Research Update: Hybrid energy devices combining nanogenerators and energy storage systems for self-charging capability

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Abstract
The past decade has been especially creative for nanogenerators as energy harvesting devices utilizing both piezoelectric and triboelectric properties. Most recently, self-charging power units using both nanogenerators and energy storage systems have begun to be investigated for portable and wearable electronics to be used in our daily lives. This review focuses on these hybrid devices with self-charging combined with energy harvesting storage systems based on the most recent reports. In this research update, we will describe the materials, device structures, integration, applications, and research progress up to the present on hybrid devices.

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The past decade has been especially creative for nanogenerators as energy harvesting devices utilizing both piezoelectric and triboelectric properties. Most recently, self-charging power units using both nanogenerators and energy storage systems have begun to be investigated for portable and wearable electronics to be used in our daily lives. This review focuses on these hybrid devices with self-charging combined with energy harvesting storage systems based on the most recent reports. In this research update, we will describe the materials, device structures, integration, applications, and research progress up to the present on hybrid devices. © 2017 Author(s).

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I. INTRODUCTION

The harvesting, storage, and utilization of energy are becoming a worldwide issue due to the energy crisis, environmental pollution, and the fast development of electronics for our daily life. It is very urgent to find sustainable and renewable energy sources because of shortages of fossil fuels and rapid increases in our energy consumption. In fact, there are considerable wasted energies in our environment, such as wind energy, and thermal, tide, solar, and various forms of mechanical energy. Among these energy sources, mechanical energy would be the most widely distributed energy form, existing all over our living environment including the human body (Fig. 1).

Researchers have developed nanogenerators with piezoelectric and triboelectric properties that can be used to convert mechanical energy into electricity, which are effective devices to harvest low-frequency mechanical energy. As well-known, the nanogenerator device and its material have the advantages of high output, simple design, low cost, flexibility, thin device packages, light weight, and excellent harmony with other materials and devices. To date, the development of device structures, materials, systems, and micro- and nano-technology has been intensively investigated for improving the performance and achieving new applications of nanogenerators. Nevertheless, nanogenerators have a fatal drawback: to be used for practical electronics, they need to generate continuous power, although a nanogenerator can generate instantaneous or pulsed power (i.e., alternating current) when it takes mechanical energy as an input. Otherwise, conventional energy storage systems (ESSs) with high power density, long cycle lifetime, and high capacitance, such as...
FIG. 1. Recent developments in hybrid devices based on nanogenerators and energy storage systems through integration, hybridization, and all-in-one designs for self-charging energy systems for future electronics.

supercapacitors\textsuperscript{23–38} and batteries\textsuperscript{39–42} can supply continuous power to operate the electronics\textsuperscript{43,44}. To overcome this critical problem of currently developed nanogenerators, the utilization of ESSs with nanogenerators should be essential for practical use in the future.

Based on this background, researchers have tried to integrate nanogenerators and ESSs to efficiently utilize the generated electricity for portable electronic devices such as mobile phones, tablets, bendable displays, portable electronic paper, wearable personal multimedia devices, and some medical devices\textsuperscript{45,46}. Very recently, within the last one and a half years, many researchers have tried to fabricate and reported hybrid devices containing nanogenerators and ESSs that were developed via simple integration (circuit connection), hybridization of materials and systems, and all-in-one device design (Fig. 1). Here, we summarize the most recent research results on hybrid devices with self-charging capability and introduce their materials, concepts, and progress.

II. NANOGENERATORS

Basically, the nanogenerator can be divided into representative two devices depending on piezoelectric and triboelectric properties, called the piezoelectric nanogenerator (PENG) and triboelectric nanogenerator (TENG), respectively. The first demonstrated nanogenerator was the PENG using a ZnO nanowire (NW),\textsuperscript{17} which was operated by the piezoelectric property. The formation of a piezoelectric potential or piezopotential was arisen from the breakage of central symmetry in the ZnO crystal structure by external force. In detail, the ZnO crystal with wurtzite structure has a stacked structure of tetrahedrally coordinated O\textsuperscript{2−} and Zn\textsuperscript{2+} along the c-axis (Figs. 2(a)–2(i)). The structure is deformed while an external force is applied. Therefore, the charge centers of the cations and anions are isolated and an electric dipole is formed, resulting in the generation of a piezopotential. The ZnO-based PENGs have been developed as various types, as shown in Fig. 2(a).\textsuperscript{13,18–30} This basic principle and the model of power generation apply to other PENGs based on various piezoelectric materials, such as lead zirconate titanate (PZT)\textsuperscript{51–53} BaTiO\textsubscript{3} (BTO),\textsuperscript{54} poly(vinylidene fluoride) (PVDF),\textsuperscript{55–58} and various two-dimensional (2D) materials (i.e., MoS\textsubscript{2}\textsuperscript{59–61} (Figs. 2(b)–2(d))). Material selection and structural design are the key factors for the development of PENGs, which are based on the coupling of piezoelectric materials and flexible substrates.

The all-polymer-based flexible TENG based on triboelectrification and electrostatic induction was invented in 2012,\textsuperscript{21} which could convert mechanical energy into electricity. Interestingly, triboelectrification can be found everywhere in the surrounding environment and in most common materials used every day in our daily lives. In detail, the working mechanism of TENGs can be
explained as shown in Fig. 2(e). When the TENG is pressed by external energy, the two materials come into contact. Because of their different electron-attracting abilities, electrons could be transferred from one material to the other. Then, there are net negative charges on the surface of the layer with a strong electron attracting ability and net positive charges on the opposite layer. When the two materials are isolated, the tribo-charges in the interfacial regions are separated. This can generate an electrical potential in the interfacial region and electrons in the attached induction electrodes can be driven to flow from one side to the other. In this process, electrons continue flowing until the TENG is fully released. At this stage, both the induced potential difference and the amount of transferred charges can be obtained as their maximum values. When the two materials approach, the potential difference decreases and the electrons flow back. Therefore, the entire process generates an alternating current pulse output. In order to provide stable power using nanogenerators, ESSs such as supercapacitors and rechargeable batteries are essential for future electronic devices (Fig. 2(f)). Thus, the selection of the materials, structural design, and circuit connections in designs for hybridizing ESSs and nanogenerators should be properly considered and designed because most nanogenerators have thin, lightweight, flexible substrates, and require bendable, and stretchable device formation. In addition, wired and textile ESSs and other new functions are required according to the developments of advanced nanogenerators with higher output power and new application in future.

III. INTEGRATION OF NANOGENERATORS AND ESSs

To test the efficient charging of the battery for hybrid devices, recently, researchers have studied how to integrate the TENGs with lithium-ion batteries (LIBs) using a rectifier. In 2016, Pu et al. demonstrated efficient charging of LIBs by a rotating TENG with pulsed output current, as shown in Fig. 3(a).

Fast Li-ion extraction from the typical electrode materials LiFePO$_4$ and Li$_4$Ti$_5$O$_{12}$ was achieved by the TENG at a rotation speed of 250 rpm. The estimated coulombic efficiency of the TENG charging and the following 0.5 C discharging can be higher than 90%, comparable with constant current charging. Interestingly, improvement of the power utilization efficiency (up to 72.4%) in transferring power from the TENG to the LiFePO$_4$-Li$_4$Ti$_5$O$_{12}$ full battery was achieved by optimizing the coil ratio of a transformer. High efficiency was achieved when the impedance of the TENG was
FIG. 3. Hybrid devices based on integration of TENG and battery. (a) Charging of LIB with pulsed output current from rotating TENG [(i) scheme of the rotating TENG, (ii) comparison of cycling performances of LiFePO$_4$ (at 160 mA g$^{-1}$) and Li$_4$Ti$_5$O$_12$ (at 350 mA g$^{-1}$), both with Li-metal as counter electrode, charged by pulsed and constant current, (iii) equivalent circuit for battery charging by the TENG with the aid of a transformer and a rectifier (upper) and The effect of the transformer coil ratio on matched impedances of the TENG, and the power utilization efficiency of the TENG at 250 rpm when charging an LFP–LTO full cell (lower)] Reprinted with permission from X. Pu, M. Liu, L. Li, C. Zhang, Y. Pang, C. Jiang, L. Shao, W. Hu, and Z. L. Wang, Adv. Sci. 3, 1500255 (2016). Copyright 2016 Author(s), licensed under a Creative Commons Attribution 3.0 Unported License. Ref. 64. (b) Charging of LIB with pulsed energy from traditional TENG. [(i) TEM image of synthesized cathode material Li$_3$V$_2$(PO$_4$)$_3$/C and compared the storage efficiency with most popular cathodic materials: LiCoO$_2$, LiFePO$_4$, and LiMn$_2$O$_4$ (Fig. 3(b)) Ref. 65. They simply prepared the integrated hybrid device using a radial-arrayed rotary TENG and coin cell type batteries containing different cathodic materials. The Li$_3$V$_2$(PO$_4$)$_3$/C nanocomposite possessed a discharge capacity of 128 mAh g$^{-1}$ at 1 C, which is very close to the theoretical capacity of Li$_3$V$_2$(PO$_4$)$_3$ (133 mAh g$^{-1}$) in the potential range of 3.0-4.3 V vs. Li/Li$^+$. The capacity retention was up to 90% after cycling for 1000 times at a rate of 5 C, corresponding to a capacity fading of 0.01% per cycle. The Li$_3$V$_2$(PO$_4$)$_3$/C composite showed outstanding energy conversion efficiency when it was charged by a large high frequency current produced by the TENG, far better than for any other known cathode materials, including LiCoO$_2$ (66.1%), LiFePO$_4$ (74.4%), and LiMn$_2$O$_4$ (73.6%). These results show that the selection of electrode materials is important for efficient charging in hybrid devices.

reduced to close to that of the battery cell. In addition, they showed that a 1 h charging of a commercial LIB by the rotating TENG (600 rpm, 36.7 transformer coil ratio) can exhibit a discharge capacity of 130 mAh.

In fact, the energy conversion efficiency is very important when the generated output power is stored in ESSs. Nan et al. prepared the cathodic material Li$_3$V$_2$(PO$_4$)$_3$/C and compared the storage efficiency with most popular cathodic materials: LiCoO$_2$, LiFePO$_4$, and LiMn$_2$O$_4$ (Fig. 3(b)). They simply prepared the integrated hybrid device using a radial-arrayed rotary TENG and coin cell type batteries containing different cathodic materials. The Li$_3$V$_2$(PO$_4$)$_3$/C nanocomposite possessed a discharge capacity of 128 mAh g$^{-1}$ at 1 C, which is very close to the theoretical capacity of Li$_3$V$_2$(PO$_4$)$_3$ (133 mAh g$^{-1}$) in the potential range of 3.0-4.3 V vs. Li/Li$^+$. The capacity retention was up to 90% after cycling for 1000 times at a rate of 5 C, corresponding to a capacity fading of 0.01% per cycle. The Li$_3$V$_2$(PO$_4$)$_3$/C composite showed outstanding energy conversion efficiency when it was charged by a large high frequency current produced by the TENG, far better than for any other known cathode materials, including LiCoO$_2$ (66.1%), LiFePO$_4$ (74.4%), and LiMn$_2$O$_4$ (73.6%). These results show that the selection of electrode materials is important for efficient charging in hybrid devices.

IV. ALL-IN-ONE HYBRID DEVICES AND INTEGRATING NEW FUNCTIONALITIES

The term “all-in-one hybrid device” means that a single device has both an energy harvesting nanogenerator and an ESS in the device without complicated connections. The all-in-one hybrid device design has many advantages for compact, simple, and portable devices, but more efforts are needed to fabricate the devices because of the problem of matching device and material characteristics, such as flexibility, coatings, compositions, electrochemical properties, etc., between the nanogenerator and the ESS. Very recently, researchers have reported and suggested new concepts for all-in-one devices.
In 2016, Wang et al. reported a new nano-energy cell (NEC) that uses high density piezoelectric nanowires to harvest mechanical energy and has a large electrolyte (phosphoric acid/polyvinylalcohol (H₃PO₄/PVA) gel electrolyte)-nanowire interface to store electricity in the all-in-one system consisting of a PENG and an electric double-layer supercapacitor (EDLC), as shown Fig. 4(a). The device achieved a continuous output current for over 90 s, and the mechanical-electric energy conversion efficiency of the NEC was over 10 times higher than that of the PENG without increasing the device volume or reducing the efficiency. Interestingly, Ramadoss et al. made an all-in-one device from a PENG and a pseudocapacitor based on PVDF-ZnO and MnO₂, respectively (Fig. 4(b)). The device exhibited self-charging capability under palm impact (aluminum-foil-based device to 110 mV over 300 s; fabric-based device to 45 mV over 300 s). As shown in Fig. 4(c), most recently, Song et al. demonstrated an integrated sandwich-shaped, self-charging power unit (SCPU) with a wrinkled poly(dimethyl siloxane) (PDMS) based TENG and a carbon nanotube (CNT)/paper-based solid-state EDLC. During vibrations, the device can be utilized to simultaneously harvest and store the mechanical energy as electrochemical energy, and it could be charged to 900 mV in 3 h under the compressive stress at 8 Hz. This study showed that their developed novel all-in-one device is a promising candidate for flexible electronics and wearable devices.

Another strand of important research progress in all-in-one hybrid devices can be explained as achieving new functions, such as devices that are stretchable, bendable, twistable, waterproof, and

transparent. Within 1 year, several interesting research programs have been reported as follows. Guo et al. reported the concept of an all-in-one shape-adaptive self-charging power package based on a TENG and EDLC that has been simultaneously demonstrated for harvesting body motion energy to sustainably drive wearable/portable electronics Fig. 5(a). By utilizing the kirigami architecture, an ultra-stretchable kirigami paper-based supercapacitor with 100% stretchability and specific capacitance of $\sim 1 \text{ mF cm}^{-2}$ and $\sim 12 \text{ F g}^{-1}$ was designed and fabricated, which showed good capacitance and superior mechanical stability for application as a super-flexible energy storage device. By utilizing silicone rubber and Ag nanowires, an ultra-stretchable and shape-adaptive TENG was fabricated with an output of $\sim 160 \text{ nC per half working cycle and open-circuit voltage of } \sim 250 \text{ V in the 100% stretching state.}$ By assembling the kirigami-EDLC into the TENG with a full-wave rectifier, an all-in-one shape adaptive device was achieved for harvesting the energy from hand flapping and continually powering an electric watch. As shown in Fig. 5(b), Yi et al. also developed a soft, stretchable, and waterproof all-in-one device based on a TENG and an enclosed polypyrrole-based pseudocapacitor. The fabricated device was washable and waterproof due to the outer surface of the device. This creative concept demonstrated a promising route to develop stretchable and wearable energy harvesters and power sources for stretchable and wearable electronics. A transparent and flexible all-in-one device would be applicable to future portable, wearable, and smart electronics.

V. WEARABLE DEVICES CONSISTING OF ESSs AND NANOGENERATORS

Recently, wearable electronics, such as smart phones, smart watches, healthcare sensors, smart glasses, etc., have been developed and become an important part of our lives. To utilize these electronics, wearable energy devices are consequently required. Conventional ESSs (batteries and supercapacitors) require frequent and inconvenient charging, however. Therefore, wearable self-charging power devices that combine energy-harvesting and energy-storage technologies could be potential solutions.

Pu et al. reported wearable hybrid devices combining nanogenerators with a supercapacitor and a flexible battery, respectively. As shown in Fig. 6(a), they reported the facile and scalable fabrication of an all solid-state flexible yarn supercapacitor and its integration with a TENG for a self-charging power textile. A conformal Ni layer and reduced graphene oxide (rGO) film were successively coated on the surfaces of common polyester yarns (with the product denoted as rGO-Ni-yarn). The resulting symmetric yarn supercapacitor achieved both high capacitance (13.0 mF cm$^{-1}$, 72.1 mF cm$^{-2}$) and stable cycling performance (96% for 10 000 cycles). Moreover, stable performance was observed for 1000 cycles of 180$^\circ$ bending. This result is valuable because they were the first to demonstrate a textile self-charging power system by the yarn supercapacitors with a TENG cloth. In addition, this cost-effective and industrially scalable approach to yarn supercapacitors and self-charging power textiles paves the way to wearable electronics. To fabricate wearable a TENG-battery self-charging power textile, a flexible battery will be essential. Fig. 6(b) shows a wearable power unit integrating a whole-textile TENG-cloth and a flexible LIB belt. Insulating polyester fabrics were transformed into conductive ones with an electroless plating of a conformal Ni film, and they were then utilized both as electrodes in the TENG-cloth and as current collectors in the LIB belt. The TENG-cloth demonstrated its capability of converting the mechanical energy of various types of human motion into electricity, and the LIB belt showed good electrochemical performance, even when it was severely folded at 180$^\circ$ for 30 times. Importantly, the LIB belt was charged by the TENG-cloth for 3 cycles, and it powered a heartbeat meter strap capable of remote communication with a smartphone, verifying the viability of the whole wearable and self-charging power unit for future wearable smart electronics.
VI. MULTI-FUNCTIONAL HYBRID SELF-POWERED WEARABLE DEVICES

Very recently, Wen et al. reported a multi-functional hybridized self-charging power textile system designed to simultaneously collect outdoor sunshine and random body motion energy by a dye-sensitized solar cell (DSSC) and a TENG, respectively, and then storing the energy in a fiber-shaped supercapacitor (Fig. 7). A single fiber-DSSC unit showed an overall power conversion...
efficiency of 5.64% and fiber-TENG can take advantage of human motions, such as jogging, to deliver an output current of up to 0.91 mA. The fiber-supercapacitor showed excellent pseudocapacitance of 1.9 mF cm\(^{-1}\) from RuO\(_2\)-xH\(_2\)O. Due to the properties all-fiber-shaped devices, this textile system can be easily woven into electronic textiles to fabricate smart clothes that can operate wearable electronic devices. These multi-functional hybrid self-powered devices will result in practical human benefits.

VII. SUMMARY AND OUTLOOK

In this review, we have introduced and updated the progress on hybrid devices based on the simple integration of nanogenerators and ESSs to achieve all-in-one systems, wearable designs, new functions, and multifunctional approaches. It is clear that the hybridized devices containing nanogenerators and ESSs are essential for smart electronics in the future. To develop and improve the new systems and performances of hybridized energy devices, further research efforts are needed on the design of devices, materials, integration, and better understanding of various energy harvesting and storage devices. Through this creative hybridization technology, it is expected that other energy harvesting devices using clean environmental energy sources will improve the harvesting capability of nanogenerators. In addition, we hope that superior energy-harvestable nanogenerators with excellent charging efficiency can be applied for integration with next-generation ESSs with high energy storage capability, such as the metal-air battery, Li-sulfur battery, Li-silicon battery, etc.

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