High-Performance Flexible Piezoelectric-Assisted Triboelectric hybridized nanogenerator Based on Ecoflex/Mxene/$LiNbO_3$ Composite Films

Biswajit Mahanty and Dong-Weon Lee

Abstract— **With an emerging energy crisis and global warming, energy harvesting technologies have attracted an attention as an alternative to replace the fossil fuel-based energy generation methods because they can generate the carbon-free and sustainable energy. Among various energy harvesting technologies, mechanical energy harvesters are regarded as an attractive harvesting method because they can convert the abundant mechanical energy surrounding us to electrical energy. However, the low electrical performance of the mechanical energy harvester is hindering its practical utilization. Hybridization of the two different mechanical energy harvesters can provide the solution for this issue because the electrical performance of the energy harvester can be improved by harvesting the applied mechanical energy in two harvesters, simultaneously. Herein, first time a triboelectric and piezoelectric hybridized nanogenerator is fabricated by embedding MXene and Lithum Niobate filler in the Ecoflex matrix. The role of MXene as the functional conductive filler and Lithum Niobate as a piezoelectric filler are experimentally investigated and the optimum point of MXene and Lithum Niobate content is investigated and the high open-circuit voltage of 437 V, short-circuit current of 140 μA and power density of 18 Wm^{-2} are obtained. The hybrid nanogenerator (HNG) is capable of driving more than 50 LEDs and can run five different sensor modules.**

I. Introduction

Recently, energy harvesting devices have been widely investigated to handle the global energy crisis and to realize of self-powered electronics such as mobile electronics, wearable sensors, implantable medical devices, and Internet of Things (IoT) based intelligent applications [1-5]. The applications of wearable sensors in IoT-based smart applications are increasing and require a constant power supply. Conventional batteries have some limitations as they need recharging or replacing after specific intervals and contain harmful chemicals that are toxic in nature and vulnerable to society. Hence, sustainable power sources and self-powered electronics have attracted significant attention, and have been extensively investigated to realize sustainable, uninterrupted, or non-exhaustible energy sources. To achieve this goal, various piezoelectric and triboelectric sensors/nanogenerators that can generate electricity from environmental mechanical energy sources (e.g., body movements, vibrations, and rotations, etc.) have been proposed to achieve the long-term functioning of wearable electronic devices. To improve the electrical output of the energy harvesters, a hybridization of triboelectric nanogenerators (TENGs) and piezoelectric nanogenerators (PENGs) can be an attractive solution. Recently, tremendous research work has been carried out in hybrid nanogenerator (HNG). However, the power output is still insufficient, and further improvement is required. In this work, to enhance the deformability and dielectric permittivity with a lowered dielectric loss factor in a triboelectric material, a microstructure composite film was designed using Ecoflex and Lithium niobate ($LiNbO_3$) (LN) nanoparticle coupled 2D MXene ($Ti_3C_2T_x$) nanosheets (referred to as EC/M/LN). Lithium niobate ($LiNbO_3$) is a ferroelectric that exhibits piezoelectric effects. So here, with a polydimethylsiloxane (PDMS) microstructure film and as-prepared Ecoflex/MXene/$LiNbO_3$ microstructure composite film as the triboelectric pair, the fabricated HNG can achieve an open-circuit voltage (V_{oc}) of 437 V, short-circuit current (I_{sc}) of 140 μA.

II. Results and Discussion

Ecoflex composite films (MXene and $LiNbO_3$) with 3 wt. % of each filler were fabricated using additives pre-mixed Ecoflex A and B (1/1, w/w) on rough sandpaper (grit 60). All casted films were cured for 24 h at room temperature. After curing, the films were carefully removed from the sandpaper and stored in a desiccator for further use. The Ecoflex and composite coatings with 3wt% MXene and $LiNbO_3$ were named as EC3M3LN. PDMS elastomer and cross linker were mixed in a 10:1 ratio, and mixed thoroughly for five minutes, and finally, casted on sand paper (grit 60) and dried at 90°C for one hour. After curing the micro-patterened film was removed from the sandpaper. The hybrid nanogenerator (HNG) was fabricated with EC3M3LN based composite film as tribo-positive and tribo-negative (PDMS films) layers without any spacer. Initially, both tribo layers were cut into 2.1 × 2.1 cm^2 pieces and then stuck on one side of adhesive 2 × 2 cm^2 Cu-Ni polyester electrodes followed by encapsulation with PET films. The PET film encapsulation protects the device from the surrounding noise and humidity. The slightly sized composite film protect the interference of the top and bottom electrodes. The energy harvesting performance of HNG is shown in Fig. 1.

Research supported by the National Research Foundation of Korea (NRF).
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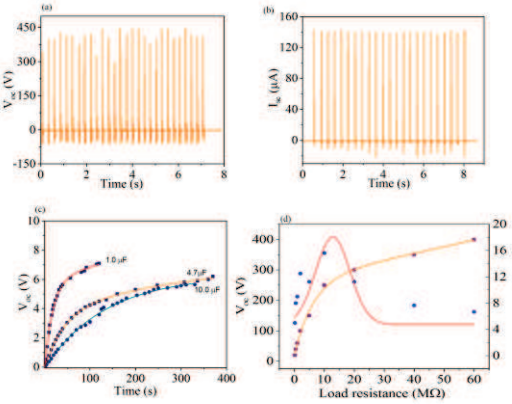

Figure 1. Energy harvesting performance. (a) The open circuit output voltage (V_{oc}) of HNG. (b) The sort circuit output current (I_{sc}) of HNG. (c) Capacitor charging behavior of the HNG at different capacitor values. (d) The variation in the output voltage and power density and external load resistances of HNG.

The open circuit output voltage (V_{oc}) and short circuit output current (I_{sc}) of 437 V and 140 µA under human hand imparting force are achieved as shown in Fig. 1a, b. To the best of our knowledge, the open circuit output voltage (V_{oc} ~437 V) and short circuit output current (I_{sc} ~140 µA) of the HNG is higher than those of previously published HNGs [6-9]. A synergistic improvement in the energy-harvesting performance was observed via lowering the dielectric loss by ferroelectric coupling of the LN and enhancing the dielectric permittivity by using 2D-MXene nanosheets. To demonstrate the potential application of the HNG in energy storage and fast charging systems, it was connected to three commercial capacitors (1.0, 4.7, and 10 µF) individually, as shown in Fig. 1c. The energy (W) stored in the capacitor was estimated as $P_c = \frac{CV_S{}^2}{2}$, where C is the capacitance of the charging capacitor and V_S is the saturation voltage. The energy stored in the capacitors was determined to be 3.5, 14.1, and 28.5 µJ respectively. Furthermore, the instantaneous voltage drop (V_L) of the HNG with varying external load resistance (R_L) was measured under a constant pressure condition, and is presented in Fig. 1d. It is worth mentioning that the generated load voltage (V_L) gradually increased as the function of load resistance increased until it reached saturation at an infinitely high resistance (R_L; ~60 MΩ), corresponding to the open circuit voltage (Fig. 1d). The instantaneous power density (P) of the HNG was estimated using $P = \frac{V_L{}^2}{A.R_L}$, where A is the effective surface area and V_L is the voltage drop across the load resistance R_L. The variation in P with a change in the external load resistance (R_L) is shown in Fig. 1d. The results revealed that P increased naturally with an increase in R_L, and achieved a maximum value of 18 W.m^{-2} at an R_L of ~13 MΩ which is superior to the recently published works on HNG [10-11].

III. Conclusion

In this work, an HNG was fabricated by embedding MXene (conductive filler) and $LiNbO_3$ (piezoelectric filler) in the Ecoflex matrix. As a result, the V_{oc}, I_{sc}, and instantaneous power density of the HNG were found to be 437 V, 140 µA and, 18 W.m^{-2} respectively. In this work, we used MXene and LN as conductive fillers to derive the synergistic effect of piezoelectricity and triboelectricity in a hybrid nanogenerator. The HNG is capable of driving more than 50 LEDs and can run five different sensor modules.

Acknowledgment

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korean government (MSIT) (No. RS-2022-00165505) and (No. 2020R1A5A8018367).

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