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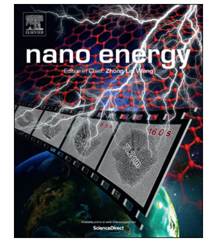

RAPID COMMUNICATION

Transparent flexible stretchable piezoelectric and triboelectric nanogenerators for powering portable electronics

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Abstract
Transparent flexible and stretchable nanogenerators (NGs) that harvest various types of mechanical energy exhibit a great potential for powering low-power portable devices and self-powered electronic systems. Integration of transparency, flexibility and stretchability to NGs has gained a lot of interest for realizing the energy harvesting systems in practical life. Flexible piezoelectric nanostructures, which can generate electrical signal when mechanically deformed, are the most promising candidates for piezoelectric NGs, which offer sufficient power to drive portable electronics and cardiac pacemakers. Moreover, triboelectric NGs enlighten a new technique to harvest mechanical energies with high conversion efficiency. This review highlights the recent research progress of transparent and flexible ZnO nanorods/nanowires, two-dimensional ZnO nanosheets, stretchable micro-patterned P(VDF-TrFE) polymer, $ZnSnO_3$ nanocubes-based piezoelectric NGs along with graphene and hydrophobic sponge structure-based triboelectric NGs, and their potential applications in powering portable electronics are summarized and presented. Finally, the power generation under different modes of pressure/friction such as vertical compressive, bending, contact-separation and stretching are collected and the involved future challenges are discussed in terms of device configuration and efficiency.

Introduction

The development of new renewable electricity generation technologies is immensely important for preserving the global

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environment and assuring sustained economic growth [1-5]. The nanogenerators (NGs) that can harvest various forms of energy from the living environment offer a possible solution [6-9]. From the year 2006, various kinds of NGs have been fabricated to convert mechanical energy into electricity using piezoelectric effect for the realization of the sustainable and self-powered operation of low power-consuming devices and micro-/nano-systems [10-15]. In 2006, Wang et al. first demonstrated electric output from vertically aligned ZnO nanowires (NWs) using atomic force microscopy (AFM) studies [16]. After that, considerable effort was focused on the development of piezoelectric energy harvesters [17-27].

Especially, transparency, flexibility and stretchability of NGs provide several advantages over others such as self-powering with high transparency, multi-shape transformability, easy operation even under ultra-low pressures of other forms of mechanical energies such as bending, folding and stretching [25-30]. Flexible NGs driven by heart beating, blood flow, contraction of the blood vessels, muscle stretching or eye blinking can serve as self-powered implanted sensors such as ultrasensitive sensors for the real-time monitoring and remote patient monitoring [31-34]. In addition, integration of transparency, flexibility and stretchability in NGs offer a fascinating application in rolling-up power source units, self-powered tactile sensors, transparent display panels, transparent electronic skins, robotic sensory skins, electronic papers, wearable power sources, flexible sensors, touch screen devices, smart mobile phones, computers prosthetics and biomedical implantable devices [35-39]. These NGs can also maintain their original properties/shapes after removal of high stresses, and are designed to meet the anticipated requirements of specific environmental conditions.

Among various piezoelectric materials, ZnO nanostructures have been intensively used because of coupling of piezoelectric and semiconducting properties of ZnO. Further, ZnO nanostructure can be easily integrated with transparent flexible plastic substrates, and has been proven to be one of the excellent candidates for next generation self-powered transparent and flexible electronics devices [16,20,21]. Biodegradable and biocompatible properties of ZnO make them suitable for body implantable medical devices [40,41]. Pioneering work on the fabrication of flexible transparent ZnO-based NGs has been reported from the year 2008. Fully integrated one-dimensional (1D) ZnO nanorods (NRs)-based large-scale piezoelectric charge-generating devices with high transparency and flexibility were first demonstrated [17]. Afterwards, a number of flexible piezoelectric NGs based on various morphologies of 1D and two-dimensional (2D) ZnO nanostructures have been demonstrated by many research groups [24,41-47].

However, low electric power output from ZnO-based NGs has restricted their practical applications. For instance, the piezoelectric potential screening effect induced by the presence of a high free carrier density in ZnO is one of the most important and fundamental issues preventing the realization of stable and large power output performance [48-50]. For the dramatic improvement of power-generating performance of NGs, it was found that controlling the defect and free charge carriers in ZnO are essential. In this view, many researchers have focused on the passivation of free carriers and donor defects in ZnO, and various methods have been suggested including thermal annealing treatment, doping with vanadium (V) element and hybridization of ZnO nanostructures with p-type semiconducting polymers to increase the output performance of the NGs in air atmosphere and also to stabilize the electric output under UV light illumination [50-53]. In addition, direct current (DC)-type NGs have also been reported for direct utilization of its generated power for self-powering devices, and to reduce the total size of the power package and power degrade [20,24,47]. Various applications such as self-powered UV detectors and magnetic, temperature, mechanical sensors have been recently demonstrated by flexible NGs [54-57].

However, low piezoelectric charge coefficient (d_{33}) of ZnO nanostructures restricted the realization of high power conversion efficiency [47,56]. Therefore, to resolve this problem, polymer-based poly(vinylidenefluoride-co-trifluorethylene) [P(VDF-TrFE)] NGs have been reported because of their large d_{33} value, pyroelectric properties, low weight, flexibility, transparency, chemical inertness and easy synthesis procedure [22,25,57-59]. Further, acoustic, mechanical and thermal energies are successfully harvested by P(VDF-TrFE)-based NGs with high electric power output by using its coexisting piezoelectric and pyroelectric properties [57-60]. Extremely stable and large-scale energy generation from various hybrid-type composite piezoelectric NGs such as composite of single-crystalline piezoelectric perovskite zinc stannate ($ZnSnO_3$) nanocubes and polydimethylsiloxane (PDMS) without any external electrical poling treatment has also been demonstrated [61-65].

Recently, a new type of simple, cost-effective and robust, power-generating device named triboelectric NGs (TNGs) that converts mechanical energy into electricity using triboelectrification was intensively studied, where the energy conversion is achieved by the periodic contact between two materials that differ in the polarity of triboelectricity yielding surface charge transfer [66-71]. The TNGs offer not only a high electric power output, but also a practical solution for self-powered devices and active sensors. Various applications such as detection of mercury ions, detection of object motion in a tube, real-time tracking of the movement location, self-powered distress signal emitter and ethanol sensor have been successfully realized [72-77].

This review focuses on transparent, flexible and stretchable piezoelectric and triboelectric NGs, and their novel applications in powering portable electronics are summarized. Recent progress in applications of 1D and 2D ZnO nanostructures and ZnO thin film for high performance flexible piezoelectric NGs through doping process, thermal treatment and passivation of free charge carriers via hybridization with p-type polymers will be discussed. This review also summarizes research progress of flexible and stretchable micro-patterned P(VDF-TrFE), $ZnSnO_3$:PDMS composite structure-based piezoelectric NGs as well as triboelectric NGs based on graphene and hydrophobic sponge structure. Finally, the working principles of piezoelectric, pyroelectric and triboelectric NGs under different modes of external stimuli are presented, and their involved future challenges are discussed.

Piezoelectric NG

Piezoelectric NG is an eco-friendly energy-converting system in which waste mechanical energy, such as tiny vibrations or the

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