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Nanosheet Zinc Oxide Synthesized by Solution-Immersion Method for Triboelectric Nanogenerator

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ABSTRACT

Most global problems are being solved by using sustainable energy harvesting technologies to retain the social ecosystem in great condition. The triboelectric nanogenerator (TENG) which is a renewable energy harvesting device, collects the waste mechanical energy from its surroundings and performs the electric signal conversion. TENG have garnered increased attention in recent years by offering prospective use in energy harvesting technology. Particularly, there is a need for flexible energy conversion that serves as a power supply for portable electronic equipment. In this study, ZnO nanosheet thin film prepared on the flexible conductive aluminium foil through a low temperature immersion technique was used to generate electrical energy. The effect of heat treatment on ZnO nanosheet thin film was also investigated on the surface morphology, structural properties and nanogerator performance. The high density of interconnected ZnO nanosheet were observed before and after heat treatment as confirmed by FESEM studies. The analysis using XRD confirmed that ZnO nanosheet thin film was successfully deposited on the aluminium foil. Additionally, the ZnO nanosheet thin films improved significantly with heat treatment, enhancing their crystalline quality. The TENG was successfully constructed in contact and separation

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mode using Kapton tape on top and ZnO nanosheet thin film on the bottom to generate electricity by a force of hand pressing. The output electrical voltage of the device doubled from around 2 V to 4 V after underwent the heat treatment. This study provides an essential insight into fabrication of TENG using the ZnO nanosheet thin film through a clean and effective method for nanogenerator applications.

Keywords:
Triboelectric; nanogenerator; Zinc Oxide; Aluminium; electrical output

1. Introduction

The explosive development of flexible and portable electronics in the booming industry of IoT technology, 5G technology, and big data for numerous different uses including in medicine and health, communication and networking, household appliances and utilities have brought to source of energy concerns and a severe environmental risk. The development of sustainable, low-cost and portable power sources for the current and the future was crucial for ensuring long-term economic growth. This situation causes an increased demand for sustainable energy that can produce electricity from the natural atmosphere. As alternatives to fossil fuels, numerous energy sources, including solar energy, mechanical energy, heat energy, bio- and chemical energy, have been effectively captured and utilized as power sources. Among such energy sources, mechanical energy such as vibrating, flow of water, drop of rain, wind and human body movement indicated a promising resource as the resource is widely available but generally being neglected [1]. Many attempts have been made to energy harvesting technology in powering miniaturized electronics, including self-powered systems, and wearable devices which need to generate a small-scale power output as alternative of using battery [2-5].

Since the invention of nanogenerators in 2006 by Wang and Song [6], zinc oxide (ZnO)-based nanogenerators that can transform mechanical energy into electricity for powering nanoscale devices have garnered considerable interest. Then after, the same team had success in introducing the triboelectric nanogenerator (TENG) in the year 2012 [7]. They reported employing the combination effects of triboelectric charging and electrostatic induction as an energy source for electricity. The generation of charges, separating charges, and electrostatic induction phenomenon can be accomplished as the mechanical bending was applied on the TENG that consisting of polyimide film (Kapton) and polyethylene terephthalate (PET) are stacked one on top of the other. At about 3 V output voltage, a power density of 10.4 mW/cm^2 has been achieved. Since then, numerous triboelectric materials have been utilized in the fabrication of TENG. However, recently reported TENGs had some drawbacks, including high-cost materials, complicated processes, and low stability and durability [8,9]. As a result, scientists are always searching for new materials that could potentially overcome this limitation [10-14]. From the point of view of realizing the high-efficiency TENG, it can be concluded that three important factors need to be considered which are the material selection, contact surface morphology and device structure. The surface structure of materials has an impact on the output behavior of TENG. This factor is related to the surface charge density to facilitate the charge transfer mechanism. One way approach to enhance the surface density is fabricating the nanostructured surface, which at the same time can increase the contact area [15]. Besides, metal oxide-polymer nanocomposites are another way to enhance the charge generation and the dielectric constant value, which is reported to have an excellent surface charge density due to the synergistic effect between polymers and metal oxide nanomaterial.

Nanostructured zinc oxide (ZnO) is one of the metal oxides extensively studied these decades for many applications ranging from electronics, textile, pharmaceutical and cosmetic, photocatalysis, and rubber industries. In electronics industries, ZnO is widely used for electronics, photonics, acoustics and sensing due to its outstanding properties of being a semiconductor with a wide bandgap and a piezoelectric material [16]. ZnO has the potential for usage in power generation and harvesting energy [17-19]. Remarkably, ZnO can be prepared using different fabrication methods into various morphologies such as nanowires, nanobelts, nanorod, nanotubes and nanosheets [20-24]. ZnO nanostructures have a more significant surface-to-volume ratio characteristic that provides more charge trapping sites and results in higher output performance. ZnO nanosheet formations are the most stable among various ZnO nanostructures when subjected to high external mechanical loads due to their geometrically interconnected structure [25]. However, the nanogenerator performance enhancement with nanostructures is challenging due to the complex and unfavourable synthesis technique which makes it difficult for large-scale production and high stability. Synthesizing ZnO nanostructures can be accomplished by a variety of processes such as sol-gel, microwave-assisted method, hydrothermal, chemical vapor deposition and precipitation methods [26-28]. Synthesizing metal oxide nanostructures by the solution-immersion approach has been favored due to low cost and able to synthesize in large scale production [29-34].

While ZnO nanostructures were extensively researched, the heat treatment effects on the TENG performance using a ZnO nanosheet thin film on an aluminium foil substrate deposited through the solution immersion method is not commonly discussed. Therefore, the objectives of this work were to investigate the impact of heat treatment on the surface and structural characteristics of the synthesized ZnO nanosheet thin film deposited on aluminium foil using a solution immersion method. Furthermore, the TENG device using ZnO nanosheet thin film and Kapton tape as triboelectric materials were fabricated to examine the TENG's output performance. The mechanism of TENG device was also discussed.

2. Methodology

The zinc nitrate hexahydrate ($Zn(NO_3)_2 \cdot 6H_2O$) was supplied by Friendemann Schmidt, while the hexamethylenetetramine (HMT) ($C_6H_{12}N_4$) was purchased from Sigma-Aldrich. Both chemicals had a purity level of 99%. The chemicals were used in the synthesis without undergoing any further purification steps. A flexible aluminium (Al) foil which is commonly used for the general purpose was employed as the substrate to deposit ZnO nanosheet thin film. The entire experiment used Millipore's deionized water (DI water).

Synthesizing ZnO nanosheet thin film required the preparation of a mixture of DI water, HMT and zinc nitrate hexahydrate. The molarity of both compounds was set at 0.1 M and diluted with 500 ml DI water. The mixture underwent ultrasonic mixing at 50 °C for several minutes and following with stirring process at 250 rpm for 5 min to facilitate better mixing. Once the solution was mixed well, it was transferred to a Schott bottle and tightly sealed. Then, the Schott bottle underwent heating in a tank of water at 95 °C. The 2×2 cm^2 Al foil substrate was placed inside the Schott bottle before the solution was poured. This immersion procedure took 4 hours to complete. The ZnO nanosheet thin films were taken out from the immersion solution and dried after a cooling period of 30 min. The ZnO nanosheet thin film was further heated at 150 °C for 60 min in an ambient atmosphere to examine the heat treatment impact on the surface morphology and structural characteristics. For the preparation of a ZnO nanosheet thin film-based TENG device, the ZnO nanosheet thin film functions as the positive triboelectric layer, while the Al foil substrate served as the bottom electrode. The Kapton tape was employed as an opposing triboelectric material that had been attached to Al foil, which was the upper electrode. These triboelectric layers were put together with a 2 mm space between them, which works as an air gap.

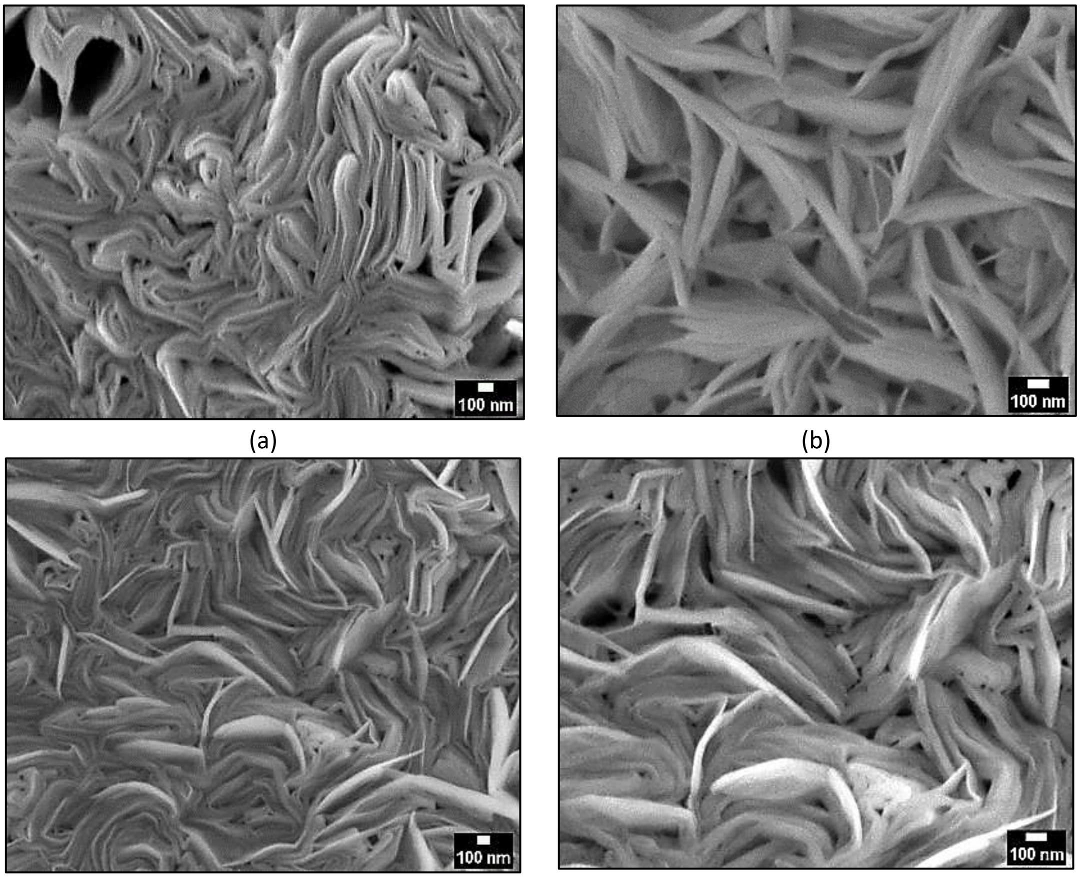
A field emission scanning electron microscope (JEOLJSM-7600F) was used to examine the ZnO nanosheet structures. The sample was placed on carbon tape sealed at the stage holder for the surface morphology examination. The PANalytical X'Pert PRO X-ray diffraction (XRD) instrument with copper K-alpha wavelength, the crystallographic structure properties of ZnO nanosheet thin film were determined. The XRD spectra have been collected at diffraction angles (2θ) ranging from 20 ° to 80 °. For the investigation of the electrical TENG performance, the triboelectric responses were captured using an oscilloscope (Keysight InfiniiVision DSOX2022A), where the applied force using repeated hand tapping and push-pull solenoid tapping.

3. Results and Discussion

3.1 Structural and Morphological Properties of ZnO Nanosheet Thin Film

The formation of an interconnected sheet-like structure in the synthesized ZnO which prepared on an aluminium foil for both before and after heat treatment can be seen clearly in Figure 1. Based on the Figure 1(a) and (b), the linked sheet-like structure was effectively spread across the Al foil substrate. This was accomplished without exposing the film to any heat treatment. Figure 1(c) and (d) demonstrate the appropriately dense and homogeneous nanosheet surface morphology that was achieved after heat treatment. This morphology was associated with fewer voids in comparison to the as-prepared ZnO nanosheet thin film. The produced ZnO nanosheet had an average thickness of 17.3 nm prior to heat treatment and 12.2 nm following heat treatment. The surface morphology of ZnO nanosheet thin film after heat treatment indicates the stronger interaction between nanosheet structures and also increases the surface area. The resulting images clearly demonstrate how the heat treatment significantly affects the development of ZnO nanosheet thin film. The ZnO nanosheet structure is highly useful for usage in TENGs because it provides access to large surface areas that can be put to beneficial use in the generation of charges.

Additionally, XRD measurement was employed in order to investigate the crystallinity of ZnO nanosheet structures that were produced by a simple solution-immersion technique. The XRD pattern that was acquired from the ZnO nanosheet thin films deposited on Al foil is displayed in Figure 2. The prominent peaks on the crystal structures (100), (002), (101), (110), (103), (200), (112), (201), and (004) allow for the identification of the hexagonal wurtzite structure of ZnO, based on JCPDS card no. 01-075-1526 [35]. The diffraction peaks in the aluminium crystal structures are located at 38.6°, 44.8°, 65.2°, and 78.3°, respectively. The (111), (200), (220), and (311) planes are all associated with these peaks (JCPDF No.001–1180) [36]. Extra diffraction peaks are found at 33.6° and 60.1° (denoted by an asterisk [*]) are planes of zinc aluminium, confirming that the ZnO nanosheet thin film had been deposited on aluminium foil [37,38]. The diffraction pattern shows no signs of any other contaminants. Following the heat treatment at 150 °C, it was found that the diffraction peaks' intensities increased. This agrees with the finding revealed by Gaddam *et al.,* [39] suggesting that heat treatment led to the effective crystallisation of ZnO nanostructures.



(a) (b)

(c) (d)

Fig. 1. The surface morphology of the ZnO nanosheet thin film prepared on the Al foil (a) as-prepared ZnO nanosheet at magnifications 30,000X (b) 50,000X, (c) 150°C heat treatment at magnifications 30,000X (d) 50,000X

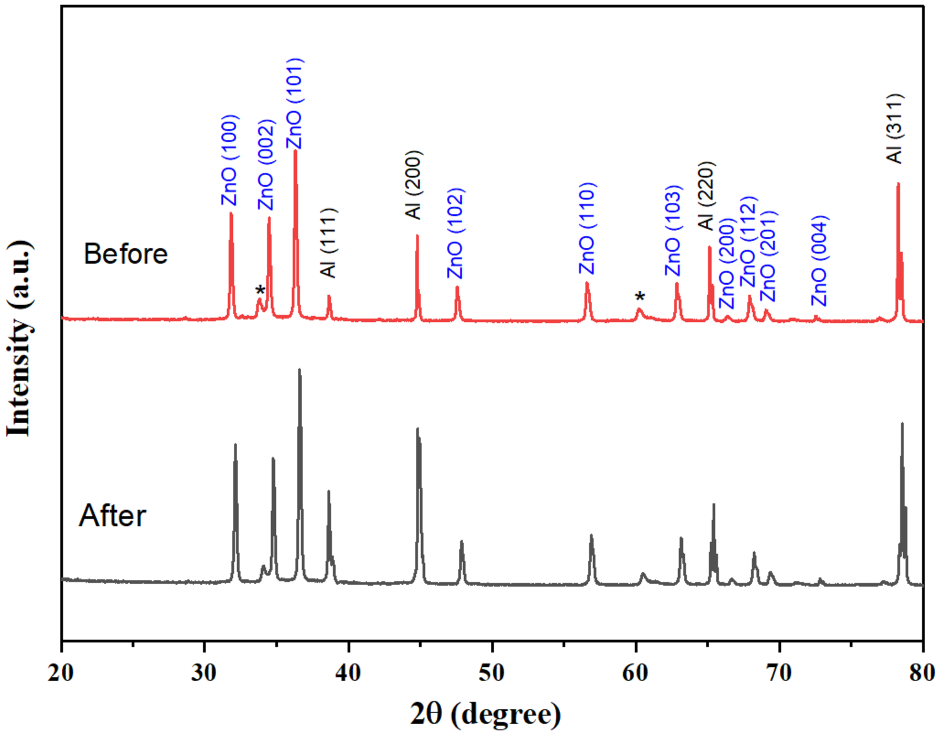
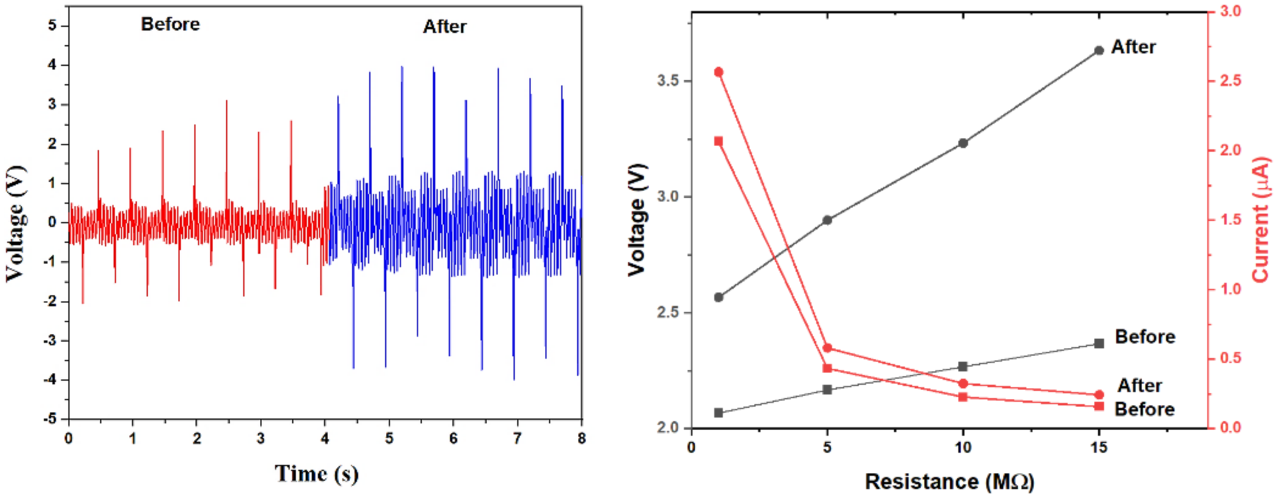


Fig. 2. The x-ray diffraction peaks of the ZnO nanosheet thin film synthesized using Al foil substrate on heat treatment effect

3.2 The Energy Harvesting Performance of ZnO Nanosheet Thin Film based TENG

Figure 3(a) displays the electrical energy produced by TENG based on ZnO nanosheet thin films. In the vertical contact-separation mode, the triboelectric response of ZnO nanosheet thin films TENG was observed. The mechanical impacts exerted on the TENG device result in the formation of positive and negative peaks (AC output signal). The output electrical voltage produced by the ZnO nanosheet thin film based TENG (size = 2 × 2 cm^2) against hand pressing. The average open circuit voltage values for ZnO nanosheet thin film fabricated TENG before and after heat treatment are approximately 2 V and 4 V respectively. Such an enhancement in the output electrical performance of ZnO nanosheet thin film based TENG with the heat treatment might be observed due to the abundance of triboelectric charges accumulating across the high density of interconnected ZnO nanosheet as confirmed by FESEM images. Thus, increased charge-attracting capability and raised the output electrical voltage. In addition, the electrical output with different resistances ranging from 1-15 MΩ was recorded to determine the output power density of the fabricated TENG. As seen in Figure 3(b), the voltage value increased with increasing load resistance. When load resistance increased, the value of the current dropped as predicted by ohm's law. The maximum current is determined using a 1 MΩ resistance and the minimum current is measured using a 15 MΩ resistance.The maximum power density for both ZnO nanosheet thin film based TENG before and after heat treatment was 1.07 $\mu W/cm^2$ and 1.65 $\mu W/cm^2$ respectively.



(a) (b)

Fig. 3. (a) The output electrical voltage of the TENG constructed using ZnO nanosheet thin film for comparison between before and after heat treatment was applied, (b) Voltage and current signal measured with different resistances

An explanation of the voltage generation phenomena in contact-separation mode was presented, and a functioning mechanism for the constructed TENG device was provided, as illustrated in Figure 4. TENG is made up of two dissimilar materials, and each one has an electrode attached to it, as illustrated in Figure 4(a). Materials that produce triboelectric charges often have a low electrical conductivity. To put it another way, they act as insulators. Based on Figure 4(b), the introduction of an external force allows these two dielectric layers to come into contact with one another, which leads to the formation of triboelectric charges on the interfaces of both positive and negative triboelectric layers. The generated charges flow through the upper conductive layer to the bottom conductive layer when the device was connected to an external load due to a potential difference created among the both layers, as depicted in Figure 4(c). When both layers are entirely separated in a certain gap when lifted by an external force, there is no longer any movement of charge at this stage. The difference in potential of these two triboelectric layers was zero (potential drop) as presented in Figure 4(d). At this point, both layers have moved closer together upon the externally applied force, which causes the current to flow oppositely (Figure 4(e)).

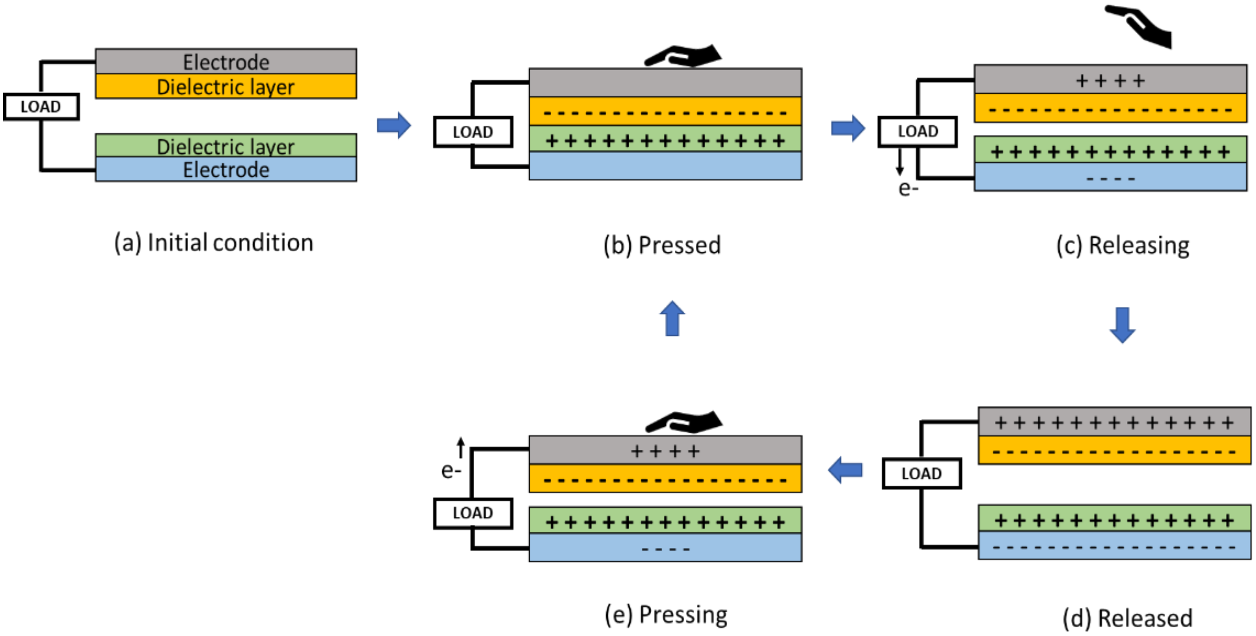


Fig. 4. The TENG device in vertical contact-separation mode functioning mechanism

The output electrical voltage was measured at different tapping solenoid frequencies between 5 Hz and 20 Hz to further examine the frequency responsiveness of the built TENG device. The frequency response of the assembled TENG device was further investigated by measuring the output electrical voltage at varying tapping solenoid frequencies between 5 Hz and 20 Hz. Figure 5 displays the response of TENG under a variety of tapping solenoid frequencies. Up until 10 Hz, an increase in frequency caused the TENG's average output voltage to rise, but after that point, the voltage began to fall as the frequency increased. The incomplete neutralization of charges led to the increase of triboelectric potential which is attributed to the increased output voltage that occurs with an increase in frequency [40]. The open circuit voltage dropped at high frequencies (more than 10 Hz) might be due to the compressed frequency cycle being too high. In that case, the nanogenerator will not be able to recover to its initial location before the subsequent applied force, which will result in a decrease of output voltage [41].

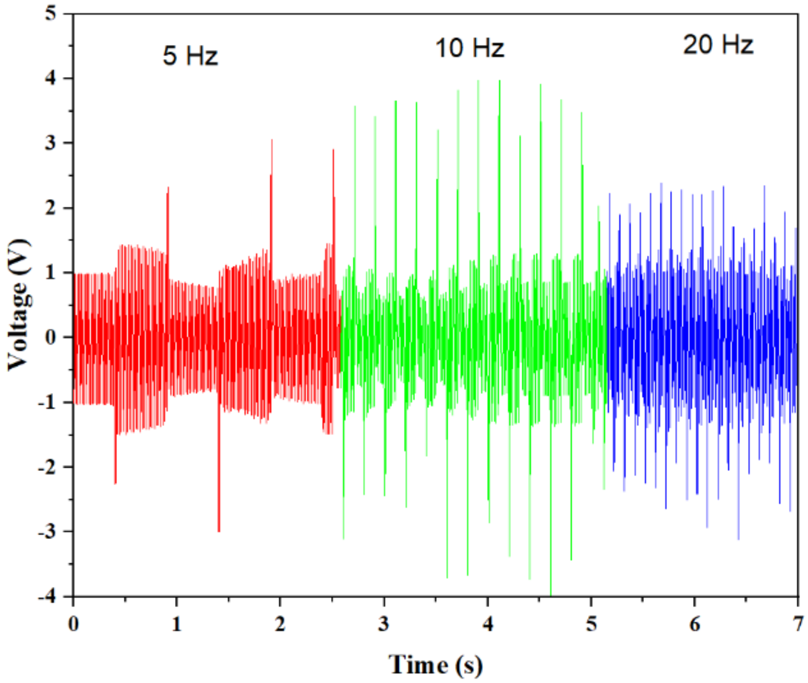


Fig. 5. TENG output electrical voltage with different frequency

4. Conclusions

In conclusion, a thin film of ZnO nanosheets was synthesized on Al foil substrate by a simple solution immersion approach. Heat treatment at 150 °C has an impact on the surface morphological and structural characteristics of ZnO nanosheet thin film. Different surface morphologies were seen for both before and after heat treatment samples. The thickness of ZnO nanosheet structures was decreased after heat treatment, which led to a larger surface area. The XRD analysis demonstrated the successfully deposited ZnO nanosheet thin film on Al foil. Both unheated and heated ZnO thin films were used to create the TENG devices, and a fixed Kapton tape surface served as the opposing triboelectric layer. An increment of the average output electrical voltage of the prepared TENG was obtained from about 2 V to 4 V after heat treatment. The different pushing frequencies additionally impacted the efficient performance of the TENG device. The structural morphology as well as the functionality of TENG were significantly influenced by the heat treatment of ZnO nanosheet thin film. In order to harvest energy that is both affordable and capable of serving as an electrode, this work introduces a new triboelectric layer that uses ZnO nanosheets that have been deposited directly onto Al foil.

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