection of their electric materials (Fig. 3). This section will give a specific introduction into the electric materials of different NGs.

#### 4.1.1. Triboelectric materials

The fact that a material becomes charged after frictional contact with another material is a triboelectric effect induced by contact [134–137]. Physical contact between materials usually produces opposite charges and the strength of the charges varies from material to material. The triboelectric series is a tool used to describe triboelectricity, which ranks different materials according to their tendency to gain or lose electrons, depending on the material's physical properties [138,139].

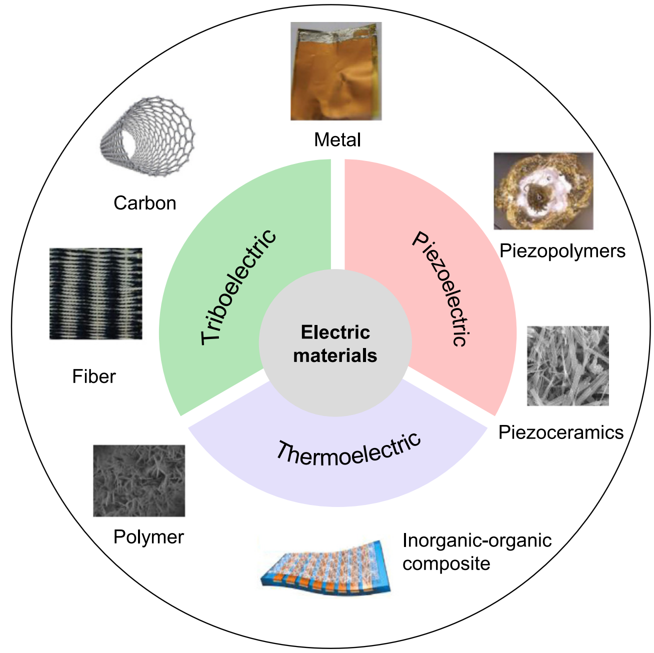


**Fig. 3.** Typical electric materials of three NGs.

Based on the working mechanism and structure, the selection of triboelectric materials for TENGs considers the ability to gain and lose electrons; the larger the electron affinity gap between the friction layer materials, the higher the charge transfer efficiency and the higher the electric energy generated through the triboelectric effect [140–142]. The electrical output properties of TENGs depend on the induced potential difference between the two paired tribomaterials. To get a larger contact surface area, the surface of the friction layer material is usually roughened so that more charge transfer can be accepted to improve the TENG's sensitivity and electrical output performance [143,144]. The triboelectric series helps select which materials can generate the most static electricity through friction for the preparation of high-performance TENGs. In the earliest period, Wilcke et al. established the first triboelectric series by listing around ten common materials in order of polarity [145]. Shaw and Henniker extended the triboelectric series by adding natural and synthetic polymers to the sequence [146,147]. Recently, Zou et al. updated the triboelectric series by normalizing the triboelectric charge density (TECD) of a wide range of polymers, regarding their triboelectric polarization in a well-controlled environment to show the intrinsic physical properties of the materials [148]. In addition, the reports of an expanded series of textile-related triboelectric materials, including 21 commercial and new fibers, have significant application potential in TENGs [149,150].

In general, common friction layer materials in TENGs mainly include polymers, metals, fibers etc [151]. Zhang et al. summarized 100 articles about TENGs, among which 14 commonly used electron acceptor materials and 20 electron donor materials were detailed [152]. In addition to these traditional materials, Hajra et al. combined non-toxic, biocompatible and edible organic cyclodextrin in their selection for a TENG. They used a cost-effective ultrasonic synthesis to fabricate alpha, beta and gamma cyclodextrin metal–organic frameworks (CD-MOFs) at temperature. The highly crystalline CD-MOFs were pressed uniformly on a copper substrate as the triboelectric layer of the TENG, which showed great potential application value. This work effectively expands the application of MOF materials in TENGs [153]. Due to the lack of a multifunctional material in the triboelectric series, combining feasible new materials is one of the effective methods to improve the performance of TENGs. Therefore, researchers can try the feasibility of applying new materials in the field of TENGs.

#### *4.1.2. Piezoelectric materials*

Piezoelectric materials generate voltage/current by sensing external pressure. Electrical output properties of PENGs mainly depend on the polarization charge of the piezoelectric materials. Generally, piezoelectric materials can be divided into piezoceramics and piezopolymers. Piezoceramics have a high piezoelectric coefficient, rigidity and brittleness, which limit their application in some flexible devices. The piezoelectric effect is associated with noncentrosymmetric crystals. Natural materials like quartz and berlinite have the piezoelectric effect. In engineering materials, zirconium titanate (PZT), zinc oxide (ZnO) and barium titanate ($BaTiO_3$) will be endowed with piezoelectricity after the process of polarization. In addition, the most studied systems are perovskite, ilmenite, bismuth-layer and tungsten bronze structure ferroelectrics. Among many similar materials, piezoelectric materials with a perovskite structure usually exhibit a more obvious piezoelectric effect [154,155]. Hao et al. reviewed perovskite type piezoelectric ceramics. It is pointed out that compositional engineering and microstructural engineering approaches can enhance the strain and stability properties of perovskite piezoelectric ceramics [156].

Concerning piezopolymers, polyvinylidene fluoride (PVDF) and its copolymer polyvinylidene fluoride-trifluoride (PVDF-TrFE) are common materials. This kind of material has a good comprehensive performance, such as high sensitivity, high chemical resistances and high resistance to mechanical damage [157–159]. This type of piezoelectric polymer is flexible, but its piezoelectric coefficient is low. To further improve the mechanical endurance, piezoelectric output performance and match to ambient vibrations, piezoceramics can be incorporated into the piezoelectric polymer, by adjusting the composition and the proportion of the two, to get the ideal performance for a PENG, such as PZT, BT, ZnO, KNN-based PVDF [160–162]. For example, Tian et al. design a self-powered rich lamellar crystal baklava-structured PZT/PVDF piezoelectric sensor for real-time monitoring of table tennis training and scalable sports. This composite developed sensor has an excellent sensitivity of 6.38 mV/ $N$ and an ultra-fast response time of 21 ms [22].

#### *4.1.3. Thermoelectric materials*

Thermoelectric materials can be divided into inorganic, organic and inorganic–organic composite thermoelectric materials, at present. Like piezoelectric materials, inorganic thermoelectric materials usually have a high thermoelectric conversion efficiency, however, their poor flexibility limits their application in flexible devices. Organic thermoelectric materials usually have good flexibility, but a low conversion efficiency [163,164]. Common inorganic thermoelectric materials are mainly metal compounds, such as bismuth telluride ($Bi_2Te_3$), tin selenide ($SnSe_2$), silicon germanium alloy etc [165,166]. Organic thermoelectric materials include graphene, carbon nanotubes (CNT) and poly (3, 4-vinyl dioxy thiophene):poly (styrene sulfonic acid) (PEDOT: PSS) etc [167–169]. To account for the conductivity, conversion efficiency and flexibility, some studies have combined organic and inorganic thermoelectric materials, which is currently the most common material usage of PyNGs [170–173].

In addition, the pyroelectric coefficient is a property of the material itself, which is related to the output performance of the PyNG, and the pyroelectric coefficient can be changed by controlling the crystallinity, strain coupling effect and polymer modification. For example, the pyroelectric properties of a PyNG can be enhanced by ion modification, forming a self-polarized ferroelectric $\beta$ phase and a porous structure [174–177]. Therefore, in order to increase the performance of PyNGs, researchers consider on how to select and improve materials, enhancing the pyroelectric coefficient [178,179]. For example, Kim et al. controlled the crystallinity of P(VDF-TrFE) with a high dipole moment solvent to enhance the pyroelectric coefficient [180]. Lee et al. introduced stretchable PyNGs with a thermally induced stain coupling effect in order to enhance the output performance of the PyNG using P(VDF-TrFE) and PDMS [181].

### *4.2. Electrode materials*

The electrode materials play the role of induced charge, and they has no major influence on the performance of NGs. According to the requirements of the application field, the selection of electrode materials should take into account the conductivity, flexibility, service life and cost factors. Besides, some nanogenerators also need to consider environmental factors, so the preparation of nanogenerators should also take into account the corrosion resistance and wear resistance [182–185]. Nowadays, the most commonly used electrode materials are metal, carbon-based materials and conductive polymers [186–188]. Among the metallic electrode materials, aluminum and copper are popular because of their high conductivity and low cost. Here, we summarize the electric materials and electrodes of different kinds of NGs, as shown in Table 1. Some typical examples are listed here and the application of these NGs is also briefly summarized.

In addition to the above introduction of the basic content, there are several hybrid types of NGs. Various characteristics of the materials are core to the preparation of hybrid NGs, such as the design of the preparation materials, at the same time as the triboelectric, pyroelectric and piezoelectric properties etc. At present, many researchers tend to study hybrid NGs, due to hybrid NGs having the advantages of the different kinds of NGs at the same time, which can provide important value for applications in the field of NGs. This kind of hybrid NGs has a lot of related reviews, so this will not be expanded more here [189–193].

## 5. Applications

The applications of NGs are ubiquitous and endless, in this chapter, we focus on three applications: there are sensors (one of the first applications to appear), wearable electronics (currently the most common application) and artificial intelligence (future application trend).

### *5.1. Sensors*

The application of NGs in sensors has attracted wide attention due to its obvious effect. NG sensors can be divided into strain sensing and thermal sensing, according to the way the NGs' materials respond, and the common one is strain sensing. In addition, NG sensors can be divided into mechanical motion and fluid sensors, according to the solid–liquid state of the NG material; the mechanical motion sensor has been studied relatively more [210–213]. NGs sensors have many application scenarios, such as intelligent sports, health monitoring and security. In this section, we introduce the NGs used as sensors applications from above mentioned areas.

When a NG sensor is used in the monitoring of biological health signals, the sensor collect signals from biological motion tracking and gesture recognition to reflect the real-time statistics of biological motion and behaviors. Liu et al. fabricated a self-powered and miniaturized endocardial pressure sensor (SEPS) based on TENG. They implanted the SEPS into a pig heart model and tested its sensitivity to 1.195 mV mm $Hg^{-1}$. Experiential and

**Table 1**
Electric and electrode materials for different kinds of NGs.

| Types of NGs | Electric materials | Electrode materials | Performance | Ref |
|---|---|---|---|---|
| | Cu, FEP | Copper foil | The acceleration of the object can be detected, the sensitivity is 0.080 V $s^2$/ m | [194] |
| TENGs | PDMS | Multi-wall carbon nanotubes | Can recognize finger movements, with the sensitivity of 1.5 $kPa^{-1}$ | [195] |
| | FEP, Acrylic acid | ITO | Can detect human eye movement and release an electrical signal of 750 mV | [196] |
| | PDMS, PMMA | PMMA with gold films and nanoparticles | Self-lighting shoes and self-lighting tiles which can be made luminous upon footfall | [197] |
| | PDMS | Graphene | Detects finger movements by touch with a minimum pressure of 1.3 kPa | [198] |
| | PET, PDMS | ITO | The voltage and current density can reach 18 V and 0.13 μA, excellent stability (∼100000) | [199] |
| | PDMS, Al | Al | A tremendous current density of 0.10 mA $cm^{-2}$ and a voltage output of up to 130 V could be obtained | [200] |
| | PVDF, FET | Silver ink coating | The sensor that can be monitored at least 300 Pa | [36] |
| PENGs | PVDF | PZT | The sensitivity and ultra-fast response time of sensor are 6.38 mV/ N and 21 ms | [22] |
| | IDE@PE | PZT-GFF | Efficient energy harvesting (∼60 V, ∼500 nA) and multi-mode (bending and pressing) energy harvesting ability | [201] |
| | $BaTiO_3$ nanoparticles, P(VDF-TrFE) | Silver flakes | Can be stretched to more than 300% strain | [202] |
| | Nanofiber mat | Cu foil | Producing an outstanding open-circuit voltage (∼68 V) and short circuit current (∼0.1 μA) with a power density of 53.2 μW/$cm^2$ | [203] |
| | P(VDF-TrFE), $CsPbBr_3$ QD | Cu | Exhibit an optimized piezoelectric coefficient (24.5 pC $N^{-1}$) | [204] |
| PyNGs | PEDOT: PSS | silver | Can produce a voltage of 0.52 V and a power of 0.32 μW | [205] |
| | PVDF | Al | Self-powered human respiration and temperature sensor, producing a 42 V open-circuit voltage | [206] |
| | Graphene | Graphene | Can detect the strain of objects, and the response time is less than 0.6 s | [207] |
| | P(VDF-TrFE), PDMS | Ag/Ag NWs, Au | Enhance output performance | [208] |
| | PDMS-CNTs | Graphene nanosheet | Realize stretchable hybrid PyNGs for wearable electronic devices | [209] |

theoretical studies were performed to verify that SEPS has good mechanical stability and responds well to stress (Fig. 4a) [214]. Yi et al. prepared a highly flexible and self-powered TENG with a sandwiched structure to achieve biomechanical energy harvesting and real-time biometric authentication. They sewed the TENG sensor onto socks and used the frequency difference caused by foot speed to distinguish between walking and running. They also developed a self-powered wearable keyboard that could be used to record physiological signals by integrating the large-area TENG sensor array. The results show that the TENG sensor prepared in this study can monitor the movement signals of human body very well (Fig. 4b) [215].

There are many similar works that have been done using various kinds of materials and these studies will not be covered in detail here [216–220]. In addition to the common types of materials described in the previous chapter, there are several other materials studied. For example, Shuai et al. used hydrogel to fabricated a TENG sensor to monitor hand gestures [221]. Apart from the monitoring of biologically related vital signals, another interesting study is the large-scale health monitoring of structures. Zhang et al. developed a multifunctional composite rebars with an embedded TENG mechanisms and demonstrated the feasibility of embedding structural elements to detect damage patterns in concrete beams at multiple scales, providing a pioneering study in the field of architecture [222]. The research on TENGs as sensors is relatively more abundant, however, research on PENGs as well the TENG and PENG combination as sensors has also become popular in recent years. In general, the research of NGs as sensors has been perfected, but further improvement is needed in the aspects of sensing accuracy, preparation cost, time-consuming installation, etc.

### 5.2. Wearable electronics

Wearable electronics is one of the most important applications of NGs in recent years. They are often used in medical treatment and motion detection at present [37,223–225]. Wearable electronics need to be folded and worn continuously, like clothing, and can collect and store energy continuously through human activities. The energy of the surrounding environment is converted into electrical signal sensing to provide communication, interaction, monitoring, perception and other auxiliary functions for

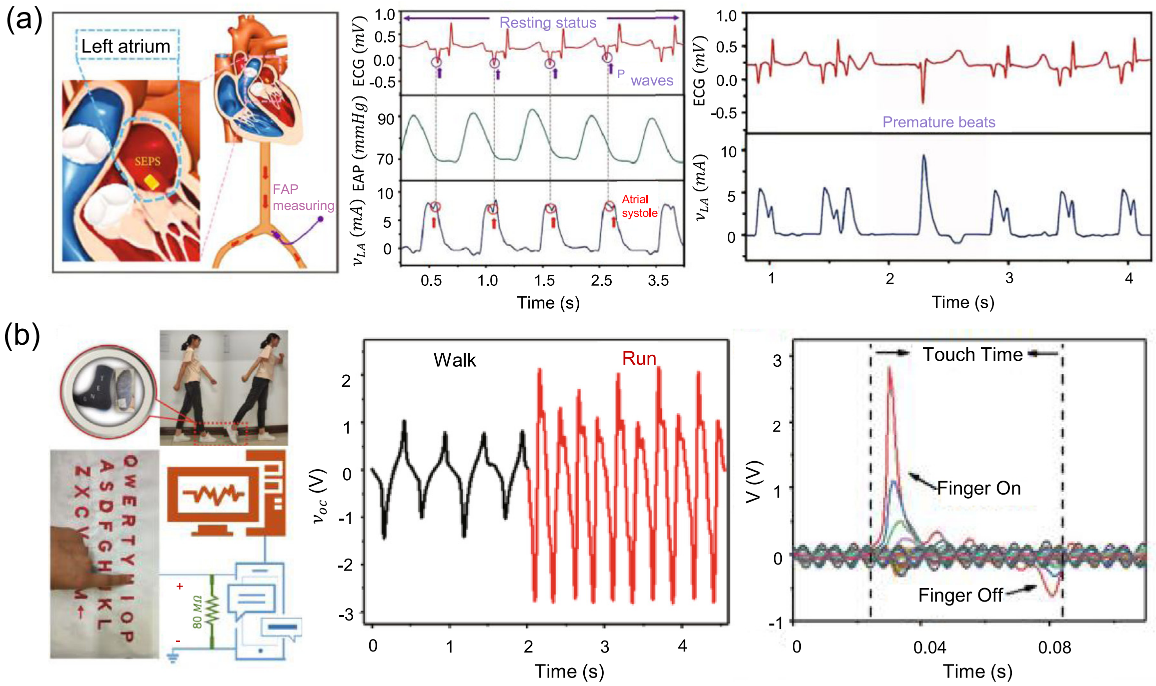


**Fig. 4.** (a) A commercial arterial pressure sensor measuring the electrocardiogram and the SEPS outputs [214]. Copyright 2019, Wiley-VCH. (b) The schematic and collected data diagram of a prepared TENG sensor used as pedometer and the self-powered keyboard [215]. Copyright 2021, the Spring Nature.

human life. Therefore, nano-generators that can be used as wearable electronic products need good energy conversion efficiency and service life [226–228]. The first structure of a wearable self-powered system was proposed by Wang's group. In this work, they prepared a supercapacitor and wearable TENG combined power system using fiber optic materials. By testing, its specific capacitance can reach 83.5 F $cm^{-3}$ and its capacitance can still be maintained at more than 94% after 5000 cycles. The main material of the wearable electronics for the TENG is PDMS, which is woven into a jacket through optical fiber materials. During jogging, the voltage can be increased to 8 mV within 10 s, realizing the power supply of wearable electronic devices [229]. Wang's group has reported a number of research projects in the field of wearable electronics using NGs and one of the studies was for medical surveillance. They developed a flexible self-powered sensor based on a TENG that can reliably measure human pulse and blood pressure on many parts of the human body (Fig. 5a). PTFE and PET are used as two friction layers, ITO as the electrode, and finally PDMS is used for packaging. The schematic diagram of the signal received from the sensor is shown in Fig. 5b. The sensor has an ultra-high sensitivity (45.7 mV $Pa^{-1}$), ultra-fast response time (less than 5 ms) and no performance degradation was observed for up to 40,000 motion cycles [230]. Their group also reported a scalable machine-programmed TENG, which was used to collect energy for human movement, with an output voltage of 232 V and a power density of 66.13mV/$m^2$. The integration of a small-size power management module, converting an irregular alternating current into stable DC output, can be very good in order to achieve a self-power supply. By testing the stretchability, washability and air permeability, it has been shown that the prepared TENG has great application prospects in the field of self-powered wearable electronics and smart textiles (Fig. 5c and 5d) [231].

There are many other studies like this and in general the application of nanogenerators in the field of wearable electronics is relatively perfect at present. It should be noted that the materials selected for the preparation of the TENG used in wearable electronics should have an excellent self-power ability, and the prepared TENG should have good service life, tensile and bending properties, and have washable resistance.

### 5.3. Artificial intelligence

Artificial intelligence (AI) is the simulation of the intelligence process of human beings by robots, which aims to realize an intelligent working state through technology. AI can usually identify people based on their unique physiological characteristics, such as face, voice, smell, fingerprint, heartbeat, etc. In the future, NG applications will also be combined with AI, which can play an important role in medical treatment, communication, transportation and other fields. In recent years, research on the combination of NG and AI has become a trend in the field of NG applications, and some state-of-the-art advances have been published. In these researches, NGs are mainly prepared into sensors to collect signals from the human body and then combined with AI for data processing, so as to realize remote control in a human–machine interactive (HMI) system [22,234,235]. For example, Yang et al. prepared a PENG device by growing 2D $SnS_2$ through CVD, which generates different voltages (off, I, II, III) under different bending states of the human index finger, and the $SnS_2$ PENG device can reach an open-circuit voltage and short-circuit current of 6 mV and 60 pA, respectively. In addition, the PENG devices can be used as a drive unit of the synchronous human–robot interface for an advanced smart sign language system and realizes human–machine synchronous control (Fig. 6a) [234]. Nanogenerators can also be used as flexible acoustic sensors for speech recognition and artificial synapses, which convert the original speech sound into analog electrical data to further machine learning [236–241]. Liu et al. developed a distinctive structure for a self-powered synapse transistor (SPST) to emulate synapse functions. The voltage was provided by a TENG without using additional voltage to generate a pre-synapse spike. When two input spikes (−1 V, 30 ms) from TENGs were both applied on a gate as the pre-synaptic input terminals, along

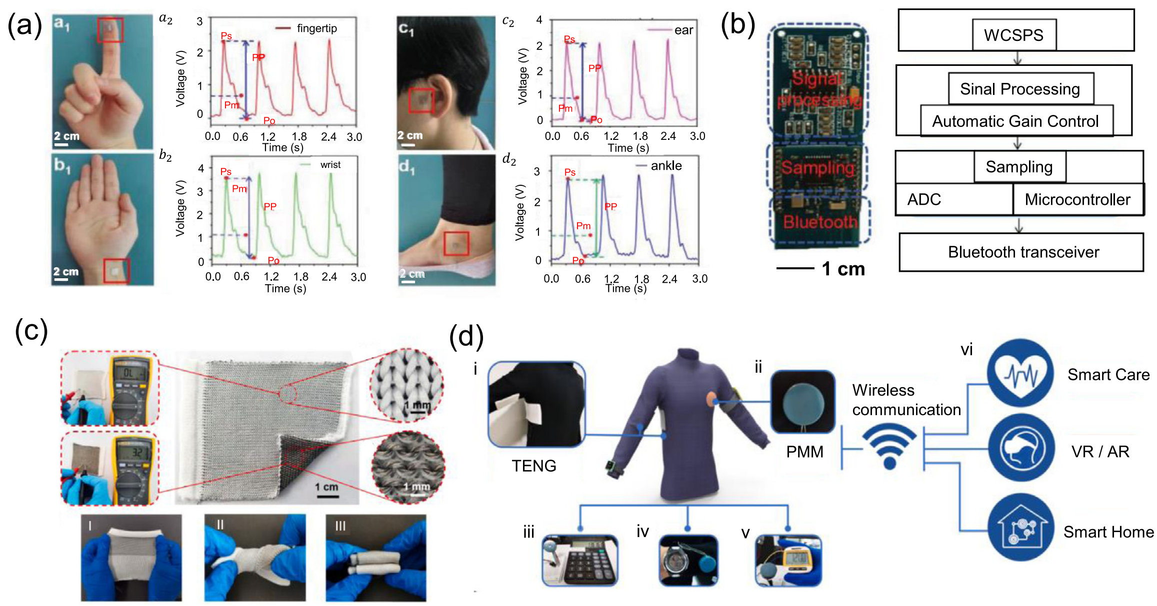


**Fig. 5.** (a) A nanogenerator used to test electrical signals in various parts of the body (fingertip, wrist, ear and ankle). (b) A schematic diagram of the signal management circuit [232]. Copyright 2018, Wiley-VCH. (c) The structure of the TENG used for wearable electrics. (d) The schematic illustration of various self-powered smart electronics driven by the TENG [233]. Copyright 2021, Elsevier Ltd.

with any input positive spike (1 V, 30 ms) from Tm, the "AND" logic function (1 μA threshold current) was achieved. The "OR" logic function can also be realized. The results show that the SPST can combine signals from multiple inputs and realize logic functions, which will be of great benefit to future AI (Fig. 6b.). Furthermore, Pavlov's dog experiment also proves that SPST can be applied to touch research experiments and has an important contribution to neuromorphological engineering (Fig. 6c) [242].

Recently, there has been related research on the combination of NGs and AI for network security. Maharjan et al. reported a new self-powered electromagnetic-triboelectric based hybrid nanogenerator for keystroke dynamics driven biometric authentication systems integrated with neural network-based AI (Fig. 6d). They developed a set of customized data acquisition and a signal processing system by acquiring and processing the key information. Their ANN-based AI model for user identification and authentication achieved a high accuracy of 99% in user authentication using hybrid nanogenerators, even under a common password among multiple users. On the same key dimension, the hybrid nanogenerators offer high accuracy and double security compared with individual nanogenerator based authentication (Fig. 6e). This work combines the nanogenerator as a sensor with the field of AI, providing a new possibility for a high cybersecurity layer in the computation world [246]. In general, the application of AI with NGs shows great potential for the future, such as flexible neuromorphic cognitive platforms, human–computer interaction, face and voice recognition. However, it is still at a preliminary stage and faces many difficulties (recognition accuracy, learning response speed, etc.) to be overcome in future developments.

## 6. Conclusion and perspectives

In this work, we have comprehensively reviewed three kinds of common NGs (triboelectric, piezoelectric and pyroelectric), specially focusing on the working mechanism, structural design, materials selection and applications (Table 2.). Moreover, it was necessary to introduce and discuss the current research status, future development trends and faced challenges of NGs.

Regarding the structure design, TENGs and PENGs are mainly developed from a single structure at the beginning and then are developed into a multilayer stacking structure later on to improve the output efficiency of the NGs further. PyNGs have similar structure as the TENGs and PENGs. PyNGs are frequently combined with other kinds of NG to obtain high performance, multifunctional compound NGs. Regarding the materials, we introduced electric and electrode materials, which are essential for the NGs' preparation. The NGs' electric material plays an important role to convert external energy into electric energy, which is related to their performance. The electrode materials just play the role of induced charge. Finally, the fields of sensors, wearable electronics and artificial intelligence have been introduced, which may prove to be possible applications for NGs. The future is likely to see an increase in the use of artificial intelligence.

The development of NGs is relatively complete in terms of theoretical research, structural design material selection and application, however, there is still a large gap between research and practical commercial applications. To produce high-output, multifunctional, low-cost NGs, the future development trend will be around the structural design and different functional materials selection. The structure design of NGs needs to be concerned with the use situation, and another trend will be towards higher performance output and miniaturization. Regarding material selection, improving output, promoting multi-functionality and low cost is the core of all NG development. TENGs can expand the sequence of friction materials by developing new materials. PENGs currently have issues with poor reliable stability, large hysteresis behavior with a strong nonlinearity, which can be solved by manipulating the grain size and configurations of domains. To improve the efficiency of PyNGs, substances with high thermal conductivity can be doped with electrode substrates on the top and bottom, or by combining with other types of NGs. At the same time, developing new materials with tribo-electric/piezoelectric/pyroelectric power generation and hybrid NGs will help advance the progress in the field of NGs. Regarding the applications of NGs, it is necessary to improve the sensing sensitivity and accuracy in the field of sensors. Stretchability, flexibility, comfortability, biocompatibility, washability and

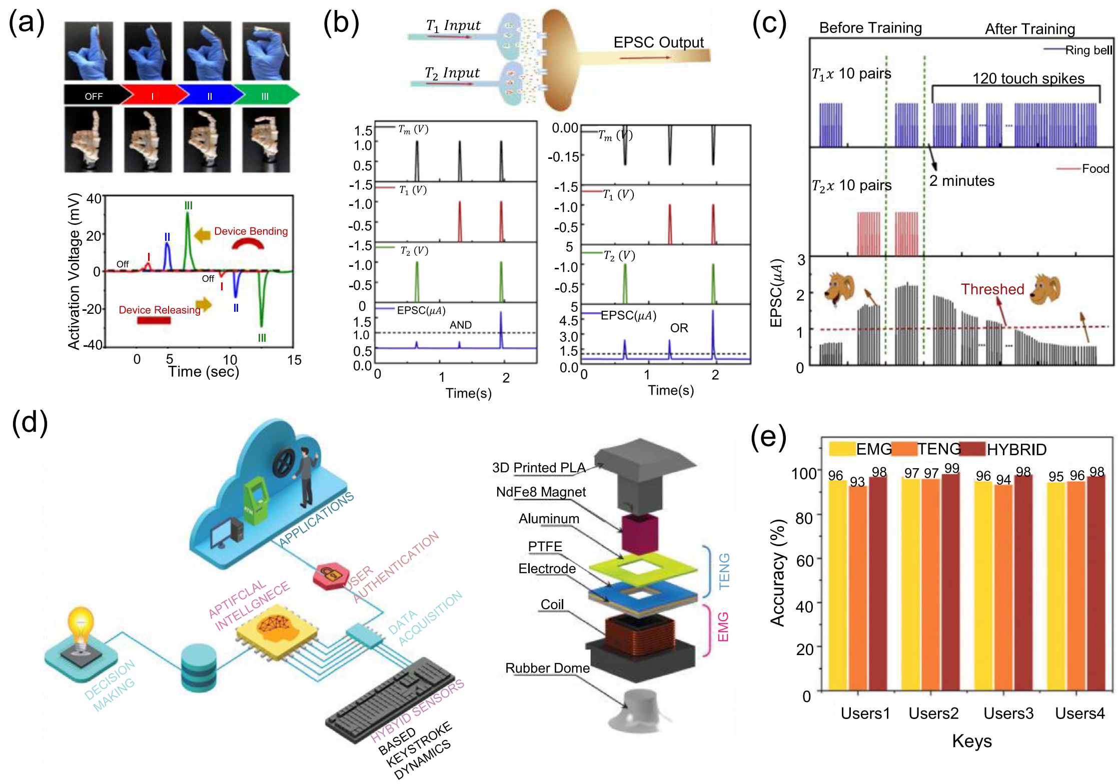


**Fig. 6.** (a) $SnS_2$ PENG device corresponds to activation voltage under different bending states of the human forefinger. Copyright 2020, Wiley-VCH [243]. (b) The schematic of synapse with multiple pre-synaptic inputs and Input–output characteristics of the "AND" and "OR" pulse logic tuned by positive spikes. (c) The schematic of the concept of Pavlov's dog experiment for associative memory [244]. Copyright 2019, Wiley-VCH. (d) The self-powered hybrid sensors driven keystroke dynamics based biometric authentication system framework using AI. (e) Accuracy score matrices for different nanogenerators [245]. Copyright 2021, The Authors. Advanced Science published by Wiley-VCH GmbH.

**Table 2**
Comparative summary of the compositional structures, materials and applications of various NGs.

| Category | Compositional structures | Materials | Applications | Ref |
|---|---|---|---|---|
| TENGs | Single structure | Metal, Carbon fiber | Wearable electronics | [247] |
| | | Metal rubber, Stainless-steel fiber | Sensors | [248] |
| | 2D/ 3D forming structure | Fiber, Polymer | Wearable electronics | [249] |
| | Multilayer stacking structure | Metal, Fabric | Wearable keyboards | [250] |
| | | | Wearable cloth | [251] |
| | | Metal, Polymer | Wearable electronics | [252] |
| | | | Artificial intelligence | [253] |
| PENGs | Single structure | Metal nanosheet | Artificial intelligence | [254] |
| | 2D/ 3D forming structure | Polymer nanofiber | Sensors | [255] |
| | | Liquid metal | Understanding mechanical properties | [256] |
| | | Fiber | Energy harvesting | [257] |
| | Multilayer cladding structure | Nanowire | Wearable Energy-Harvesting | [258] |
| PyNGs | Multifunctional hybrid structure | Polymer, Nanofiber | Sensors | [259] |
| | | Polymer nanowire, Nylon | electronics | [260] |
| | | Metal, Polymer film | sensors | [261] |

permeability are important factors to be further considered to fabricate wearable electrodes. In addition, pairing artificial intelligence with NGs may provide an avenue for further research and improvements of NGs.

As a whole, the NGs make use of the semiconductor properties of nanowires and the piezoelectric effect to realize sustainable and accessible energy generation. NGs have simple and diverse structures, stable output and high energy conversion efficiencies. NGs play an important role in the sustainable development of human beings and the design of NGs provides a new direction for green energy. However, there is still a big gap between the practical application effect of NGs and the ideal effect at present. NGs are an emerging research area and there are many challenging issues to be solved. (1) Developing multiple structural couplings and miniaturizing the structures of NGs to meet more application situations. (2) Reasonable design, selection and development of new NG materials, so that NGs can have increased practical application. (3) Improving the output efficiency, stability and service life of the NGs' self-powered system to enhance the performance

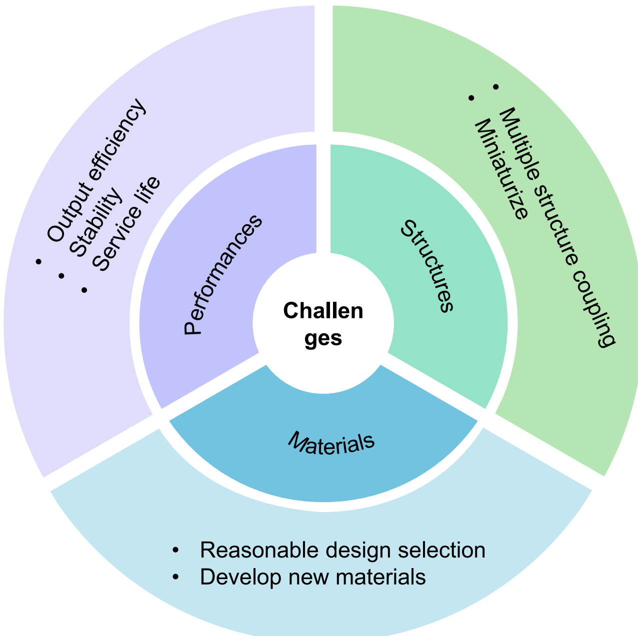


**Fig. 7.** The challenging issues facing NGs.

of NGs and expand the development process of NGs in the future (Fig. 7).

## CRediT authorship contribution statement

**Xiaoxiao Dong:** Writing – original draft, Visualization, Investigation. **Zhen Yang:** Data curation, Writing – original draft, Visualization, Investigation. **Jiayi Li:** Writing – original draft. **Wenquan Jiang:** Writing – original draft. **Jianmin Ren:** Supervision, Writing – review & editing, Funding acquisition. **Yanting Xu:** Writing – original draft. **Travis Shihao Hu:** Conceptualization, Writing – review & editing, Funding acquisition. **Ming Li:** Supervision, Writing – review & editing, Funding acquisition.

## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Jianmin Ren reports financial support was provided by Scientific research fund Project of Education Department of Liaoning Province. Travis Shihao Hu reports financial support was provided by U.S. National Science Foundation.

## Data availability

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

## Acknowledgments

This work was supported by the Talent scientific research fund of LSHU (No. 2023XJJL-008) and Scientific research fund project of the Education Department of Liaoning Province, China (LJKMZ20220725) and the U.S. National Science Foundation, USA (Nos. HRD-2112554, HRD-1547723). Y. X. appreciates the support from the NSF CREST-CATSUS fellowship, USA. M.L. acknowledges the financial assistance from Imperial College London, UK in the form of the "President Scholarship" (01790264). All authors discussed and commented on the manuscript.

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