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In the past few years, triboelectric nanogenerator-based (TENG-based) hybrid generators and systems have experienced a widespread and flourishing development, ranging among almost every aspect of our lives, e.g., from industry to consumer, outdoor to indoor, and wearable to implantable applications. Although TENG technology has been extensively investigated for mechanical energy harvesting, most developed TENGs still have limitations of small output current, unstable power generation, and low energy utilization rate of multisource energies. To harvest the ubiquitous/coexisted energy forms including mechanical, thermal, and solar energy simultaneously, a promising direction is to integrate TENG with other transducing mechanisms, e.g., electromagnetic generator, piezoelectric nanogenerator, pyroelectric nanogenerator, thermoelectric generator, and solar cell, forming the hybrid generator for synergetic single-source and multisource energy harvesting. The resultant TENG-based hybrid generators utilizing integrated transducing mechanisms are able to compensate for the shortcomings of each mechanism and overcome the above limitations, toward achieving a maximum, reliable, and stable output generation. Hence, in this review, we systematically introduce the key technologies of the TENG-based hybrid generators and hybridized systems, in the aspects of operation principles, structure designs, optimization strategies, power management, and system integration. The recent progress of TENG-based hybrid generators and hybridized systems for the outdoor, indoor, wearable, and implantable applications is also provided. Lastly, we discuss our perspectives on the future development trend of hybrid generators and hybridized systems in environmental monitoring, human activity sensation, human-machine interaction, smart home, healthcare, wearables, implants, robotics, Internet of things (IoT), and many other fields.

1. Introduction

With the gradual rollout of the 5G (5th-generation mobile networks) technology across the world, the role of the Internet of things (IoT) is becoming more and more essential in both industrial and commercial developments [1]. Entering the IoT era, wireless and portable electronics are undergoing explosive advancement, with the total number increasing tremendously and power consumption decreasing significantly. Affected by the nature of incredibly huge numbers, small power consumption, and ultrawidely distributed location of IoT devices, energy supply in the new era should be changed from the centralized, immobile, ordered, and large-scale mode to the distributed, mobile, in situ, and small-scale mode. Yet batteries as the primary choice of conventional energy supply exhibit apparent drawbacks such as the limited lifespan that needs frequent replacement or recharging, large volume and weight, rigidity, biological incompatibility, and environmental pollution. Thus, to meet the above requirements, the ideal energy supply units should possess the characteristics of portability, sustainability, miniaturization, wearability, and implantability depending on the applications [2–7]. In this regard, scavenging energy from the ambient surroundings by energy harvesters can provide a green, portable, and sustainable solution. There exist various types of energy forms that are normally wasted in the environment, including mechanical energy associated with diverse nature vibrations and human activities, thermal energy, and solar energy. Accordingly, different types of energy transducing mechanisms and generators (i.e., energy harvesters) have been developed to scavenge these wasted energies, such as the electromagnetic generator (EMG), piezoelectric nanogenerator (PENG), and triboelectric nanogenerator (TENG) for mechanical energy, the thermoelectric generator (TEG) and pyroelectric nanogenerator (PyENG) for thermal energy, and the solar cell (SC) for sunlight energy. In most scenarios, multiple energy forms coexist in the ambient environment, and the strength of a single-type energy form may vary significantly from time to time, such as solar energy varying with time and weather, random wind/wave energy, and different human activities. Therefore, generators employing a single-energy transducing mechanism may greatly suffer from the unstable energy source, low energy utilization rate, low conversion efficiency, and low adaptability in different scenarios. On the other hand, hybrid generators utilizing integrated transducing mechanisms can be more effective in harvesting both single-type and multiple energies. Through synergetic designs, hybrid generators can compensate for the shortcomings of each mechanism, improve the space utilization efficiency, enhance the energy utilization rate, and thus can be applied in diverse scenarios as the energy supply units [8–10].

Among different energy forms, mechanical energy is one of the most ubiquitous energy sources, widely existing in water wave, wind, sound/ultrasound, machinery vibration, human motions, etc. Considering that most IoT devices are adopted in the environment- or human-related applications where they exhibit abundant mechanical energy, generators with mechanical energy harvesting ability will be most desirable. Since the first invention in 2012 by Prof. Wang and his team [11], the TENG technology has been extensively explored for mechanical energy harvesting, due to its superior advantages of high output performance, versatile operation modes, broad material availability, wearable/implantable compatibility, simple fabrication, high scalability, and low cost [12–15]. Benefitting from these merits, integrating TENG with other transducing mechanisms yields a promising research direction for developing hybrid generators, which has received flourishing development in the past few years [16–19]. Furthermore, after integrating the TENG-based hybrid generators with power management circuitry, energy storage units, and functional components, a variety of hybridized systems with self-sustainability can be achieved for broad applications. Here, in this review, we systematically introduce the key technologies and the recent progress in the TENG-based hybrid generators and hybridized systems: (1) principles of different types of transducing mechanisms and generators; (2) strategies for enhancing the output performance of TENGs; (3) applications of TENG-based hybrid generators in the outdoor, indoor, and on-human-body scenarios; (4) strategies for achieving efficient power management and energy storage; and (5) demonstrations of functional and self-sustainable hybridized systems for various applications. These sections summarize the development trend of the TENG-based hybrid generators and hybridized systems in the aspects of operation principles, structure designs, optimization strategies, power management, system integration, and applications. In the end, conclusions and perspectives on the existing challenges and future trends are also provided, which give a glimpse of the further development of hybrid generators and hybridized systems in the IoT era.

2. Transducing Mechanisms and Generator Principles

To scavenge different types of ambient available energies, generators based on different transducing mechanisms have been developed. As shown in Figure 1, TENG/EMG/PENG, PyENG/TEG, and SC are the most common generators employed for mechanical, thermal, and solar/light energy harvesting, respectively. Through synergetic integration, their hybrid generators can be further employed for complementary and effective energy harvesting of both single-type and multiple energies.

2.1. TENG

The first TENG was invented in 2012, based on the coupling effect of contact electrification (triboelectrification) and electrostatic induction, which can convert mechanical energy into electricity [11]. The origin of TENG and other types of nanogenerators (e.g., PENG and PyENG) is Maxwell's displacement current theory, i.e., external current induced by a time-varying electric field in the nanogenerators [20]. More specifically, when two dissimilar materials contact with each other, due to their different electron affinity, surface charges are generated at the contact interface. Then, upon separation, the built-up electric potential will drive electrons on the respective electrodes to flow in the external circuit until a new balance is achieved. If the two materials are brought into contact again, the electric potential will disappear and electrons will flow in a reverse direction. Thus, under periodic contact and separation, alternating current (AC) can be generated on the external load, and a rectification circuit is normally required for energy storage to convert the AC output into the direct current (DC) output [21–23]. There are also some advanced designs of DC-TENGs that do not need a rectification circuit before energy storage [24–28], but they exhibit more complicated structures and most developed TENGs are still AC-based. According to a material's ability to lose or accept electrons (electron affinity), different materials can be arranged into a sequence called the triboelectric series, from the most positive material to the most negative material [29, 30]. Along these years, different models including the electron-cloud-potential-well model have been developed to explain the origin of contact electrification among polymers, metals, semiconductors, and even liquids [31–34]. It is revealed that in most cases, electron transfer is the dominating effect of the contact electrification process. Specifically, for the solid-liquid contact electrification, a two-step formation of the electric double layer (EDL) at the solid-liquid interface is proposed by Prof. Wang, which is also known as the Wang model [34, 35]. The Wang model indicates that the electron transfer is required at the very first contact to create the first layer of electrostatic charges on the solid surface, and then, the ion transfer in solution dominates in the second step due to the electrostatic interactions with the charged solid surface. The surface charge density, as one of the most important parameters determining the output performance of TENGs, can be enhanced through several strategies, e.g., proper material selection (contact materials with a larger difference in electron affinity), surface modification (like micro/nanostructures, chemical treatment, and ion injection), structural optimization, middle layer insertion, and circuitry assistance [36–38]. With the generated surface charges, the output voltage on an external load can be given by [39] where is the open-circuit voltage that is greatly determined by the surface charge density, is the transferred charges across the two electrodes, and is the instant capacitance of the TENG at a separation distance of . In general, there are four basic operation modes of TENG, i.e., vertical contact-separation mode, lateral sliding mode, single-electric mode, and standing triboelectric mode [40]. All these four modes share the same output equation given by Equation (1). Based on these operation modes, a large variety of TENGs have been developed for both the mechanical energy harvesting and self-powered ambient parameter monitoring/intervention [41, 42], such as micro/nanopower sources [43], blue energy [44], physical/chemical sensors [45, 46], human machine interfaces [47, 48], nerve/muscle/brain stimulators [49, 50], air filters [51], droplet manipulation [52], and high-voltage applications [53, 54]. Hence, TENGs can contribute to be not only power sources but also various functional components in self-sustainable hybridized systems.

2.2. EMG

The mechanism of EMG is based on Faraday's law of electromagnetic induction, in which the voltage on a closed loop is proportionally induced by the loop's magnetic flux variation over time (, where is the magnetic flux and is the time). Due to the high energy conversion efficiency, EMGs have been widely adopted in modern energy farms for centralized and large-scale electricity generation. Besides, EMGs have also been developed as distributed and small-scale energy sources for harvesting the in situ energy from various machinery vibrations and human motions [55–57]. When there are relative movements between the magnets and coils in the EMGs, induction current will be generated in the coil. According to the difference in relative movements, EMGs can be classified into two basic operation modes, i.e., movable magnet-fixed coil mode and movable coil-fixed magnet mode. Compared to TENG, EMG normally exhibits small impedance with large current but small voltage outputs. Due to these distinct output characteristics and the similar triggering forms of EMG and TENG (both by the relative movements of different components), EMG can be a good complement to TENG in a hybrid generator and hybridized system.

2.3. PENG

The fundamental mechanism of PENG is the appearance of an electric potential (electric dipole moment) on a piezoelectric material when it undergoes external pressure, which is also known as the direct piezoelectric effect [58–60]. The common piezoelectric materials can be categorized into two classifications—inorganic piezoelectric materials and organic piezoelectric materials [61]. The popular inorganic materials include piezoelectric ceramics and crystals, such as lead zirconate titanate (PZT), aluminum nitride (AlN), barium titanate (BaTiO₃), lithium niobate (LiNbO₃), zinc oxide (ZnO), and quartz. Meanwhile, the representative organic piezoelectric materials are polyvinylidene fluoride (PVDF) fiber/thin film and its copolymers, with good flexibility and suitability for wearable electronics. One of the key parameters determining the output performance of piezoelectric materials is the piezoelectric coefficient, or , which is the generated charge density normalized by the applied stress (unit: C·N⁻¹). The "3" here denotes the polar axis direction of the material, and "1" can be used to denote one of the directions that are perpendicular to the polar axis due to symmetry. Thus, depending on the direction of the applied stress, PENGs can be classified into two modes, 33-mode (stress along the polar axis) and 31-mode (stress perpendicular to the polar axis). Benefitting from the advanced transferring technology in recent years, rigid piezoelectric materials with normally higher piezoelectric coefficient can also be transferred to flexible substrates, greatly improving the output performance of PENGs in wearable and implantable applications [62, 63].

2.4. PyENG

The operation mechanism of PyENG is based on the pyroelectric effect of a material, referring to the spontaneous polarization change under the temperature variation over time (, where is the temperature and is the time) [64, 65]. Generally, the spontaneous polarization intensity of a pyroelectric material will remain unchanged (no pyroelectric current) when there are no temperature variations over time, no matter how high/low the temperature is or with/without a temperature gradient over space. Once there is an ascent or descent in temperature over time, the spontaneous polarization intensity of a pyroelectric material will then change accordingly, generating a pyroelectric current in the external circuit until a new equilibrium is achieved. Since most piezoelectric materials also exhibit pyroelectric property, thus the same material can be used to harvest both mechanical and thermal energies under different usage scenarios, based on the direct piezoelectric effect [58–60].

2.5. TEG

If a temperature gradient exists in a thermoelectric material, an electric potential will be built up at the two ends of the material, according to the Seebeck effect [66]. The Seebeck effect refers to the buildup of an electric potential difference across a thermoelectric material (typically semiconductor or conductor), because of the existence of a temperature gradient and the resultant diffusion of charge carriers from the hot end to the cold end. The output performance of a thermoelectric material is closely relative to its figure of merit () and the temperature difference (). The figure of merit can be given by [67] where is the Seebeck coefficient of the material, is the electrical conductivity, is the absolute temperature, and is the thermal conductivity. Depending on the major charge carriers, thermoelectric materials can be divided into -type (electron charge carriers) and -type (hole charge carriers). Under the same temperature difference, -type materials and -type materials will have the built-up electric potential of opposite polarity. Thus, multiple couples of -type and -type thermoelectric materials can be connected in series to improve the overall output performance of a TEG.

2.6. SC

The underlying mechanism of SC is the photovoltaic effect, where the electron and hole pairs are generated after the absorption of lights. After the separation of the electron and hole pairs by the internal built-in electric field, an electric potential is created between the two electrodes [68]. The power conversion efficiency () is one of the key parameters to evaluate the performance of an SC, which is given by [9] where , , , and are the maximum output performance, the input solar energy, the open-circuit voltage, the short-circuit current density, and the fill factor, respectively. Nowadays, state-of-the-art SCs can reach an efficiency of more than 40% [69]. Other than the conventional rigid SCs, flexible SCs have also been developed for wearable and more diverse applications [70].

3. Output Enhancement Strategies for TENGs

For the TENG-based hybrid generators, the output performance of TENG components plays an important role in determining the overall output of the hybrid generators. Therefore, it is of great significance to enhance the performance of the integrated TENG components. The surface charge density of TENGs is inevitably limited by the air breakdown effect between two triboelectric surfaces and thereby is severely detrimental to their practical applications [70, 71]. As the earliest strategy to enhance performance, TENG devices are improved by material selection [72], structure optimization [73], surface modification [74], ion injection [75, 76], and environment control [77]. Charge improvement from material modifications is finite while a vacuum strategy limits applications of TENGs; therefore, more effective methods are desired to improve the charge density in air for broad applications of TENGs. There are still urgent needs for developing advanced mechanisms in enhancing the output, which should also be facile to be integrated into the practical devices for different working environments. Recently, there are a few effective methods proposed to obtain an optimum contact structure and improved output performance.

Li et al. proposed a standard method to precisely evaluate the contact status of TENG to optimize the contact of two tribo-surfaces, as shown in Figure 2(a) [78]. They illuminate the strategies of enhancing the charge output for the charge excitation TENG, including the reduction of the thickness of dielectrics, the increment of external capacitor, and the control of the atmospheric environment. With the quantified contact, an arched composite soft structure is designed by using a homemade carbon/silicone gel electrode, which can enhance the contact efficiency of devices from 6.16% to 54.98% for a 4 μm dielectric film. As a result, the average charge and the energy density for the TENG with charge excitation are achieved up to 2.38 mC m⁻² and 286.7 mJ m⁻², respectively, in the ambient atmosphere with 5% relative humidity. Furthermore, the experimental result indicates that the actual charge density in the electrode is over 4.0 mC m⁻², showing a large possibility for further promotion of output charge density using this method. As illustrated in Figure 2(b), Wang et al. proposed a high-performance TENG based on the shuttling of charges [79]. The charge shuttling TENG consists of a pump TENG, a main TENG, and a buffer capacitor. The electrodes of the main TENG and the buffer capacitor form two conduction domains, presenting a quasisymmetrical structure with a side and a side. The capacitance of the main TENG changes upon contacts and separations, while that of the buffer capacitor remains constant, inducing voltage differences between them. Therefore, the charges would be shuttled between the main TENG and the buffer capacitor in a quasisymmetrical way, generating electricity on the two loads. When the main TENG changes to the contact state, the capacitance of the main TENG grows, causing the voltage on it to descend. Therefore, charges return from the buffer capacitor to the main TENG via the loads. Consequently, an ultrahigh projected charge density of 1.85 mC m⁻² is obtained in the ambient conditions. Based on this mechanism, an integrated device for water wave energy harvesting shows the feasibility of the charge shuttling TENG as a fundamental device to be applied in complex structures for various practical applications. TENG in the lateral sliding mode provides an effective approach for the in-plane low-frequency mechanical energy harvesting. However, as the output enhancement strategies such as surface modification and charge excitation are not well applicable for this mode, the strategy to promote its output performance is rarely proposed [80, 81].

As shown in Figure 2(c), He et al. developed a new strategy by a shielding layer and alternative blank-tribo-area-enabled charge space accumulation (CSA) design for enormously improving the charge density of a sliding mode TENG [82]. The CSA-TENG consists of polytetrafluoroethylene (PTFE) and a nylon (PA) film, which also incorporates a shielding layer on the slider and an alternating blank-tribo-area structure on the stator. With the grounded conductive layer covered on the back of the slider, air breakdown can be contained to a great extent, and by further introducing the extra blank-tribo-area structure with charge-dissipated tribo-material on the stator, the CSA effect can be achieved. The CSA process can be clearly observed in the charge output with a fast increment to a stable state, which is consistent with the theoretical analysis. With experimental optimization, the stable output charge density reaches 1.63 mC m⁻². Additionally, when the grounding state of CSA-TENG is cut off, a rapid decline in the charge output is observed, proving the inhibitory effect on the air breakdown of the shielding electrode. In addition, based on the principle of the linearly sliding TENG unit, CSA-TENG can be easily designed to a rotational working mode. Similarly, based on the CSA mechanism, Wang et al. presented an out-of-plane design to achieve high-performance TENG [83]. As shown in Figure 2(d), electrodes B and D are arranged on the left part while electrodes A and C are arranged on the right part of the designed structure. There are two different dielectric combination groups, i.e., PTFE vs. PTFE with 600 μm thickness and FEP vs. PTFE with 520 μm thickness. Both groups can generate the CSA to enhance the output performance. Since the friction material is the same for the PTFE vs. PTFE group, the voltage increases slower than that of the FEP vs. PTFE group. However, due to a thicker dielectric layer, the PTFE vs. PTFE group shows a higher maximum voltage. The maximum voltage is 1400 V and 1150 V for the PTFE vs. PTFE and the FEP vs. PTFE group, respectively. Both two groups reach the maximum peak power at around 200 MΩ with 3.3 and 2.5 mW, respectively. Another common method for increasing the output of TENGs is designing the external circuit. Owing to the advantages of easy integration and being magnet-free and lightweight, the switched capacitor converter (SCC) plays an increasingly important role compared to the traditional transformer in some specific power supply systems [84, 85]. Therefore, a power management system with higher transfer efficiency and multifunctional output mode is urgently needed and has great significance for practical applications of TENGs. As shown in Figure 2(e), Liang et al. developed a new charge excitation system based on the voltage-multiplying circuit (VMC) to achieve high-output TENGs for effectively harvesting the water wave energy [86]. Not only the output performance of a single TENG is increased by multiple times but also a scheme is proposed to realize a high-output TENG network through integrating with the charge excitation circuits (CECs). When connecting with the CEC, the AC outputs of a TENG can be increased by many times and transformed into DC outputs simultaneously. When triggered by real water waves, the outputs of the charge excitation TENG are found to be controlled by the water wave frequency and amplitude. Under the optimal water wave condition with a frequency of 0.6 Hz and an amplitude of 10 cm, the output current and power can reach the maximum values of 25.1 mA and 25.8 mW, respectively. Furthermore, a TENG network integrated with the CECs is proposed and fabricated to harvest the water wave energy, presenting a maximum output current of 24.5 mA and a power of 24.6 mW. The CEC can improve both the output current and power of a TENG, which is the most important difference from an ordinary voltage doubler circuit that only increases the output voltage. Finally, the high-output TENG network is utilized to drive a thermometer to work continuously and realize wireless communication with a mobile phone for remote environmental monitoring. However, the high output impedance and switching loss largely reduce the SCC's power efficiency, due to the imperfect topology and transistors. To address this issue, as shown in Figure 2(f), Liu et al. proposed fractal design-based switched capacitor converters (FSCC) with characteristics including high conversion efficiency, minimum output impedance, and electrostatic voltage applicability [87]. Considering the low charges (~200 nC) in TENG, large switching loss, and zero gate voltage drain current (>15 μA) of MOSFET and superlow leakage current (

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