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Full paper

Fully stretchable and highly durable triboelectric nanogenerators based on gold-nanosheet electrodes for self-powered human-motion detection

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ABSTRACT

A patchable triboelectric nanogenerator (TENG) is highly promising for self-powered human-motion detection, but it may undergo repeated stretching/releasing cycles during daily activities of a human, which may lead to mechanical fracture of each component and degradation in electrical output performance of the TENG device. Here, we report a fully stretchable and durable triboelectric nanogenerator (TENG) with gold (Au) nanosheets (NSs) embedded into both PDMS matrix and micropyramid-patterned PDMS. It was found that a new design of the Au NS electrodes dramatically improves the mechanical flexibility and stretchability, enabling to achieve the outstanding output stability of the Au NS electrode-based TENG (Au NS-TENG) during 10,000 cycles of repeated pushing and stretching tests. Our fully stretchable and durable Au NS-TENGs were successfully applied to the hand joints that can be used in the self-powered human-motion detection processes for wearable applications.

1. Introduction

Recent interest in wearable or portable electronic devices has raised concerns about high-quality flexibility and stretchability, which are of interest to next-generation electronic device applications such as flexible displays, flexible and/or stretchable circuits, artificial electronic skins (e-skins), and various form-type sensors [1–6]. Accordingly, the need to replace or charge the battery must be removed regarding such electronic devices; otherwise, a heavier, separate power supply will remain a technological hurdle to user comfort over a long time period [7,8].

Recently, a new type of power-generation device called a triboelectric nanogenerator (TENG) has been developed based on the coupling mechanism between the triboelectric charge and the electrostatic induction for which mechanical energy, which is one of the most common energy sources in the surrounding environment, is used [9–12]. Because of the many advantages of the TENG, such as a low cost, a simple manufacturing process, and a high power density [13–18], the TENG can be successfully demonstrated without a battery or an external power supply due to the use of promising device and application energy-harvesting technologies such as voice-recognition devices, distress-signal emitters, trace-memory systems, velocity sensors, electroplating, electropolymerization, and water splitting [19–23]. In the TENGs, the triboelectric charging occurs at the contact interface through a periodic repetitive physical contact between two different materials of different polarities in terms of the triboelectric series, so the contact surface must be able to generate a robust and stable electrical output [24–26]. Although many TENGs have been previously reported, the reports of TENGs that are highly elastic, durable, and robust against repetitive external movements are rare.

To achieve devices that are 100% human-wearable, all of the components that make up the device must be flexible and stretchy [27–30]; particularly, the flexibility and stretchability of the electrode portion that serves to transfer the carrier to the external circuit to drive the device must be excellent [31–33]. However, most of the previously reported TENGs are lacking because the metals of the conductive electrodes in the TENGs such as gold (Au), aluminum (Al), or copper (Cu), which are not stretchable generally, are considered as the counterparts of triboelectric films [34–39].

To overcome this hurdle, it has become necessary to develop flexible and stretchable electrodes that are mechanically robust, electrically stable, and reliable for repetitive external forces. In this study, a fully stretchable and durable TENG is developed using Au nanosheet (NS)-embedded electrodes. The Au NS electrodes were built into the following two layers: the opposite side of the patterned polydimethylsiloxane (PDMS) for the top electrode and the friction layer (if it also serves as the bottom electrode). Also, the operating mechanism of the Au NS electrode-based TENG (Au NS-TENG) was confirmed in the

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pushing and stretching modes using the COMSOL software. The new design of Au NS electrodes dramatically improves the mechanical flexibility and stretchability, demonstrating the proposed TENG's outstanding output stability over 10,000 cycles of repeated pushing and stretching tests. Therefore, the proposed Au NS-TENG can be successfully applied to the hand joints that can be used in the self-powered human-motion detection processes for wearable applications.

2. Material and methods

2.1. Synthesis of the Au NS

According to the typical synthesis of Au NSs, 5 mL of aqueous solution containing 1.7 mg of L-arginine (Sigma Aldrich) was hosted in a 20-mL vial at room temperature and the solution was heated to 95 °C. Meanwhile, 2 mL of aqueous solution containing 13.5 mg of hydrogen tetrachloroaurate trihydrate ($HAuCl_4 \cdot 3H_2O$) that was obtained from Alfa Aesar was injected into the L-arginine solution using a pipette. The reaction mixture was maintained at 95 °C for 2 h and then cooled down to room temperature.

2.2. Fabrication of the Au NS-embedded electrodes

A Petri dish was filled with deionized water, and the Au NS dispersion in 1-butanol was dropped onto the water surface using a pipette. When the monolayer film of the Au NSs was formed at the water surface, the Au NS film was brought into contact or scooped up by an Si-wafer coated with a Teflon substrate. The multilayer Au NS film was obtained by repeating this process. Then, the Au NS film on the substrate was annealed at 100 °C for 10 min. The Sylgard 184 PDMS (Dow Corning) was prepared by mixing the base and the curing agent with a weight ratio of 10:1. The liquid PDMS was poured onto the Au NS film and then cured at 80 °C for 6 h. After the PDMS curing, the Au NS film-embedded PDMS matrix was peeled off from the glass substrate. Cu wires were connected to the Au NS film-embedded PDMS matrix using a silver paste, and the wire was coated with PDMS glue. The Au NS-embedded electrode with the micropyramid-patterned PDMS (100 μm in width) was fabricated using the same protocol; however, the micropyramid-patterned Si wafer was covered with the Au NS film/liquid PDMS on the Si-wafer substrate.

2.3. Characterization of the Au NS-embedded electrodes

Field emission scanning electron microscope (FE-SEM) images were captured using the SUPRA 55VP device (Carl Zeiss). Atomic force microscope (AFM) images were obtained using the XE100 device (PSIA) under tapping modes. The electrical performance of the Au NS electrode was measured using the Keithley 2634B device (Tektronix).

2.4. TENG measurements

All of the device measurements were performed under ambient conditions (temperature = 23 °C and relative humidity = 25–40%). A DPO 3052 digital phosphor oscilloscope (Tektronix) and the SR570 low-noise current preamplifier (Stanford Research Systems, Inc.) were used to measure the electrical properties of the TENG.

3. Results and discussion

Fig. 1a shows the schematic illustration of a highly stretchable and durable electrode based on a multilayered Au NS film-embedded PDMS matrix. The Au NSs that were used in this study were synthesized using the previously reported method, and the thickness and lateral size are 20 nm and 20–50 μm, respectively (Fig. S1) [6,40,41]. First, an Au NS multilayer film was prepared by transferring a monolayer of the Au NSs that were floating on a water surface onto a silicon (Si) wafer coated with polytetrafluoroethylene (PTFE) film followed by a repeating of the transfer process. After each transfer, the film was annealed at 100 °C for 10 min. Thermal annealing at this mild temperature increases the contact between stacked Au NSs. Then, the liquid PDMS was cast onto the Au NS film, which filled the spaces between the NSs. After the PDMS curing, the Au NS film-embedded PDMS matrix was detached from the substrate. The weak adhesion between the Au NS film-embedded PDMS matrix and the PTFE-coated substrate facilitated a clean

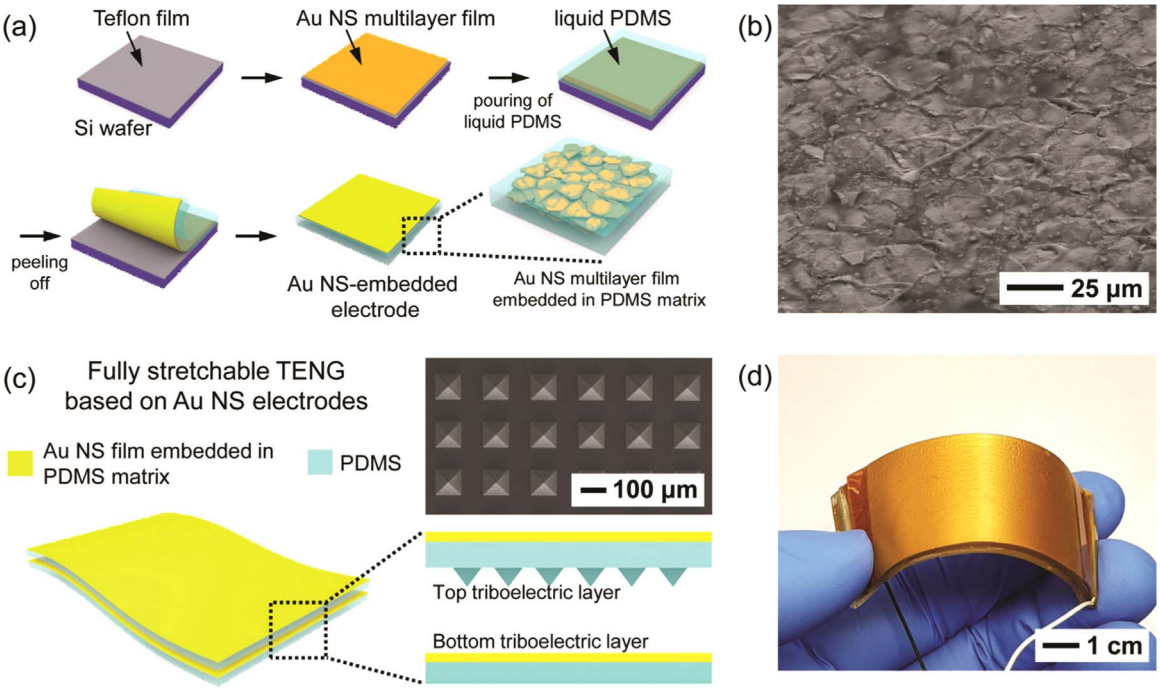

Fig. 1. (a) Schematic illustration of the fabrication process of the Au NS-embedded electrode. (b) Top-view FE-SEM image of the Au NS-embedded electrode. (c) Schematic illustration of the structure of the fully stretchable TENG based on the Au NS-embedded electrodes. The FE-SEM image shows the micropyramid-patterned PDMS (10 μm width) of the top triboelectric layer of the TENG. (d) Photograph of the Au NS-TENG.