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Overview

1.1 Introduction

Nanogenerators are based on the use of the displacement current of Maxwell as the driving force to convert environmental energies into electric signals, exhibiting various potential applications in wearable electronics, sensor systems, robotics, and other energy-related science. Prof. Z. L. Wang and coworkers invented the first piezoelectric nanogenerator in 2006 [1], and invented the first triboelectric nanogenerator (TENG) in 2012 [2]. Both these nanogenerators are based on the polarization \mathbf{P}_s produced due to the mechanical motions induced electrostatic surface charges, where it is not the electric field produced medium polarization \mathbf{P} .

Usually, Maxwell's equations can be expressed by

$$\nabla \cdot \mathbf{D} = \rho \quad (1.1)$$

$$\nabla \cdot \mathbf{B} = 0 \quad (1.2)$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \quad (1.3)$$

$$\nabla \times \mathbf{H} = \mathbf{J} + \frac{\partial \mathbf{D}}{\partial t} \quad (1.4)$$

where \mathbf{D} is the electric displacement vector ($\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P}$). Z. L. Wang added an additional term \mathbf{P}_s in \mathbf{D} in 2017 [3, 4]. Thus, \mathbf{D} can be given as

$$\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P} + \mathbf{P}_s \quad (1.5)$$

where the polarization vector \mathbf{P} is associated with the appearance of the external electric field, while the additional term \mathbf{P}_s is associated with the appearance of the surface charges that can be independent of external electric fields [5]. Maxwell's displacement current is then expressed as

$$\mathbf{J}_D = \frac{\partial \mathbf{D}}{\partial t} = \epsilon \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial \mathbf{P}_s}{\partial t} \quad (1.6)$$

where the first term $\epsilon \frac{\partial \mathbf{E}}{\partial t}$ (electric field induced) is associated with the electromagnetic waves theory, while the $\frac{\partial \mathbf{P}_s}{\partial t}$ added by Wang (called **Wang term**) is due to the non-electric field-induced, strain-related polarization; this is the

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practical application of Maxwell's equations in the energies scavenging field as nanogenerators [5].

Various energies such as thermal, mechanical, chemical, and solar energies exist in the living environment. However, the occurrence of these energies depends on some working conditions such as weather or some other factors. The purpose of developing hybridized nanogenerators is to scavenge the different energies at the same time by integrating the different energy scavenging units into a system, so that we can obtain stable and sustainable power supply, regardless of whatever energy is available in the environment [6]. Prof. Z. L. Wang and coworkers invented the first hybrid energy cell in 2009 [7]. The first electromagnetic–triboelectric hybridized nanogenerator has been reported to scavenge one vibration energy by two different energy scavenging units in 2015 [8], which can largely enhance the efficiency of conversion from mechanical energy to electric energy. The existing hybridized nanogenerators are based on effectively stacking individual nanogenerators together in parallel or in series, where the individual nanogenerator has independent device structures and output electrodes. This is not suitable for miniaturization of the device dimension and for massive production. It is highly desirable to utilize multifunctional materials to obtain multi-effects coupled nanogenerators with the same structure, material, and electrodes. By using piezo–tribo–pyro–photoelectric effects, Prof. Ya Yang and coworkers invented the first coupled nanogenerators in 2015 [9], which have the same materials, the same electrodes, and simultaneous different energies scavenging abilities. This book will give a detailed summary about the design, performance, and applications of the hybridized and coupled nanogenerators.

1.2 Hybridized Nanogenerators

Hybridized nanogenerators are based on integrating the different energies scavenging units into a system for realizing simultaneous multiple energies scavenging, which has two advantages as compared with the reported individual energy scavenging techniques: (i) increasing the total output electric performances; (ii) providing a more stable and sustainable small power source. By integrating the electromagnetic generators (EMGs) and TENGs, the research group of Prof. Ya Yang reported various electromagnetic–triboelectric hybridized nanogenerators to scavenge different types of mechanical energies [8, 10–19], where all the hybridized nanogenerators have the same concept of integrating different nanogenerators to scavenge one mechanical energy for realizing obvious enhancement of energy conversion efficiency. Moreover, there have been more reports about hybridized nanogenerators in other countries such as Korea and Singapore in recent years [20–23].

1.2.1 Hybrid Energy Cells

In 2009, mechanical and solar energies have been reported to be scavenged with a hybrid device by using a piezoelectric nanogenerator at the bottom surface and

a dye-sensitized solar energy harvester at the top surface of the device [7]. The output voltage of the solar energy harvester can be about 0.591 V and the output current density can be about $6.9 \mu\text{A}/\text{cm}^2$. When the piezoelectric nanogenerator is working, the increased output voltage can be about 0.6 V and the output current density still remains at a fixed value, where the enhancement of the voltage is due to the piezoelectric nanogenerator [7]. By integrating a piezoelectric nanogenerator, the used dye-sensitized solar energy harvester has been also utilized to create a hybrid energy cell for concurrently scavenging solar and mechanical energies [24]. The corresponding output voltage of the solar cell was found to increase from 0.415 to 0.433 V due to the piezoelectric nanogenerator.

From 2011 to 2014, various hybrid energy cells were reported to scavenge multi-types of energies at the same time for realizing the enhancement of the conversion efficiency of the related energy devices, exhibiting practical applications in sensor networks and electrochemical reactions. In 2013, Yang et al. reported a hybrid energy cell consisting of a TENG and a micropyramid Si solar cell, which can be utilized to scavenge mechanical and solar energies at the same time [25]. As displayed in Figure 1.1a, the working of the TENG is due to the periodical contact and separation between the indium tin oxide (ITO) film and the polydimethylsiloxane (PDMS) nanowires. The fabricated solar cell consists

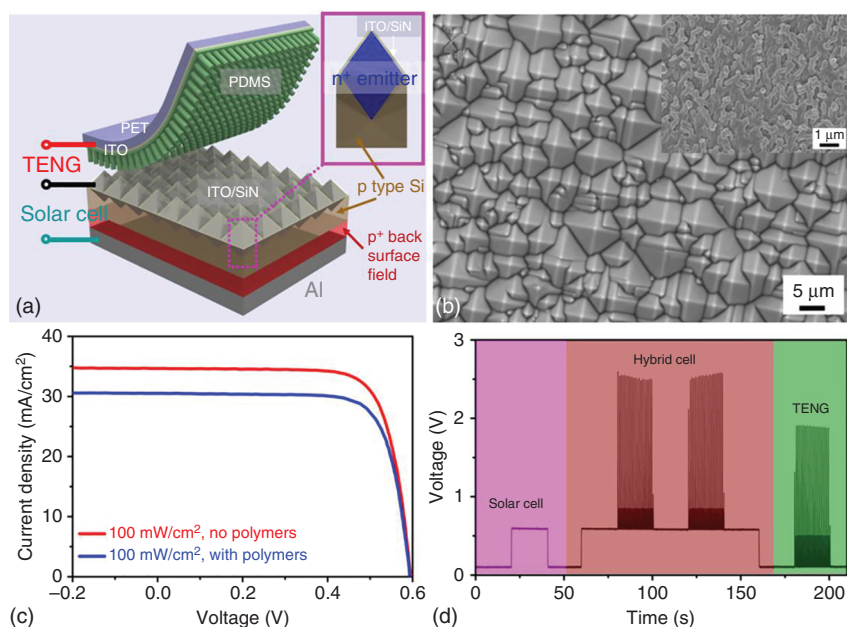


Figure 1.1 Hybrid energy cell for scavenging solar and mechanical energies. (a) Schematic diagram of the designed device. (b) Scanning electron microscopy (SEM) image of the Si pyramid structures. The inset displays an SEM image of the PDMS nanowire array. (c) J - V curves of the solar cell covered with and without PTFE (polytetrafluoroethylene)-PDMS polymers under a light illumination intensity of $100 \text{ mW}/\text{cm}^2$. (d) V - t curves of the solar cell, hybrid energy cell, and TENG. Source: Reproduced with permission from Yang et al. [25]. Copyright 2013, American Chemical Society.

of Si-based micropylramid p–n junctions, where the used Si pyramids have sizes ranging from 1 to 10 μm , as presented in Figure 1.1b. The inset of the Figure 1.1b shows the used PDMS nanowires. Figure 1.1c presents the J – V curves of the used solar energy harvester with and without the transparent PDMS nanowires on the surface of the device, where the output voltage of the device can be about 0.6 V and the output current density can be about 35 mA/cm^2 under simulated sunlight illumination, respectively. After using the PDMS nanowire array on the solar cell, the corresponding conversion efficiency of the fabricated device was decreased from 16% to 14%. Figure 1.1d nanomaterial displays the V – t curves of the hybrid energy cell, indicating that the rectified output voltage signals of TENGs exhibit DC peak characteristics.

Yang et al. also reported many other hybrid energy cells [26–29], including TENGs, piezoelectric nanogenerators, solar cells, thermoelectric nanogenerators, and pyroelectric nanogenerators. All the hybrid energy cells are based on effectively integrating the multimode energy scavenging units into a system to obtain sustainable power supply. The purpose of developing hybrid energy cells is to maximize the energies obtained from our living environment. How to effectively integrate the different energy scavenging units is still a challenge in practical devices. Moreover, the ratios among different energy scavenging abilities also need to be considered to make sure that this integration is useful in the system.

1.2.2 Electromagnetic–Triboelectric Hybridized Nanogenerators

The electromagnetic effect is due to the electromagnetic induction in Faraday's law, where the magnet and the coil have relative movements to induce the voltage/current signals. The working of a TENG is based on the coupling effect between the triboelectrification effect and the electrostatic induction in the periodical mechanical motion process [2]. In 2015, Prof. Ya Yang and coworkers first developed a method to integrate an EMG and a TENG in one device [8], so that the same mechanical motions can generate more electric energy due to the use of two energy scavenging devices. The corresponding energy conversion efficiency from mechanical energy to electric energy can be largely increased. This method has been rapidly extended to scavenge many kinds of other mechanical energies, such as wind energy [30], biomechanical energy [31], rotational energy [11], and so on.

To harvest vibration energy, Wu et al. demonstrated a spring-based hybridized nanogenerator including an EMG and a TENG [8], as illustrated in Figure 1.2a. In the vibration process, the electromagnetic and triboelectric effects can coexist in the same mechanical motions, which can effectively enhance the energy conversion efficiency. Figure 1.2b displays a photograph of the device, where the TENG can work under the periodical mechanical motions. Moreover, the working of an EMG is based on the relative mechanical motions between the magnet and the coils. Figure 1.2c illustrates the photograph of the vibration part. The pyramid microstructures with average sizes smaller than 10 μm can be found on the

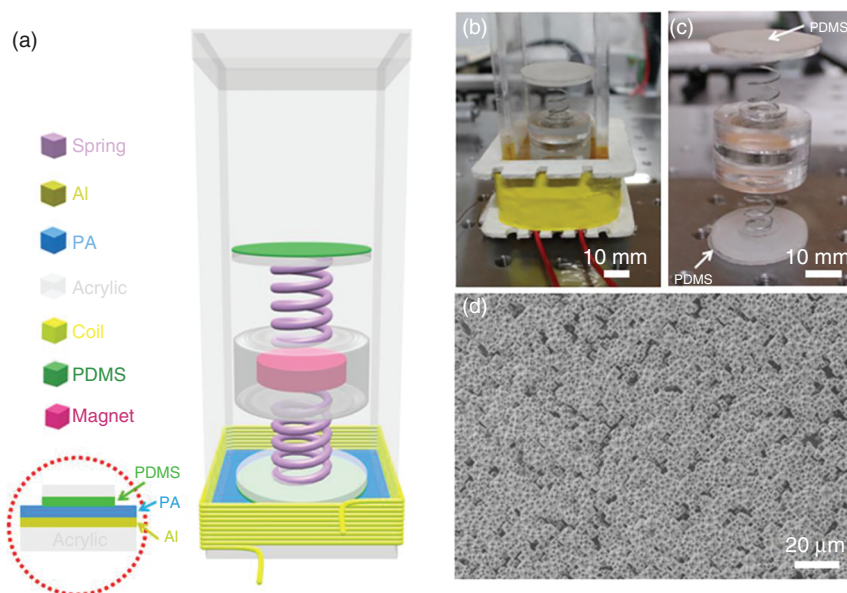


Figure 1.2 Electromagnetic–triboelectric hybridized nanogenerator for scavenging vibration energy. (a) Schematic diagram of the electromagnetic–triboelectric hybridized nanogenerator. (b) Photograph of the fabricated device. (c) Photograph of vibration part in the device. (d) SEM of the PDMS surface with micro/nanostructures. Source: Reproduced with permission from Wu et al. [8]. Copyright 2015, Elsevier.

surfaces of the PDMS films, as shown in Figure 1.2d. The output voltage of the TENG can be up to 600 V, and the corresponding output current can be about 3.5 μA . Under the same mechanical motions, the output voltage of the EMG can be about 3 V and the output current can be about 1 mA.

To scavenge wind energy, Wang et al. designed an electromagnetic–triboelectric hybridized nanogenerator, which is based on a tube structure [12]. As displayed in Figure 1.3a, the Kapton film can vibrate in the tube when wind goes through the tube from the right side of the device to the left side of the device [32]. By utilizing a high-speed camera, we can find that the effective contact between the two electrodes and the Kapton film for the TENG is mainly at the left side of the acrylic tube (marked with red dot line). There is no contact between the Kapton film and the two electrodes at the right most region of the tube, where most of the vibration energy was wasted due to the required effective contact/separation for the working of the TENGs. By integrating an EMG at the right region of the device in Figure 1.3b, the wind-driven vibration of the organic film can drive the working of the EMG and generate more electricity [12]. To understand the ratios of the produced energies for the two generators, we can calculate the electricity generated in the time of 10 seconds, where the TENG can give 2.6 mJ while the EMG can give 3.2 mJ. The total energy for the electromagnetic–triboelectric hybridized nanogenerator is 5.8 mJ, which

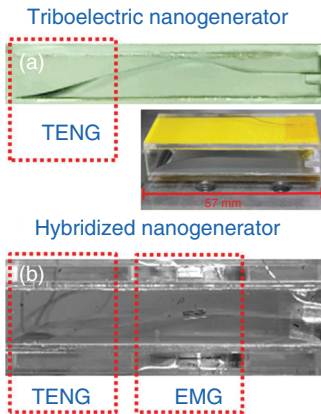


Figure 1.3 Electromagnetic–triboelectric hybridized nanogenerator for scavenging wind energy. (a) Photographs of the triboelectric nanogenerator (TENG) for harvesting wind energy. Source: Reproduced with permission from Wang et al. [32]. Copyright 2015, John Wiley and Sons. (b) Photograph of the hybridized nanogenerator including a TENG and an electromagnetic generator (EMG) for harvesting wind energy. Source: Reproduced with permission from Wang et al. [12]. Copyright 2015, American Chemical Society.

is larger than that of the individual TENG by over 130%. Moreover, Wang and Yang reported a hybridized nanogenerator, where the two ends of the Kapton vibration film have been fixed to harvest wind energy [19]. Although many kinds of electromagnetic–triboelectric hybridized nanogenerators have been reported, all these devices are based on the same idea of integrating the two kinds of energy scavenging units into one system to scavenge the same energies for enhancing the energy conversion efficiency.

1.2.3 Other Hybridized Nanogenerators

Smart cities can be realized by integrating wind and solar energies scavenging units with city buildings to power wireless sensor networks. However, it is very difficult to integrate conventional wind harvesters with Si-based solar cells, as illustrated in Figure 1.4a. The largest issue is the required rotation motion for the conventional wind harvesters, which is not suitable for integrating it with planar solar cells, as displayed in Figure 1.4b. In 2016, Prof. Ya Yang first designed a new hybridized nanogenerator by integrating silicon solar cells with a wind-driven TENG in one device for harvesting wind–solar energies at the same time [30]. As illustrated in Figure 1.4c, these hybridized nanogenerators can be effectively integrated on the top surface of houses because of the non-rotating part of the TENGs. Figure 1.4d presents an optical image of the hybridized nanogenerator, where the solar cells can be fixed on the surfaces of the wind-driven TENG. Under a device size of 12×2.2 cm, the total output power of the solar cells is 8 mW, but the corresponding output power for the wind-driven TENG can be about 26 mW. If the hybridized nanogenerator array can be arranged on the top surfaces of city buildings to simultaneously scavenge wind and solar energies, self-powered sensor networks in smart cities can be realized in future.

For this technique, the solar cells can be easily attached on the wind-driven TENGs. The most important issue is how to realize the large size of the device. To solve this issue, Prof. Ya Yang first designed the meter-scale TENG to harvest wind energy and applied the corresponding patents, as shown in Figure 1.5a, where corresponding output current signals of single device can be larger than $100 \mu\text{A}$, as depicted in Figure 1.5b. Figure 1.5c illustrates that the corresponding

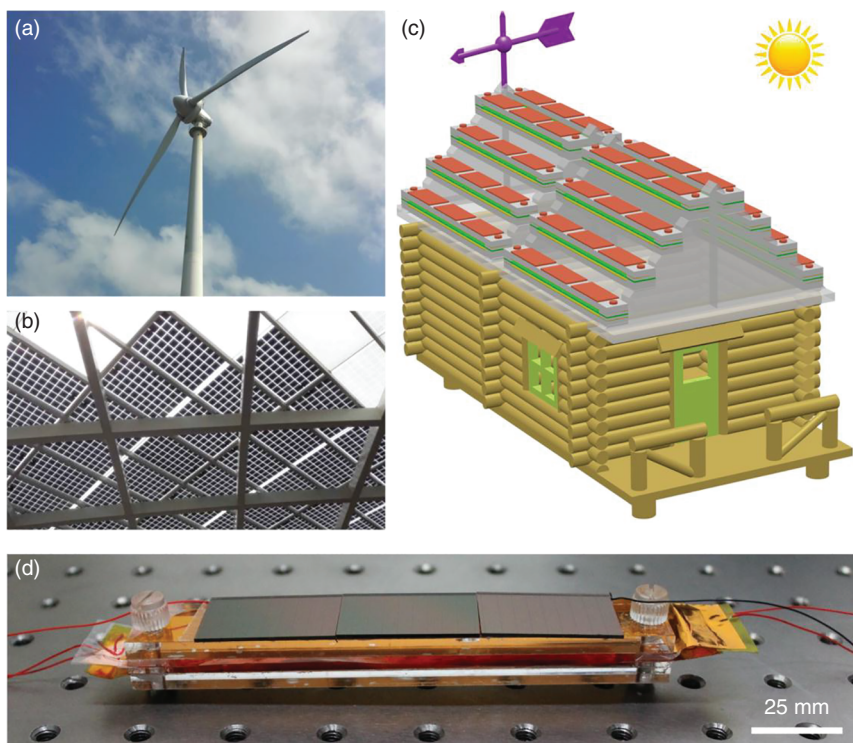


Figure 1.4 Hybridized nanogenerator for scavenging solar and wind energies. (a) Photograph of conventional wind turbine generator. (b) Photograph of the solar cells on the top of a building. (c) Schematic diagram of the hybridized nanogenerators on the roof of a house model. (d) Photograph of the fabricated hybridized nanogenerator. Source: Reproduced with permission from Wang et al. [30]. Copyright 2016, American Chemical Society.

output power of the fabricated TENG is larger than 220 mW for the single device. Figure 1.5d displays that TENGs with different sizes can be fabricated, which is very important for meeting different environmental needs. This technique has demonstrated the ability of designing different devices with sizes ranging from centimeters to hundreds of meters to simultaneously scavenge wind and solar energies, as illustrated in Figure 1.5e [33].

Figure 1.5d illustrates a schematic diagram of a power station including thousands of large-scale TENGs. Before 15 March 2018, Prof. Ya Yang first designed the large-scale “electric wall” including tens of TENGs as a wall for scavenging wind energy and applied the corresponding patents, as displayed in Figure 1.6a. Each wind wall can provide the electric power outputs ranged from 1 W to 10 W, which can easily light up tens of lamp bulbs, as presented in Figure 1.6b. One thousand of these wind walls can provide a total electric power output up to about 10 kW, which can be utilized in many windy places such as both sides of high-speed train road, desert island, sea surface, mountain area. If the solar cells were fixed on the devices, the energy produced can be much larger than 10 kW. Several megawatt or gigawatt power stations can also

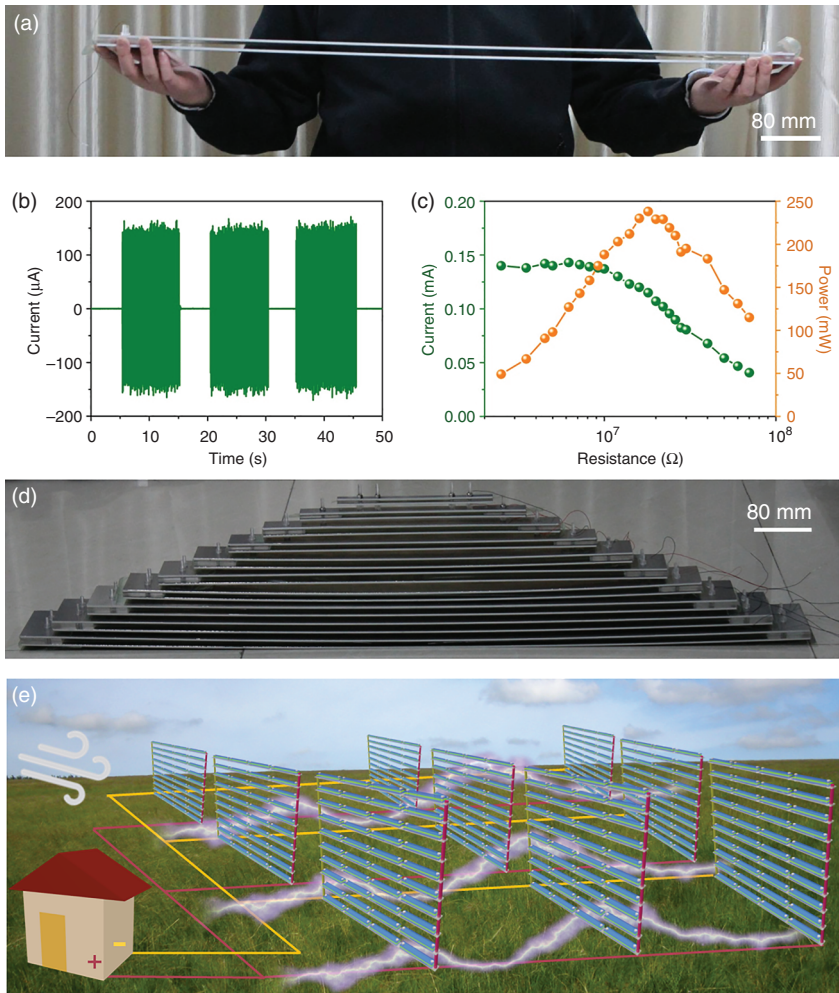


Figure 1.5 Large-scale triboelectric nanogenerators (TENGs) for scavenging wind energy. The large-scale TENG to scavenge wind energy was first designed and fabricated by Prof. Ya Yang before 15 March 2018. The corresponding patents have been also filed based on it. (a) Photograph of the designed large-scale TENG. The data were provided by Prof. Ya Yang. (b) The output current signals of the large-scale TENG. The data were provided by Prof. Ya Yang. (c) The output current signals of the large-scale TENG under different loading resistances and the corresponding output powers. The data were provided by Prof. Ya Yang. (d) Photograph of the designed different-sized TENGs. The data were provided by Prof. Ya Yang. (e) An illustrative diagram of large-scale TENGs wind walls on farm land. Source: Reproduced with permission from Chen et al. [33] Copyright 2017, John Wiley and Sons.

be designed based on this new technique. The electric power obtained can be transformed by power substations and then be transported through cables for long-distance electricity supplement. These large-scale new hybridized nanogenerators have potential applications in the wind and solar energies scavenging field.



Figure 1.6 Large-scale triboelectric nanogenerators (TENGs) for scavenging wind energy. The mobile meter-scale wind wall includes tens, even hundreds, of meter-scale TENGs, which was first designed and fabricated by Prof. Ya Yang before 15 March 2018. The corresponding patents have been also filed based on it. (a) Photograph of the large-scale TENGs together with Prof. Ya Yang. The time of taking the photo is 15 March 2018. The data were provided by Prof. Ya Yang. (b) Photograph of 16 LED bulbs lighted by the TENGs to scavenge wind energy. The data were provided by Prof. Ya Yang. Source: Courtesy of Prof. Ya Yang.

1.3 Coupled Nanogenerators

Hybridized nanogenerators can simultaneously scavenge the different energies from our living environment by integrating the different nanogenerators into a system. Different nanogenerators have different electrodes and materials, which makes it difficult to realize device miniaturization and decrease the device cost. To solve this issue, in 2015, Prof. Ya Yang and coworkers first invented the coupled nanogenerators, which have the same materials and electrodes but

different energy scavenging functionalities [9]. One-structure-based coupled nanogenerators can be fabricated by using the various physical properties to simultaneously/individually harvest various energies in the environment wherever/whenever energy types are available, where the core of the coupled nanogenerators is the multifunctional materials.

1.3.1 Pyroelectric and Photovoltaic Coupled Nanogenerators

Some ferroelectric materials have both pyroelectric and photovoltaic properties such as LiNbO_3 [34–36], $\text{Pb}(\text{Zr,Ti})\text{O}_3$ (PZT) [37–39], BaTiO_3 (BTO) [40–42], and BiFeO_3 [43–45]. Owing to the wide bandgaps of about 2.7–4 eV, these ferroelectric materials are more suitable to be used as photodetectors instead of as solar cells. When the light is illuminated on the ferroelectric materials, the light-induced heating can generate pyroelectric current, and photovoltaic current can be also observed. The pyroelectric and photovoltaic coupled nanogenerators are based on understanding the mechanisms about how to couple the two current/voltage signals in the ferroelectric materials.

Here, we used the ferroelectric BaTiO_3 material as an example to describe the pyroelectric and photovoltaic coupled nanogenerators [46]. The Ag and ITO were utilized as the bottom and top electrodes of the device, respectively. As illustrated in Figure 1.7a, there will be no current signals when the $dT/dt = 0$, where T is the temperature and t is the time. The reason is that there is no change of the polarization in the BaTiO_3 material, so that no output current signals can be seen in the external circuit. When the temperature variation dT/dt is larger than zero, the corresponding polarization in the BaTiO_3 material will be decreased, indicating that the electrons in the external circuit can move from the bottom Ag electrode to the top ITO electrode to deliver a pulsed output current signal. The pyroelectric current signals are AC signals, which can only flow in the external circuits.

For the photovoltaic effect, the spontaneous polarization-induced electric field can effectively drive the light-induced electrons and holes to separate and flow to the two electrodes, as displayed in Figure 1.7b. Owing to the built-in electric field in these ferroelectric materials, only one material can realize the appearance of photovoltaic current instead of p–n junctions. As depicted in Figure 1.7c, the pyroelectric effect induced by light illumination can modulate the charge transfer processes of the light-induced electrons and holes such as generation, separation, and recombination. As a result, the coupling enhancement “ $1 + 1 > 2$ ” for the output current signals can be seen in this system due to the coupling effect between the pyroelectric current and the photovoltaic current signals. The new physical effect of ferro–pyro–phototronic effect can be found in this process.

1.3.2 Multi-effects Coupled Nanogenerators

Multi-effects coupled nanogenerators are focused on multifunctional materials such as ferroelectric materials. As illustrated in Figure 1.8, Prof. Ya Yang and coworkers invented the coupled nanogenerators in 2015 [9], representing a new

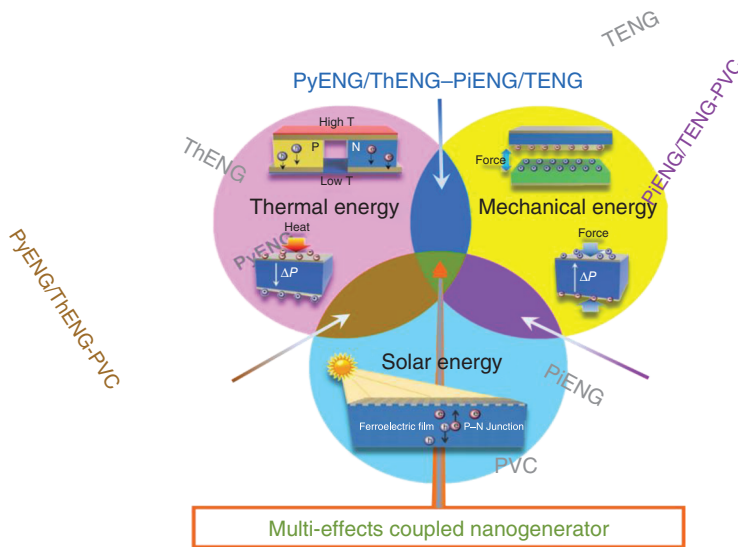


Figure 1.8 Illustration of multi-effects coupled nanogenerator toward multi-energies scavenging. Source: Reproduced with permission from Zhang et al. [9]. Copyright 2017, John Wiley and Sons.

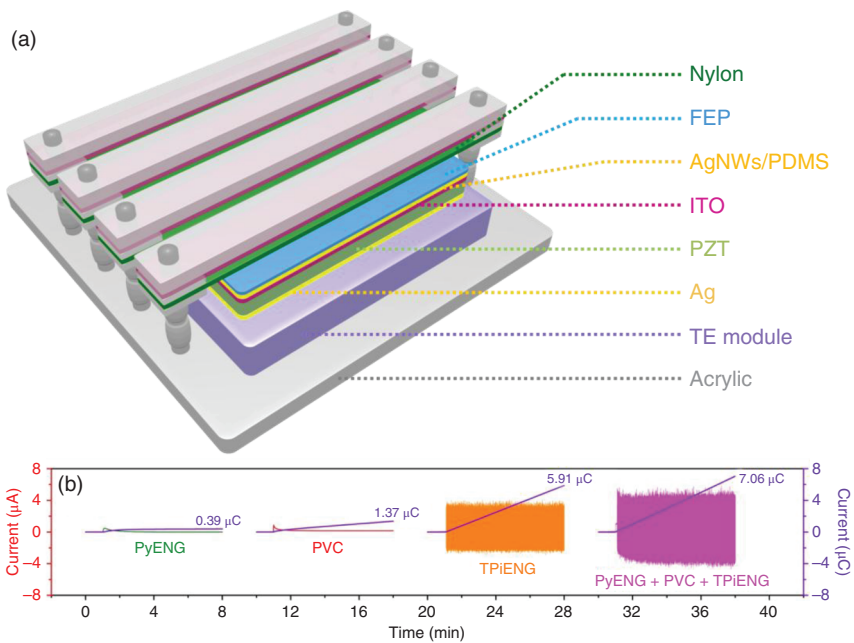


Figure 1.9 One-structure-based multi-effects coupled nanogenerator. (a) Schematic diagram of the designed one-structure-based multi-effects coupled nanogenerator. (b) Transferred charges of the different devices. Source: Reproduced with permission from Zhang et al. [9]. Copyright 2017, John Wiley and Sons.

different energies are applied on the device, the produced charge amount can be up to $7.06 \mu\text{C}$, which is much larger than that of individual energy scavenging methods.

The ferroelectric PZT material is toxic due to the presence of Pb. It is necessary to look for other high-performance ferroelectric materials without Pb for the coupled nanogenerators. It has been reported that a polyvinylidene fluoride (PVDF)-based one-structure-based coupled nanogenerator has been utilized to scavenge mechanical and thermal energies at the same time by using tribo–piezo–pyroelectric effects [47]. By charging a capacitor, the coupled nanogenerators have the best charging performance. However, the piezoelectric constant of the PVDF is too small with about 20 pC/N . Ji et al. reported a ferroelectric BTO material-based multi-effects coupled nanogenerator [48], where the piezoelectric constant of BTO can be larger than 300 pC/N . On comparing the charging curves of the different conditions, it is seen that the coupled nanogenerator exhibited faster charging performance than the other individual effects.

As compared with the other hybridized nanogenerators with simple physical integrations, the one-structure-based multi-effects coupled nanogenerators have more advantages such as simpler structure, smaller volume, and lower cost, representing a new research trend in multifunctional materials-based all-in-one multiple energy harvesting in our living environment. Moreover, these coupled nanogenerators have potential applications in multifunctional sensor systems. Owing to the multifunctionalities achieved by one material, these new sensor systems exhibit more advantages than the conventional methods such as higher resolution, smaller device size, and so on. Obtaining high-performance multifunctional material is still a challenge, and more research work is needed along this research direction.

1.4 Applications

Hybridized nanogenerators have extensive practical applications in environmental energies scavenging technologies and self-powered sensor networks. Wind and solar energies can be simultaneously scavenged by the hybridized nanogenerators, consisting of TENGs and Si-based solar cells. Moreover, the sizes of the hybridized nanogenerators can range from centimeters to hundreds of meters for harvesting large-scale energies, pushing the potential applications of the hybridized nanogenerators instead of the conventional wind–solar energy harvesters in some environments. Moreover, the energy conversion efficiency is still a challenge for these energy scavenging devices. The stability of these hybridized nanogenerators in the natural environment still needs to be considered. Hybridized nanogenerators have been used as various self-powered sensors, where the different nanogenerators can detect the different physical signals to realize simultaneous detection. Because different nanogenerators were used for detecting the different physical signals, no signal interference can be found in the detection process.

The coupled nanogenerators have potential applications in multi-energies scavenging and multifunctional sensor systems. The core of the coupled nanogenerators is the multifunctional material; therefore, looking for a high-performance material is one of the most important challenges for the applications. Ferroelectric materials-based coupled nanogenerators have demonstrated the ability of scavenging multi-energies from the environment, indicating that the coupled nanogenerators have better performances than individual energy scavenging devices. However, energy conversion efficiency of these devices still needs to be enhanced. Ways to obtain more electricity from these devices also need to be considered. For the application of multifunctional sensor systems, coupled nanogenerators exhibit more advantages than the conventional methods of integrating the different sensors in a system, such as smaller sizes, higher resolutions, and lower cost. The coupling effect among the different physical signals is very interesting due to the possible new mechanism. However, how to change the current/voltage in one circuit by the coupling needs to be investigated to quantify these signals in the multifunctional sensing process.

The hybridized nanogenerators have more chances to be used as a practical large-scale energy scavenging technique as compared with the coupled nanogenerators. However, the coupled nanogenerators have more scientific significance than the hybridized nanogenerators, where the coupling enhancement among the different physical signals is interesting for future research.

1.5 Conclusion and Prospects

Hybridized nanogenerators integrating two or more different energy harvesting units into an energy scavenging system for simultaneously harvesting multi-types of energies in our living environment have the potential for effectively enhancing the total output performance. It is one of the most significant multiple energies scavenging technologies. The development of hybridized nanogenerators is based on maximizing the harvested energy from one type of energy by integrating the different energy scavenging units and achieving complementary harvesting of multiple energies. The large-scale hybridized nanogenerator consisting of solar cells and TENGs will be a very powerful technology such as creating new wind-solar complementary energies scavenging systems.

The core of the coupled nanogenerators is based on multifunctional materials such as ferroelectric materials. The coupled nanogenerators have the same materials and the same electrodes, which can obtain various advantages of smaller sizes, lower cost, and higher integrations than the conventional integrated different nanogenerators. The coupling enhancement effect among the different physical effects has been found in the coupled nanogenerators. Considering the highlighted advantages and the ongoing related research hotspots, more breakthroughs in hybridized and coupled nanogenerators with respect to the design, performance, and applications for multiple energies scavenging and self-powered multifunctional sensor networks will be seen in future.

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