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Wide-ranging impact-competent self-powered active sensor using a stacked corrugated-core sandwich-structured robust triboelectric nanogenerator

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ABSTRACT

Herein, we present a stacked corrugated-core sandwich-structured triboelectric nanogenerator (CCS-TENG) that can be used as an enhanced self-powered active sensor that has a working capability under a broad range of external impacts in the environment. The working performance of a single-unit TENG can be tuned by varying the core diameter of the corrugated-core structure and locating it in a crosswise and/or lengthwise orientation between two sandwich plates. The assembly of the corrugated-core structure also offers a steady gap between the sandwich plates under a strain-free condition. A CCS-TENG with a crosswise assembly exhibits higher electrical output (142–153 V; 3.2–3.9 μA) than with a lengthwise assembly (107–123 V; 2.7–3.1 μA) at a fixed pressing frequency of 3 Hz and different applied forces ranging from 3.0–10.1 N. A constant output voltage of 142 V and an output current of 8.8–9.3 μA are measured at 3–10 Hz and 3.0–10.1 N when four CCS-TENGs with four different core diameters arranged crosswise are stacked in parallel. As a self-powered active hydrogen (H_2) sensor, the device can efficiently detect H_2 with a high response magnitude (~83%) and a broad detection range of 0.001–1.000 vol% at the above external frequency and force range. We expect that this exceptional advancement will pave the way for a next-generation self-powered active sensor for integrated systems that has competency under a broad range of external mechanical impacts in the environment.

1. Introduction

With the current rapid technological advancement along with the overexploitation of fossil fuels, the world's reserved energy sources are running out. Climate change and global warming due to the consumption of fossil fuels have become the most important environmental and human-kind concerns. As a consequence, harvesting green energy from the environment has attracted intense global attention as it is considered to be the best option to provide relief from the current energy crisis [1]. Nanogenerators and solar cells are capable of filling the current needs by harvesting green energy from mechanical vibration, heat, and light, and they have been used for some time to provide independent, uninterrupted, maintenance-free power supplies to numerous applications, such as personal electronics, implantable sensors, and environmental monitoring [2,3]. Among the various energy sources, mechanical motion/vibration is abundant in our environment, and it can be harvested from wind, water flow, and daily life activities, such as human motion, using various nanogenerators based on piezoelectric [4,5], electrostatic [6,7], and electromagnetic [8] properties. Most recently, harvesting green energy using a triboelectric nanogenerator (TENG) that employs triboelectrification has attracted great interest due to its simple and cost-effective fabrication process, miniature size, light weight, easy scalability, outstanding flexibility, diverse formats of mechanical triggering, and biocompatibility [9–11]. The triboelectrification process of a TENG can be realized based on the charge separation and the transfer between the two different polarities of the charged materials after a periodic mechanical contact–separation and/or friction. To date, a number of TENG devices have been explored using various polymer materials and design structures.

The performance of the TENGs mainly depends on the triboelectric charge density on the contact materials, which can be enhanced by selecting the proper combination of contact materials that are far from each other in the tribo-series (i.e., large triboelectric charging polarity), as well as increasing the friction area by modifying the contact surface. To raise the friction areas of the

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tribo materials, nanostructures and surface modifications on polymers have frequently been adopted, and investigations have shown significant enhancements of the devices' output levels [12–17]. Various metals [18,19] and metal oxides [20,21] with controlled surface roughness have also been used to enhance TENG performances. Cellulose is one of the most versatile and widely found eco-friendly biopolymers in nature, and it is attracting increased interest in a wide range of scientific and commercial applications. It exhibits a fascinating natural structure comprising crystallites with interspersed amorphous regions having a low degree of order and linear chains of β-1, 4-D anhydroglucopyranosyl units. More importantly, it exhibits both direct and converse piezoelectricity [22,23]. Further, the deconstruction of native cellulose through acid hydrolysis can yield highly crystalline cellulose that can be used as a soft-electroactive material for various applications, such as energy generation, sensors, optical balancing, and actuators.

A suitable device structure plays a vital role in enhancing the applicability and durability of TENGs. Researchers have proposed TENGs based on a multilayer wavy-structure [24], elastic bellows [25], case-encapsulated cylinder [26], rotating-cylinder [27], interlocking kirigami patterns [28], and origami structure [29]. These reports have demonstrated that, with a proper design configuration, TENGs are capable of harvesting green energy efficiently in both low- and high-frequency ranges, as well as in both small and large scales of external applied forces.

Chemical/gas sensors are a demanding application in various sectors, used for environmental and personal safety purposes. Traditional solid-state or flexible sensors require external power sources to operate, which ultimately limits their usability and portability as wearable devices, such as for smart skin applications. Their continuous maintenance, accuracy, and durability are major concerns. Therefore, researchers are interested in developing self-powered active sensors that harness energy from ambient sources. Recently, studies have extensively investigated piezoelectric and TENG-based self-powered active sensors that detect various gases/chemicals [30–41]. TENGs-based sensors exhibit numerous advantages over piezoelectric-based sensors, such as high output performance, a convenient design mechanism and structure, simplicity, and biocompatibility. However, the reported self-powered active sensors can only work well under certain specific external impacts.

In this paper, we present a corrugated-core sandwich triboelectric nanogenerator (CCS-TENG) for potential application as a self-powered active sensor. The corrugated-core sandwich panel comprises a corrugated sheet of polyimide (PI)-Al-polyethylene terephthalate (PET)-Al-PI film and two flat sheets of microcrystalline cellulose (MC) film. So that this self-powered active sensor will perform well under a broad variety of external impacts, we stacked four CCS-TENGs together and investigated the sensor in a hydrogen (H_2) environment. We expected the proposed sensor to show enhanced workability under a broad variety of impacts from the environment.

2. Experimental

2.1. Preparation of MC suspension and film

To obtain an aqueous suspension of microcrystalline cellulose (MC), commercial grade cellulose powder (~20–100 μm; cotton linters pulp) obtained from Dae Jung Chemicals Co. Ltd. was first hydrolyzed in 35% hydrochloric acid (HCl). Using a typical process, 2.4 g of cellulose powder was added to the 20 wt% HCl pre-heated at 40 °C in an Erlenmeyer flask and allowed to hydrolyze the cellulose at 55 °C in a hot water bath under continuous stirring at 700 rpm for 1 h. The cellulose suspension was then diluted with cold de-ionized (DI) water (~10 times the volume of the acid solution used) to stop the hydrolysis, and was kept overnight to settle. The suspension was then cleaned by DI water and a thick white precipitate of MC was obtained via several times centrifugation. Last, the MC precipitate was diluted into 40 mL DI water with a concentration of 0.09 g/mL. To prepare the film, 6–14 mL of MC aqueous suspension was poured into a soda-lime glass Petri dish and allowed to evaporate in an oven at a controlled temperature of 35 °C for 26 h. Finally, flexible MC films of basis weights 70–80 gm^{-2} with thicknesses of 35–60 μm were obtained.

2.2. Fabrication of the CCS-TENG

Polyethylene terephthalate (PET) film (thickness ~90 μm) was periodically bent into a corrugated shape using a set of metal rods. Specifically, PET film was wrapped sinusoidally or periodically onto a set of metal rods and tied tightly, placed into a furnace at 170 °C for 3 h, and cooled naturally. Four different corrugated-core-shaped PET films were prepared using metal rods with varying diameters of 1, 2, 4, and 6 mm. Then, 230 nm thick aluminum (Al) was deposited on both side of the corrugated PET films via a thermal evaporator (thermal evaporator system, KVT-438). Subsequently, polyimide (PI) film (thickness ~70 μm) was attached onto the Al on both sides of the corrugated films to obtain a PI-Al-PET-Al-PI corrugated-core structure. Consecutively, two pieces of MC film with 230 nm thick Al deposited on one side was attached on the PET substrates. Finally, the PI-Al-PET-Al-PI corrugated-core film was sandwiched using the two MC-Al-PET substrates with MC films facing inside, and the corrugated-core sandwich structure was packaged using scotch tape. The wire bonding of the fabricated device was then completed because the device could be considered as two capacitors connected in parallel. The dimensions of the as-fabricated corrugated-core sandwich TENG (CCS-TENG) was 4 cm × 2.5 cm (length × width). Importantly, two different sandwich structures were fabricated by orienting the corrugated-core film lengthwise and crosswise inside the sandwich panel. Furthermore, to realize the CCS-TENG as a self-powered active sensor, 10–15 nm thick palladium (Pd) film was sputtered (RF and magnetron sputtering system, KVS-C4055) on each trough (bottom of corrugation) of the corrugated film. The overall schematic illustration of the as-fabricated device is shown in Fig. 1.

2.3. Characterization of the CCS-TENG

To measure the output performance of the CCS-TENG, we mounted the device to a linear motor (FASTECH, Ezi-Servo) that can provide constant contact force at tunable frequencies. The open-circuit voltage and short-circuit current of the device were determined using a functional oscilloscope (Lecroy Wave Runner 610) and a low-noise current measurement system (Keithley 6485 picoammeter) at pressing forces of 3–20 N at between 0.5 and 15 Hz. To characterize the sensing properties of the CCS-TENG-based self-powered active sensor, a mixture of target gas (hydrogen, H_2) and synthetic air was supplied onto the device at a constant flow rate of 50 sccm (standard cubic centimeters per minute), with different target gas concentrations. The response magnitude of the device was calculated using the following formula: $S(\%) = \left(V_a - V_g\right)/V_a \times 100$, where S denotes the response of the device, and V_a and V_g are the voltages in air and in the presence of a certain amount of gas, respectively.

3. Results and discussion

The detailed fabrication process of the device is described in the experimental section. Specifically, the length and width of the sandwich panel were fixed at 4 cm and 2.5 cm, respectively, while