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Overview of Triboelectric Nanogenerators

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Ambient energy is abundant in the environment and takes various forms, such as solar irradiation [1], thermal gradient [2], mechanical deformation [3], and so on, which can be converted into electricity using ambient energy-harvesting techniques [4]. The approach to harvesting energy from the environment is one of the ideal solutions to respond to the energy demands of distributed autonomous microsystems, which should be sustainable, renewable, and of high performance [4]. Ambient energy-harvesting technology provides an attractive future vision to realize fully integrated self-powered microsystems that overcome the drawbacks of batteries, which currently need to be frequently replaced, or of laying out long wires for power supply [5]. In 2012, a new ambient energy-harvesting mechanism named triboelectric nanogenerator (TENG) was developed by combining the triboelectrification effect and electrostatic induction [6, 7]. This novel technology has proved to be a robust power source to directly power commercial electronics and even regular light bulbs [7]. There has been a remarkable growth of TENG research in the past years due to its unique properties, including high-output performance, cleanness, sustainability, etc. [8, 9]. Mechanical energy from sources such as wind, raindrops, and ocean waves, as well as body motions can be efficiently converted to electric power using TENGs. Therefore, this chapter summarizes the current progress of microenergy technologies and then introduces an overview of TENG.

1.1 Energy Crisis of Microsystems

In the past half century, as the benefit of the rapid development of electronic science and technology, human society gradually changed to automation, both intelligent and digitization, and various electronic devices have become part of our life and are distributed everywhere. The featured size of modern electronic devices becomes smaller year by year down to the millimeter, and even to micrometer levels, which induces a continuous decrease in power consumption down to milliwatts and even to microwatts. Consequently, the

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great reforms in small-size and low-power consumption promote the integration of multifunctional electronic devices to realize microsystems.

Microsystems experienced a blooming development in the past decades resulting from their unique features, i.e. portable, smart, and miniature. However, the further development of microsystems suffered several critical challenges, especially the exploration of appropriate power sources. At present, batteries are still the first option, especially some of the flexible batteries, but the problems of sustainability and pollution caused by batteries cannot be ignored. In the meantime, distributed autonomous microsystems also have some new energy demands such as sustainability, renewability, high performance, and even flexibility or stretchability.

As an essential example of microsystems, Internet of things (IoT) is expected to play an important role in economic and social development of the next generation, as shown in Figure 1.1. Thus, IoT is taken as an example to describe the energy crisis of microsystems [10].

In principle, an IoT system is composed of three main parts: sensing network, interconnection network, and terminal network. The sensing network detects the various changes of environmental factors and transforms them as electronic signals. Subsequently, the electronic signals are transferred to the interconnection network and treated to form control signals. Eventually, these control signals are delivered to the terminal network to drive functional electronic devices to respond to the corresponding changes. Therefore, the sensing network serving as the interface media between environment and client is the essential component of the IoT. The sensing network consists of trillions of sensors, which are widely distributed in the environment, especially in autonomous states.

Consequently, exploring an appropriate power approach for the sensing network is an urgent issue for the rapid development of IoT. Micro energy

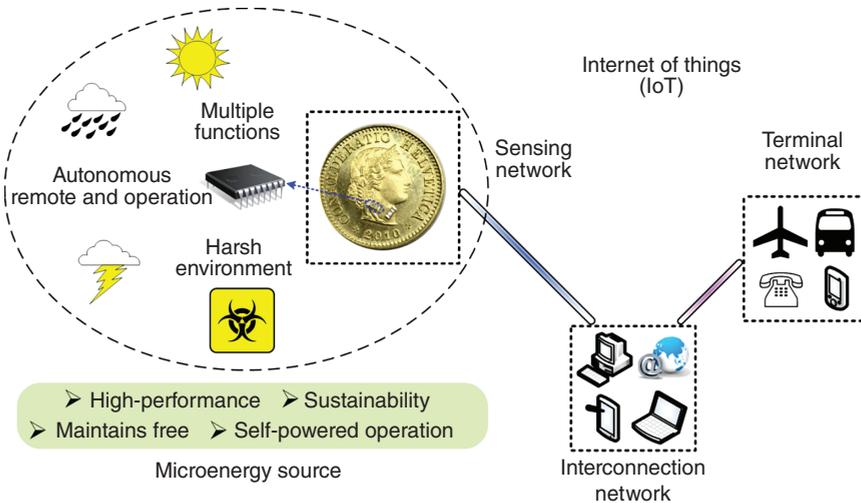
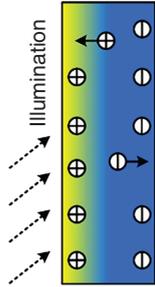
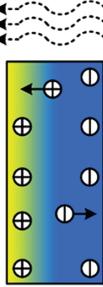
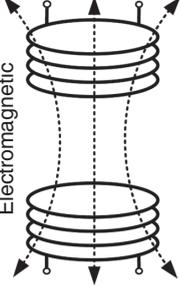
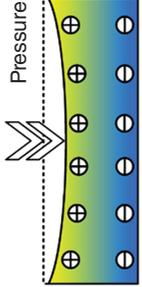
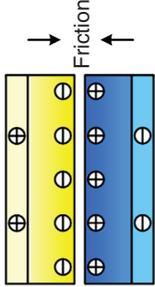


Figure 1.1 Schematic view of the Internet of things (IoT) and its power supply requirements. It is of great significance for the development of IoT to realize the appropriate microenergy source that fits the unique requirements.

Table 1.1 Comparison of five technologies for ambient energy harvesting.

Type	Schematic view	Voltage (V)	Current (A)	Power density (mW/cm ²)	Efficiency (%)	Pros versus cons
Photovoltaic (PV)		0.5–0.9	100–500	5–30	0.3–46	High output power, continuous DC output, good basis of industrial fabrication For the flexible organic solar cell, the conversion efficiency is still very low, only works under light
Thermoelectric (ThE)		0.1–1	5–30	0.01–3	0.1–25	Sustainably working as a DC power source, easy to scale down, no moving component Low conversion efficiency, low output performance, large temperature difference
Electromagnetic (EM)		0.1–10	N/A	N/A	5–90	High conversion efficiency, high current with low voltage, resistive impedance For wireless power transmission: short range, working at certain frequency; For magnet moving: heavy weight and big size, complexity
Piezoelectric		1–200	0.01–10	0.001–30	0.01–21	Highly sensitive to external excitation, easily integrates and miniaturizes in micro-/nanoscale Low conversion efficiency, low output performance, pulse output, high impedance
Triboelectric (TrE)		3–1500	10–2000	0.1–100	10–85	High output and energy conversion efficiency, no materials limitation, remarkable flexibility Pulse output, high impedance, friction damage

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area is limited for self-powered flexible microsystems, only flexible or wearable configurations of these five technologies are emphasized.

1.2.1 Photovoltaic Effect

The solar cell is one of most popular power sources based on photovoltaic effect. The basic law of photovoltaic effect is that electrons overcome the potential barrier and are excited to a higher energy state by absorbed lights. The frequency of light must exceed a certain range in order to possess sufficient energy to overcome the potential barrier for excitation, and then the separation of charges leads to the establishment of an electric potential [4, 11].

So far, solar cells can be divided into five main categories, including multi-junction cells, single-junction GaAs, crystalline silicon cells, thin-film technologies, and emerging others [12]. The first three types possess better performance, with the energy conversion efficiency (ECE) ranging from 21.2% to 46%, and silicon-based solar cells dominate the commercial market [12]. Nevertheless, because of fragile and nonflexible characterization, it is difficult to apply them in wearable microsystems.

By contrast, the latter two classifications show good flexibility by fabricating specific functional materials on polymeric substrates, but their ECEs are still at a relatively low level, with copper indium gallium selenide (CIGS), perovskite, and dye-sensitized solar cells reaching the highest values of 23.3%, 22.1%, and 11.9%, respectively [12].

1.2.2 Thermoelectric Effect

As is known, the human body itself is a perfect energy source to provide thermal dissipation and physical movement [13, 14], which is regarded as an attractive solution to satisfy the power demand of wearable electronics. The thermoelectric effect is employed to transform the energy generated by the thermal dissipation of the human body to electricity. When a temperature difference is applied to thermoelectric devices, an electrical potential that can drive the flow of electrons in the circuit loop to generate the electricity will be established, which is named the Peltier–Seebeck effect [4, 15].

Thermoelectric generators can be classified into inorganic and organic. Inorganic thermoelectric generators are made from inorganic materials, such as several alloys and intermetallic compounds based on elements like Bi, Te, Sb, Pb, etc., which are, as a matter of fact, toxic [16].

Organic thermoelectric generators are usually manufactured using conductive polymers (i.e. conjugated polymers and certain coordination polymers) and small molecules (i.e. charge-transfer complexes and molecular semiconductors) [15]. They have attracted much attention because of the properties of light weight and outstanding flexibility. However, the *ECE* needs to be strengthened, which is still at a relatively low level of less than 25%. The *ECE* of the thermoelectric generator is defined as a function of the figure of merit (ZT), average working temperature, and the temperature difference between the hot and cold ends. Thus, compared with *ECE*, the figure of merit, i.e. $ZT = S^2\sigma T/k$, is more important

to characterize the performance of the thermoelectric generator, where S is the Seebeck coefficient or thermopower, σ is electrical conductivity, κ is thermal conductivity, and T is the absolute temperature [17].

1.2.3 Electromagnetic Effect

The electromagnetic effect is a well-known Faraday's law of electromagnetic induction, which can be traced back to 1831 [4]. It reveals that the voltage induced in a closed loop is proportional to the change rate of the magnetic flux through the annular region. This is the operational principle of a traditional magnetic generator, which is the cornerstone of modern society. The *ECE* of the magnetic generator can reach 90%, which has a desirable output power. But when we attempt to use it for wearable applications, this traditional magnetic generator is obviously unsuitable because of its heavy weight and large size. Thus, electromagnetic microgenerators were developed by adopting the microfabrication technology, which makes the device miniaturized and partially realizes its flexibility by fabricating flexible coils [18–20]. However, the properties of a hard magnet make it impossible to create fully flexible electromagnetic microgenerators.

Another promising alternative is wireless power transfer (*WPT*) based on electromagnetic induction, which can be used to transfer electrical power among multiple points without requiring a physical connection [21, 22]. The method endows the powered electronics with the maximum freedom, and has been proved as a wireless power source in both laboratory and industry. Although relay coils have been developed to cope with these obstacles, the limitations of power transmission direction and short-range distance still pose challenges for further applications [23].

1.2.4 Piezoelectric Effect

The piezoelectric generator is an important approach to scavenging biomechanical energy, which has been proved to be a clean energy source [24–26]. The fundamental mechanism is a piezoelectric effect, which, as an electric potential, is established at the end of piezoelectric materials, and under external pressure is a reversible process [24].

Several kinds of materials, including specific crystals, ceramics, polymers, and biological matter, have been discovered to possess piezoelectric property. They can be simply classified into two main categories: inorganic and organic materials. The most well-known inorganic materials are piezoelectric crystals and ceramics, such as PZT (lead zirconate titanate), BaTiO₃ (barium titanate), ZnO (zinc oxide), quartz, etc. [4, 27].

The typical organic material is PVDF (polyvinylidene fluoride), which is flexible and suitable for integration with wearable electronics. In order to obtain piezoelectric property, it is necessary to implement a post process of polarization by applying an ultra-strong electric field, which requires specific equipment and manufacturing processes.

Piezoelectric coefficient, also named piezoelectric constant, is one of the essential parameters to quantify the piezoelectric property of materials, whose

variation ranges from tens to thousands. Therefore, the performance of a piezoelectric generator has a direct relation with the piezoelectric property of the selected material. The remarkable linear characteristic between input pressure stimulation and output electric signal makes piezoelectric generators suitable to play the part of self-powered transducers [5]. It deserves to be mentioned that piezoelectric nanogenerators (PENGs) based on ZnO nanowires have developed vigorously in the past decades [28].

1.3 Triboelectric Nanogenerators

The traditional techniques proposed for microenergy sources are still hindered more or less by the following limitations, such as low output performance, strict environmental requirement, and low conversion efficiency. In 2012, a novel ambient energy-harvesting technology was developed. Named TENG, it combines triboelectrification effect and electrostatic induction [4, 6].

The charge generated at the friction interface of two different materials (i.e. the triboelectric pair) is defined as the triboelectrification effect. Although the observation and description of the electrification effect can be traced back more than 3000 years ago, the question is how to accumulate charges, generate electricity, and minimize the size in an efficient way.

By setting two electrodes on the back surface of a triboelectric couple, electrification effect and electrostatic induction are combined for effective power conversion [4, 6]. In the past five years, TENGs have aroused widespread interest due to excellent properties of high-output performance, low cost, being maintenance-free, sustainability, and green power performance. In order to strengthen and extend the capabilities of TENGs, a variety of techniques have been developed, and the power density has reached tens of mW/cm^2 level [9]. Furthermore, the maximum power conversion efficiency was achieved at 85% [29].

Since the electrification effect exists between nearly all of the two different materials used, this technique has great tolerance with material selection, and plenty of polymers and organic materials can be selected to achieve flexible and even stretchable devices. Besides, micro-/nanopatterned surfaces are usually adopted to maximize the effective friction area and enhance the output performance, and TENGs serve to emphasize it [30–34].

1.3.1 Principle of Triboelectric Nanogenerators

The original prototype of TENG had a triboelectric pair made of two different materials and two electrodes placed at the back. The operation principle can be described using contact-separation-mode TENG, as shown in Figure. 1.3.

Firstly, the whole TENG is electrically neutral and the triboelectric pair materials are separate. Under an action of external force, the pair makes contact and generates friction, and then surface charges are generated at the friction interface. Because of the difference in capabilities of losing or capturing electrons during electrification, one material of the triboelectric pair loses electrons and shows positive potential, while another captures electrons and shows negative potential.

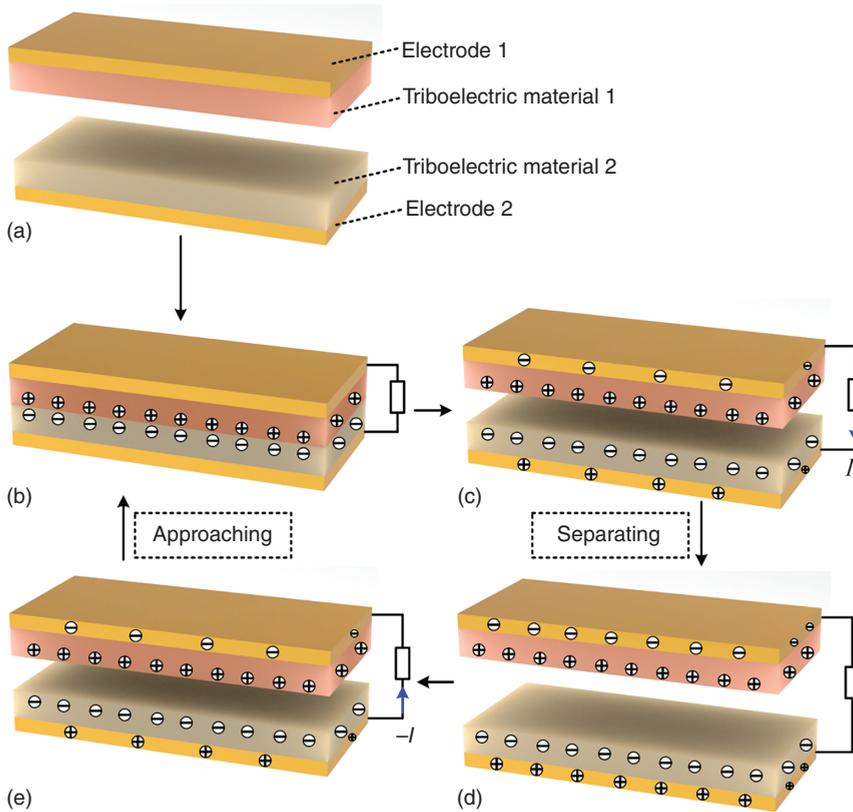


Figure 1.3 The working principle of contact-separation-mode triboelectric nanogenerators (TENGs). (a) In the beginning, the triboelectric pair made of triboelectric materials 1 and 2 are separated, and the whole device shows an electrically neutral state. (b) When a compressive force is applied, the top structures will move toward the bottom structures and have a friction with each other. Due to the phenomenon of triboelectrification effect, positive and negative charges of equal amount will be generated on the surfaces of triboelectric pair, respectively. (b–d) When the compressive force is removed, the triboelectric pair will separate from each other as a result of the mechanical recovery force. And then, an internal electric potential is established, which changes accordingly as the distance of the triboelectric pair increases and decreases. Consequently, due to the electrostatic induction, opposite charges are generated on the back electrodes, and a current (I) can be detected in the loop resulting from the charges flowing from one electrode to the other. When the triboelectric pair works cyclically, an electric output power will be generated continuously. Source: Reproduced with permission from Zhang et al. [4]. Copyright 2018, Elsevier.

In principle, the total charge amounts on the surface of the triboelectric pair are equal.

Secondly, after removing the external force, the triboelectric pair separates and an internal potential is established owing to the electrostatic induction. During the separation process, this internal potential will drive charges to flow from one electrode to the other through the connection loop to make the change of electrical potential balanced. And then, a positive current is formed, namely, electricity is generated. This is named the process of separating.

Thirdly, when applying an external force to the TENG again, the triboelectric pair moves toward each other, which will cause charges to flow back due to the opposite change of electrical potential. Thus, a negative current is formed, and this process is named approaching.

Finally, the triboelectric pair makes contact again and there is friction, and then a new cycle begins. Thus, when TENG repeats this separating–approaching cycle, it produces a periodical electric output with positive and negative parts.

The output performances of CS-mode TENGs were not so advantageous and it remained at a low level of several or tens of volts at the beginning. Subsequently, an arch-shaped geometry design was introduced, whereby surface roughness was increased. It significantly expanded the output voltages and power densities of CS-mode TENGs to hundreds of volts and several mW/cm^2 [9].

The fundamental mechanism for enhancing output by increasing surface roughness is to maximize the effect friction area, and more surface charges can therefore be generated [30]. As for the arch-shaped optimization, the basic rule of output enhancement can be roughly described by introducing a capacitance equivalent mode [35]. Assuming the surface charges (Q) are constant, the potential difference ($U = Q/C$) between two electrodes increases sharply when the capacitance (C) decreases sharply due to the stronger mechanical restoring force from the arch-shaped design. The CS-mode TENGs have the advantage of high output voltage, and their simple geometric structure makes for easy utilization, for instance, to convert most mechanical energies, such as pressing, impacting, bending, shaking, vibration, etc. However, using them for harvesting mechanically rotational energy is hard to achieve, and the frequency effect study also figured out that the CS-mode TENGs are not suitable for high-frequency applications [30].

1.3.2 Key Factor: Triboelectric Series

TENG development is based on the progress of materials. As described in Section 1.3.1, the key of electrification is the ability difference of losing or capturing electrons between the triboelectric pairs. Basically, if the ability relative difference of triboelectric pairs becomes larger, then the output performance of fabricated TENG becomes better as a result of the enhancement of generated surface charges. A table of triboelectric series, which qualitatively points out these ability differences, was established, and is summarized in Table 1.2.

The triboelectric series table is essential for constructing high-performance TENGs, since it quantitatively figures out the relative ability difference of losing/capturing electrons during the triboelectrification effect [36]. If the triboelectric pairs have a larger difference in this table, it means that it is easier for electrons to transfer from one to the other during the triboelectrification effect.

1.3.3 Material Progress of Triboelectric Nanogenerators

In the past few years, great progress was made in different triboelectric pairs for TENGs, as listed in Table 1.3. In this table, the materials in the left column, highlighted in yellow, indicate that they are easy to lose electrons and show positive

Table 1.2 Triboelectric series of different materials.

Triboelectric series						
	No.	Materials	No.	Materials	No.	Materials
POSITIVE 	1	Aniline-formol resin	17	Styrene-acrylonitrile copolymer	33	Polyacrylonitrile
	2	Polyformaldehyde 1.3–1.4	18	Styrene-butadiene copolymer	34	Acrylonitrile-vinyl chloride
	3	Ethylcellulose	19	Wood	35	Polybisphenol carbonate
	4	Polyamide 11	20	Hard rubber	36	Polychloroether
	5	Polyamide 6-6	21	Acetate, Rayon	37	Polyvinylidene chloride (Saran)
	6	Melanimeformol	22	Polymethyl methacrylate (Lucite)	38	Poly(2,6-dimethyl polyphenyleneoxide)
	7	Wool, knitted	23	Polyvinyl alcohol	39	Polystyrene
	8	Silk, woven	24	Polyester (Dacron) (PET)	40	Polyethylene
	9	Polyethylene glycol succinate	25	Polyisobutylene	41	Polypropylene
	10	Cellulose	26	Polyurethane flexible sponge	42	Polydiphenyl propane carbonate
	11	Cellulose acetate	27	Polyethylene terephthalate	43	Polyimide (Kapton)
	12	Polyethylene glycol adipate	28	Polyvinyl butyral	44	Polyethylene terephthalate
	13	Polydiallyl phthalate	29	Formo-phenolique, hardened	45	Polyvinyl chloride (PVC)
	14	Cellulose (regenerated) sponge	30	Polychlorobutadiene	46	Polytrifluorochloroethylene
	15	Cotton, woven	31	Butadiene-acrylonitrile copolymer	47	Polytetrafluoroethylene (Teflon)
	16	Polyurethane elastomer	32	Natural rubber		

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potential. The materials in the top row, highlighted in blue, indicate that they are relatively easy to capture electrons and show negative potential.

Currently, the widely used materials are PTFE (polytetrafluoroethylene), PDMS (polydimethylsiloxane), PI (polyimide), and FEP (fluorinated ethylene propylene), composed of 14, 11, 11, and 8 triboelectric pairs, respectively, as shown in Table 1.3. The main reason is that they occupy the bottom-tier levels in the triboelectric series, which leads to their outstanding abilities of capturing electrons during the electrification effect. The sequence of electron-capturing ability is listed as PTFE > PDMS > PI [7], and FEP is almost like PTFE with little difference in molecular structure.

Among known materials, PTFE, also known as Teflon, has the strongest ability to capture electrons, and possesses ultrastable chemical and physical properties.

Table 1.3 The configuration of triboelectric pairs (up to October 2017).

Triboelectric pairs	Material																							
	PTE	PDMS	Polyimide	FEP	Parylene	PVC	Rubber	PFA	Graphene	Epoxy	Polyolefin	PVDF	Polyester	Ground	PE	Polyamide	PU	PET	Alginatesodium	Human skin	Silicone	PP		
Metal	Al	2013.04	2012.11	2013.05	2013.09	2016.03	2014.03	2015.05	2013.01		2014.02	2014.06					2015.09						2017.03	
	Ag	2012.12	2013.08	2013.08	2017.04						2013.09					2015.03								
	Au	2013.04	2013.08	2014.11	2015.06																			
	Cu	2013.06	2013.08	2014.03	2015.06	2015.03		2015.03																
	Ni	2014.03	2016.08	2016.01	2015.01																			
Oxide	ITO	2016.05	2013.02																					
	ITO ₂	2013.04	2014.01																					
	Al ₂ O ₃								2015.01															
	SiO ₂	2014.05	2013.07	2014.04		2013.11																	2017.01	
Graphene oxide																								
Polymer	PET	2013.065	2012.05	2012.01			2016.06																	
	PMMA			2012.08																				
	Nylon	2013.04		2015.11																				
	PU	2015.05																						
	PPy	2015.12																						
	Urethane																							
	PVA																							
	EVA																							
	Biodegradable polymer																							
Others	Latex																							
	Carbon Black																							
	CNT	2014.04																						
	Cellulose																							
	Silk fibroin																							
	Paper	2017.01																						
	Human Liquid	2014.05	2013.01	2015.11	2014.04																			
Total pairs	14		11	3			3		2													1		

EVA: ethylene-vinyl acetate, PP: polypropylene, PPy: polypyrrole, PU: polyurethane, FEP: fluorinated ethylenepropylene, PFA: polyfluoroalkoxy, PVC: polyvinyl chloride, ITO: indium tin oxide, CNT: carbon nanotubes, PVDF: polyvinylidene fluoride, PDMS: poly(dimethylsiloxane), PMMA: poly(methyl methacrylate), PE: polyethylene, PET: polyethylene terephthalate, Biodegradable polymer: poly(L-lactide-co-glycolide) (PLGA), poly(3-hydroxybutyric acid-co-3-hydroxyvaleric acid) (PHB/V), poly(caprolactone) (PCL), poly(vinyl alcohol) (PVA). The date in the form represents the first time this triboelectric pair was reported in publications. The materials in the left and the right columns mean the positive and negative parts in each triboelectric parts in each triboelectric pair, respectively.

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PDMS is a kind of biocompatible material which can be micro-/nanopatterned easily using the molding cast process, and it also maintains a good stretchable capability. PI, also known as Kapton, can be processed into elastic thin films and made to work under high-temperature conditions.

Regarding the negative part of triboelectric pairs, materials can be classified into four groups: metals, oxides, polymers, and others. Metals can work as triboelectric pairs and electrodes at the same time [37–39], which simplifies the structure of TENGs. Oxide compounds are widely used, including ITO (indium tin oxide), TiO_2 , Al_2O_3 , SiO_2 , and graphene oxide. Among the rest, ITO exhibits a unique feature of conductivity and an outstanding property of transparency [40]. Polymers are the most important type of triboelectric materials; therefore, TENG is also called organic nanogenerator, which is firstly used to collect mechanical energy as organic materials [7].

One of the most attractive benefits of TENG is its tolerance to the material selection, since almost all of the two different kinds of materials are able to generate the electrification effect. Therefore, as is shown in Table 1.3, there are a great number of triboelectric pairs made up of different materials. However, the electrical performance of fabricated TENG depends on the ability difference of losing or capturing electrons. From this point of view, the materials are expected to possess the excellent electrical property of easily losing or capturing electrons. Exploring novel triboelectric materials still exists as an important research topic. In addition, triboelectric materials are expected to achieve excellent flexibility or even stretchability, environmental friendliness, or even biocompatibility, which fulfills the demand for constructing flexible and wearable self-powered microsystems.

1.3.4 Challenges of Triboelectric Nanogenerators

So far, the rapid development of TENG is still confronted with two challenges: one is the requirement to enhance power density, and the second is to simplify the structure to realize easier integration. Although simply enlarging the size can enhance the output power of TENG, the area power density remains constant, which is closely relevant to the fundamental mechanism of the triboelectrification effect, especially the triboelectric series that qualitatively depends on the ability of the triboelectric pair to lose or capture electrons [4]. Moreover, although four types of TENGs with different working principles have already been developed [4], such as contact-separation mode and relative-sliding mode, integrating TENG with other components to realize fully integrated self-powered microsystems still requires a lot of effort.

1.4 Summary

Through the abovementioned analyses and comparisons, we can summarize that TENGs possess much more attractive potentials and are considered as one of the promising ambient energy-harvesting approaches. In the latter part of this book, we summarize the emerging technology of TENG which serves as an important component of self-powered flexible and wearable microsystems, including

the innovation of working principle and material selection, the functionalization of sensing and actuating, and the future development direction of the all-in-one concept.

Abbreviations

CNT	carbon nanotubes
CS	contact-separation mode
ECE	energy conversion efficiency
EM	electromagnetic
EVA	ethylene-vinyl acetate
FEP	fluorinated ethylenepropylene
FS	free-standing mode
IoT	Internet of things
ITO	indium tin oxide
PCL	poly(caprolactone)
PDMS	poly(dimethylsiloxane)
PE	polyethylene
PET	polyethylene terephthalate
PFA	polyfluoroalkoxy
PHB/V	poly(3-hydroxybutyricacid-co-3-hydroxyvaleric acid)
PLGA	poly(L-lactide-co-glycolide)
PMMA	poly(methyl methacrylate)
PP	polypropylene
PPy	polypyrrole
PU	polyurethane
PV	photovoltaic
PVA	poly(vinyl alcohol)
PVC	polyvinyl chloride
PVDF	polyvinylidene fluoride
PZT	lead zirconate titanate
RS	relative-sliding mode
SE	single-electrode mode
TENG	triboelectric nanogenerator
ThM	thermoelectric
WPT	wireless power transfer

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