Nano Energy 40 (2017) 399–407

Contents lists available at ScienceDirect

Nano Energy

journal homepage: www.elsevier.com/locate/nanoen

Full paper

Fully self-healing and shape-tailorable triboelectric nanogenerators based on healable polymer and magnetic-assisted electrode

Wei Xu, Long-Biao Huang, Jianhua Hao*

Department of Applied Physics, The Hong Kong Polytechnic University, Hong Kong, China

ARTICLE INFO

Keywords:
Triboelectric nanogenerators
Self-healing
Energy harvesting
Shape-tailorable

ABSTRACT

Triboelectric nanogenerator (TENG) as a promising mechanical energy harvester has been rapidly developed recently. However, its robustness and endurance are seriously challenged by frequent and inevitable mechanical impacts during operation. Herein, we demonstrate, for the first time, a fully self-healing TENG possessing the ability to recover its performance after damage by introducing healable polymer materials and electrodes consisting of small magnets into the device. Our works imply that high-performance self-healing TENG can be readily realized attributed to not only the excellent mechanical-healing capability of the employed healable polymer but also the quick electric-healing capability of the novel magnetic-assisted electrodes designed for the TENG. The measurements indicate that both the output voltage and current of the healed device can reach up to above 95% of their original values even after the 5th breakage-healing cycle. Additionally, the presented TENG also shows shape-tailorability and object-adaptability. This maximizes the effective contact area of device and further increases the electric output performance to benefit energy harvesting and self-powered sensing of versatile mechanical motions. This research will offer feasible strategies for developing novel mechanical energy harvesting devices and self-powered sensors with recoverability, robustness and adaption.

1. Introduction

Alongside the continued growth in consumption of limited fossil energy, developing sustainable power technology has become urgent to meet the human energy requirement in modern society [1–4]. As abundant and renewable energy, the ambient mechanical energy has been attracting increased attentions due to the extensive availability of target resource and low cost in engineering [5–7]. Accordingly, a variety of mechanical energy harvesters have been developed to convert the ambient energy into usable power such as electricity [8–11]. Among them, triboelectric nanogenerator (TENG), as an emerging technology based on the triboelectric and electrostatic induction, has recently achieved rapid progress from both fundamentals and applications due to its many advantages, including simple fabrication process, expected size and flexible constituent materials choice, and therefore presents great potential as future sustainable power source [12–16].

However, during the harvesting process of mechanical energy, TENGs must be constantly and long-termly exposed to mechanical impacts, originated from not only the external mechanical stimuli but also the internal mechanical friction between contact materials of the device itself [11,17,18]. These mechanical impacts will inevitably lead to the material fracture and eventually TENG failure, generating a series of problems, such as degradation of device performance, reduction of reliability, lack of function, generation of waste, even the serious safety hazards. In order to overcome these obstacles, it is greatly desired to develop a TENG with self-healing feature to restore configuration and property's integrity once the mechanical damage is occurred.

The healable polymer, one kind of smart material allowing to repair fracture to the original state through external stimulus, such as temperature or light [19–24], could be an ideal material to realize self-healing TENG. Given its unique recoverability, the healable polymer has been considered in various functional devices with added self-healing property, such as battery, supercapacitor and electronic skin [25–28]. Compared with these attempted devices, TENGs require far more direct and frequent contact with mechanical stimuli as aforementioned. Therefore, it will be interesting and more realistically significant to conceive healable polymers as the constituent material of TENGs. Unfortunately, few attempts have been made for realizing recoverability of TENGs using such a direct and feasible approach to achieve the self-healing TENG. Besides healable polymers, fully self-healing TENG requires healable electrodes as well for electric generation. At present, most electrodes utilized in self-healing devices are formed based on the connection of conductive fillers, such as Ni particle, carbon nanotube and Ag nanowire [25–29]. Due to the lack of

* Corresponding author.
E-mail address: jh.hao@polyu.edu.hk (J. Hao).

http://dx.doi.org/10.1016/j.nanoen.2017.08.045
Received 15 June 2017; Received in revised form 23 August 2017; Accepted 23 August 2017
Available online 26 August 2017

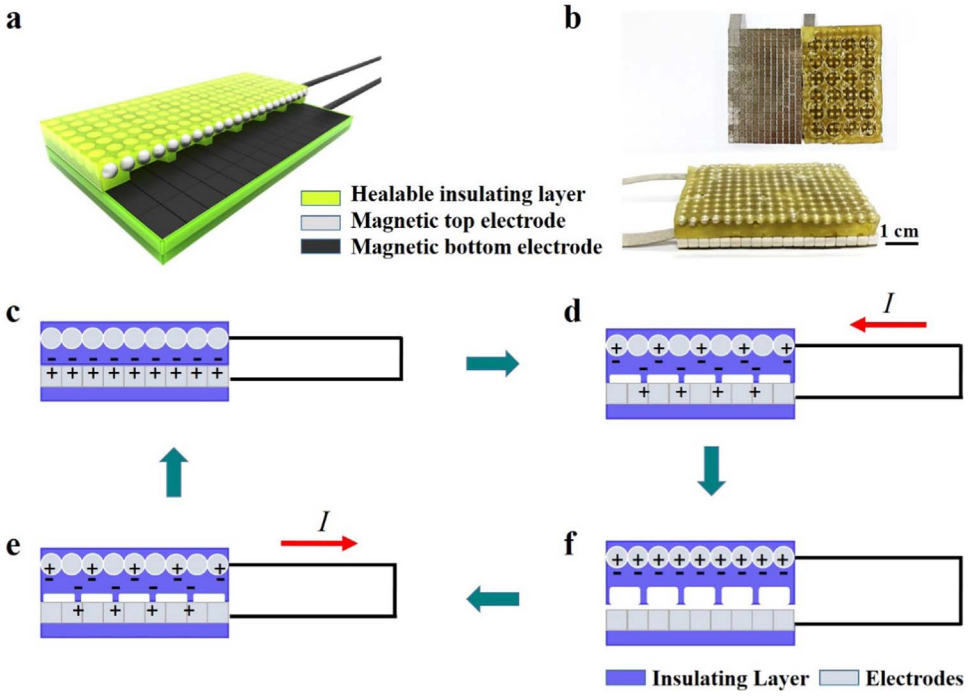

Fig. 1. (a) Schematic of the self-healing triboelectric generator (TENG). (b) Optical images of self-healing TENG (4 cm × 6 cm) with bottom electrode (left) and pattern on undersurface of insulating layer (right) illustrated in the inset. (c)-(f) Schematic illustration of operation principle of self-healing TENG. (c) Contact state (d) Separating state. (e) Contacting state. (f) Separation state.

interaction among these conductive fillers themselves, the healing of the broken electrode can only be achieved by means of the simple contact of conductive fillers around fracture surfaces. This results in the limited healing capability of the electrode and the reduced reliability of the healed device. In this regard, developing healable electrodes with intrinsic healing property is highly recommended in addition to introducing the self-healable polymers into TENG.

Herein, we present a fully self-healing TENG for the first time, which is fabricated by combining both the healable polymer material based on Polydimethylsiloxane-polyurethane (PDMS-PU) and the novel healable electrode consisting of small magnets. When the damage of TENG is occurred, the mechanical property of PDMS-PU and the electric property of magnetic-assisted electrodes can be self-recovered by simply connecting the broken ends. As a result, an excellent self-healing capability of TENG is realized, and the output performance of the self-healed device can be restored to above 95% of their original value after the 5th cutting-healing cycle. Taking advantages of self-healing features, the device is shape-tailorable and object-adaptive to match with applied mechanical stimuli. This can maximize the effective contact area of device, and furthermore increase the performance of device in energy harvesting and self-powered sensing of versatile mechanical motions.

2. Experimental

2.1. Preparation of the healable PDMS-PU

Trifunctional Poly(propylene glycol) (PPG) (Mn = 6000) was purchased from Bayer Materials Science. Hydroxyl-terminated PDMS (Mn = 2000) was purchased from Dow Corning. Isophorone diisocyanate (IPDI), dibutyltin dilaurate (DBTDL), bis(4-aminophenyl) disulde (AFD) and tetrahydrofurane (THF) were purchased from Aladdin-reagent Co., Shanghai, and used without further purification. 25 g trifunctional PPG and 25 g hydroxyl-terminated PDMS were firstly mixed in a 100 ml glass reactor. Then the mixture was heated at 120 °C for 2 h under vacuum and magnetic stirring for water removal. After cooling to 70 °C, 8.38 g IPDI and 0.3 ml catalysis of DBTDL were added to conduct the sealed reaction with the mixture for 20 min at the stirring rate of 120 r/min. Then, the reaction was further evolved at constant temperature of 95 °C for 3.5 h to form the isocyanate terminated prepolymer. The prepolymer was stored in a tightly closed glass bottle for further use after vacuum removal of residual monomers for 5 min. To obtain a healable PDMS-PU layer, 10 g prepolymer was firstly dissolved in 7 ml THF, Then, 1 ml solution of AFD in THF (0.6 g/ml) was added and adequately mixed with prepolymer. The mixture was poured in a Polytetrafluoroethylene (PTFE) dish to allow the curing process at 55 °C for 48 h. At last, the healable polymer elastomer was obtained, which can be used to fabricate insulating layer and substrate of self-healing TENG.

2.2. Fabrication of the self-healing TENG

The top and bottom electrodes were first prepared by the array of magnetic balls with diameter of 3 mm and magnetic cubes with side length of 3 mm, respectively. The utilized magnetic balls and cubes with nickel plating were purchased from Shenzhen Youci Industrial Co., Ltd., China, and Xinhongchang Magnetic Industrial Co., Ltd., China, respectively. Then the mixture of prepolymer and AFD was poured and cured on the top electrode to form a PDMS-PU film, which encapsulates the top electrode inside. A pattern was formed on the surface PDMS-PU as a gap by curing prepolymer with template. The diameter of each circular hole in the pattern is 8 mm, and the thickness of the gap is about 1.5 mm. The self-healing TENG was achieved by assembling the top electrode encapsulated by PDMS-PU film and the bottom electrode attached on another PDMS-PU film as substrate.

2.3. Measurement of the device

The open-circuit voltage and short-circuit current were characterized by LeCroy WaveRunner Oscilloscope (44MXI) with the probe resistance value of 10 MΩ and low noise current amplifier (Stanford Research Systems, SR570), respectively. During the measurement of output voltage, the top and bottom electrodes were directly connected with the both ends of probe of oscilloscope. The noise filter and the coupling of the oscilloscope were 1 bits and AC 1 MΩ, respectively. For the measurement of output current, SR570 was connected between the device and the oscilloscope on the basis of the voltage measurement. The tensile property of healable polymer was measured by INSTRON