

Fabrication of ZnO based Nanogenerator for Sustainable Biomechanical Energy Harvesting

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Abstract—Wearable nanogenerators have gained significant attention due to their capacity for transforming energy from bodily movement and the environment around them into electronic signals. These signals can then be used to power portable or wearable electronic gadgets for purposes such as monitoring, tracking movement, or observing, ultimately enhancing the overall quality of human life. A p-n junction ZnO textile nanogenerator is created on conductive textiles using PEDOT:PSS/CuSCN-coated ZnO nanorods. This combines the benefits of ZnO piezoelectricity with textile flexibility. It has been discovered that as the length of ZnO nanorods is increased, the nanogenerators demonstrate a corresponding increase in output voltage and power density. The device, equipped with ZnO nanorods of optimized length, exhibits a progressive rise in output voltage when the shaking frequency is raised. This increase in voltage is sufficient to power an LCD screen display. Furthermore, the gadget has the capability to produce a progressively higher negative voltage ranging various with the application of an increasing impacting force. The manufactured gadget may also capture other mechanical forces, such as flicking, light finger bending, and tapping. These findings are especially significant for the development of upcoming adaptable self-sustaining gadgets and portable electronics.

Index Terms— *Nanogenerator, energy harvesting, sustainable, and biomechanical.*

I. INTRODUCTION

Flexible electronics is a developing field of study that seeks to improve the durability of micro electromechanical systems (MEMS) components, including sensors, micro actuators, light-emitting diodes (LEDs), and accelerometers. The versatility and mobility of such gadgets are determined by the electrical power they need. A self-powered gadget offers a viable option for achieving great flexibility and mobility at a reasonable cost. Several studies have demonstrated several techniques of generating self-

sustaining electricity from waste energy found in the environment [1]. This involves the conversion of mechanical energy, auditory energy, wind energy, human motion, and thermal energy into electrical energy. Mechanical stimuli, such as stress, extension, movement, and tension, are plentiful in the surrounding environment. The piezoelectric nanogenerator (PENG) and triboelectric nanogenerators have been used as very promising devices for harnessing these energies [2].

PENG devices are used in real-time MEMS systems because of their intrinsic characteristics, including increased voltage, biocompatibility, and improved longevity. In addition, much research has been conducted on many applications of sensors and energy harvesting, including self-powered force sensors, monitoring systems, and energy generation from internal organ vibrations. In addition, there is a diverse range of commercially accessible piezoelectric materials, including PZT, ZnO, ZnS, BaTiO₃, and PVDF [3]. ZnO based piezoelectric materials have attracted significant attention owing to their distinctive characteristics, including a non-centrosymmetric structure, cheap manufacturing cost, and environmental friendliness. In order to enhance the efficiency of pure ZnO devices, the process of introducing III-IV group elements (B, Al, Ga, In, Pb, Sn) by doping has been used. Out of these dopants, Sn is more compatible with Zn since their ionic radii are near (Sn⁴⁺ - 0.69 Å & Zn²⁺ - 0.74 Å). This allows Sn to effortlessly integrate into the lattice with minimum deformation and increases the concentration of electron carriers [4].

Several researches have documented the physical and electrical properties of Sn-doped ZnO materials. In addition, there is a diverse range of commercially accessible piezoelectric materials, including PZT, ZnO, ZnS, BaTiO₃, and PVDF. ZnO based

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When choosing piezoelectric materials for constructing wearable or implantable energy harvesters, ZnO is particularly notable due to its many benefits, such as its high piezoelectricity, tunability, ease of production, availability, and cheap cost. ZnO is renowned for its superior biocompatibility in comparison to other often used piezoelectric ceramics including PZT, PZN-PT, and PMN-PT. While these ceramics have high d_{33} values, they are poisonous due to the presence of lead (Pb) [7].

Therefore, the later ones are not biocompatible for constructing wearable or implantable energy harvesters. Zinc oxide (ZnO) has been used in several forms for the development of energy harvesters with the assistance of nanofabrication techniques. These forms include ZnO nanoarrays, nanoparticles, and metal-doped ZnO [8]. The exceptional piezoelectric performance of ZnO nanoarray has garnered significant interest in recent years. Aligning ZnO nanorods along the c-axis of the ZnO crystalline structure leads to a substantial increase in overall electrical energy output. This is due to the cumulative effect of electrical generation from each nanorod. Nevertheless, despite the existing technologies in the development of energy harvesters using ZnO nanoarrays, there are still three significant obstacles that hinder the widespread use of ZnO nanoarrays in constructing flexible energy harvesters. The first method involves the intricate and indirect production of ZnO nanoarrays on a flexible substrate. This is achieved by first growing the ZnO nanoarrays on a rigid substrate, such as a silicon (Si) wafer or a silica substrate, and then transferring them onto a more pliable substrate.

Another difficulty is that many flexible substrates that are described are not sufficiently flexible to create wearable devices that are pleasant, since they have a substantial disparity in mechanical properties compared to human soft tissues, such as skin tissues. Common substrates used for growing ZnO nanoarray, such as polyethylene terephthalate (PET), polyethylene naphthalate, polyethersulfone, Kapton film, and paper, have mechanical properties that differ significantly from human skin. As a result, they are not suitable for comfortable use on the human body. The third problem emerges from the electrodes of devices that lack the ability to stretch. Frequently used electrodes for ZnO nanoarray consist of platinum (Pt), copper (Cu), and indium tin oxide (ITO), which possess excellent conductivity and are typically deposited onto a polymer film using a sputtering technique. Nevertheless, these electrodes made of metal are very susceptible to fracturing when subjected to stretching, resulting in a significant reduction or entire loss of their favourable conductivity [9].

To tackle these three critical issues, we have created a versatile, elastic, and compatible piezoelectric energy harvester utilising a simple manufacturing approach. The novel manufacturing technique enables the direct growth and integration of ZnO nanoarrays into a polydimethylsiloxane (PDMS) matrix, together with carbon nanotube (CNT) electrodes, without requiring annealing or polling of the material. The gadget, which has a flexible nature and an elastic modulus of 3.3 MPa, has the capability to be extended to 250% of its initial length. The energy harvester, when used as a wearable device and subjected to various movements, may generate an open-circuit, peak-to-peak voltage of up to 9.2 V. Finally, the energy harvester is integrated into a leadless pacemaker (LP) and surgically implanted into a live porcine right ventricle. Its purpose is to gather energy from the heart's contractions and transform the mechanical energy into electrical energy. The gadget demonstrated potential electrohydraulic (EH) capacity during ex vivo testing utilizing a customised cardiac simulation system. The technology suggested in this study addresses the three issues stated earlier and offers a straightforward and efficient approach for creating energy harvesters that possess sufficient stretchability, flexibility, and biocompatibility for prospective use in wearable and implantable applications.

Rest of the paper is organized as; Section 2 summarizes the literature survey followed with the proposed framework is arranged in Section 3. Section 4 describes the experimental study and the paper is concluded in Section 5.

II. RELATED WORKS:

For harvesting biomechanical energy, Paydari et al. [10] suggested embedded ZnO particles-based composite. The study examines the effect of ZnO particles of different shapes that are enclosed in a PDMS matrix on the performance of hybrid nanogenerators. The XRD examination reveals that pyramid-shaped ZnO particles have a higher density of crystals aligned along the c-axis, which corresponds to the direction of the piezoelectric action in ZnO material. The scanning electron microscopy (SEM) findings indicate that the particles may be categorized in terms of size from largest to smallest as follows: rod-shaped, pyramid-shaped, and ellipse-shaped. The pyramid and ellipse-shaped ZnO particle/PDMS composite hybrid nanogenerators provide a maximum open-circuit voltage of 161 V and a short-circuit current of 29.8 μ A. The highest power density achieved by the hybrid nanogenerators made of ZnO particles embedded in a PDMS composite is approximately 1.04 Wm^{-2} for the ellipse shape, 0.75 Wm^{-2} for the rod shape, and 0.6 Wm^{-2} for the pyramid shape. These power densities were observed when a load resistance of 10 M Ω was used.

For biothermal and biomechanical monitoring, Kim et al. [11] presented wearable fabric-based ZnO nanogenerator. This study introduces a fabric-based ZnO nanogenerator that allows air to pass through and has the ability to adapt to various deformations. The purpose of this nanogenerator is to be used in wearable piezoelectric sensors for gathering biomechanical health data. We synthesize ZnO nanorods uniformly distributed across the whole nylon fabric, with a strategically located neutral mechanical plane, specifically designed for the purpose of creating bend-sensitive electronics with a current sensitivity of 2.59 μ A mm. The hierarchically interlocked shape of the object allows for precise tactile sensing, with a sensitivity of 0.15 nA kPa $^{-1}$. Skin-mounted sensors are used to get various physiological information regarding activities, such as pulse rate, respiration, swallowing, and coughing. Additionally, the potential

of a device coupled to a mask to detect and analyze certain respiration patterns by pyroelectric sensing is shown. The wearable healthcare sensors we have developed show significant potential for continuously monitoring vital indicators connected to health. This allows people to be informed about their health condition in real-time, without any interference in their everyday routines.

Based on flexible substrates, the solution-processed ZnO energy harvester devices was presented by Martinez-Lopez et al. [12]. Renewable energy harvesting adaptable gadgets provide an alternative technology that can progressively replace electrochemical batteries. Provide a straightforward and cost-effective method for producing flexible piezoelectric energy harvesters using ZnO nanoparticles. These devices have the capability to transfer vibrational energy from green sources into electrical energy, and can be fabricated at low temperatures. The ZnO nanoparticles are applied onto a substrate made of ITO (indium-tin-oxide) and PET (polyethylene terephthalate). The manufactured harvester device exhibits a maximum open-circuit output peak voltage and root mean square (rms) voltage of about 15 mV and 2.5 mV, respectively, when subjected to mechanical vibration at a frequency of 14 Hz. This manufacturing approach enables the creation of piezoelectric energy harvesters that are both environmentally friendly and cost-effective. These harvesters have flexible substrates, are lightweight, have a simple mechanical construction, and do not need complicated performance.

For self-powered sensing and biomechanical energy harvesting, Wang et al. [13] presented sustainable triboelectric nanogenerators. This article provides an overview of the latest advancements in TENGs (Triboelectric Nanogenerators) that are constructed using recycled materials, known as RM-TENGs. It covers the many aspects of RM-TENGs, including their design, manufacturing, assessment, and application. Initially, an examination is conducted on the existing recycling techniques used for recycled materials, followed by a concise explanation of the fundamental protocols now in use. Next, this article provides a thorough examination and analysis of the energy harvesting and sensing applications of RM-TENGs, focusing specifically on several categories of

recycled materials. Furthermore, their prospective applications are also shown. Ultimately, we provide insights into the difficulties and possible remedies linked to the future recycled-material based TENGs. Utilizing recycled materials for the production of TENGs may overcome the current constraints imposed by the materials used in TENGs. Additionally, this approach can significantly enhance sustainability by creating sustainable and renewable energy from waste materials.

The study conducted by Rana et al. [14] introduces a piezoelectric nanogenerator made of a polymer nanocomposite film for the purpose of biomechanical energy harvesting and motion monitoring. We have developed a flexible piezoelectric nanogenerator (PNG) device using a polyvinyl difluoride (PVDF) polymer. This device can effectively capture energy from different human movements and transform it into usable electrical energy. In order to boost the performance of the PVDF based nanogenerator, we include hydrothermally produced nanosheets of reduced graphene oxide (rGO) and boron doped rGO into the PVDF matrix as conductive nanofiller materials. This addition improves the output performance of the device. Out of all the devices made using pure PVDF (P), PVDF doped with reduced graphene oxide (PR), and reduced graphene oxide doped with boron (PBR), the PBR device produces the highest voltage and power density, measuring 13.8 V and about 42.3 $\mu\text{W}/\text{cm}^2$ respectively. This power is then utilized to illuminate a series of commercially available LEDs. Ultimately, the use of PBR film in PNG is shown as a means to extract energy from many forms of human movement, such as finger tapping, elbow bending, foot tapping, leg folding, and wrist motions. This gadget showcases the possible use of polymer nanocomposite films in self-powered wearable devices.

Jiang et al. [15] introduced Metal-doped Functional Zinc Oxide Nanosheets for the purpose of Self-powered Mechanical Sensing. The stability and durability needs of nanogenerators after continuous operation still pose challenges in terms of fabrication processes and expanding sensory output. This study involves the preparation, characterization, and complete embedding of metal-doped zinc oxide nanomaterial-polymer composites onto flexible

substrates. The aim is to create piezoelectric sensors that have improved output performance and sensing capacities. When an applied force of 8 N is exerted on the flexible device at a frequency of 3 Hz, it is capable of producing an open-circuit voltage of up to 13.6 V. Furthermore, these composite devices that are devoid of lead have exceptional sensitivity in detecting elbow motion, which presents a significant opportunity for accurately monitoring human body movements. In addition, a biomimetic soft robotic fin equipped with mechanosensors has been created, allowing for the measurement of both the amplitude and frequency of the fin ray's flapping. These discoveries provide fresh insights into the production of efficient piezoelectric nanogenerators and provide the groundwork for enhancing the ambient sensing abilities of underwater species or robots.

III. METHODS

3.1 Synthesis ZnO based on Sn

The ZnO nanostructure was grown using Zinc nitrate hexahydrate [$\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, 98%, Sigma Aldrich] and Hexamethylenetetramine (HMTA) [$\text{C}_6\text{H}_{12}\text{N}_4$, 99%, Sigma Aldrich]. Tin Chloride II [$\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$, 98%, Sigma Aldrich] was used as a dopant. A composite thin film was created using polyvinyl alcohol (PVA) as a binding agent, specifically obtained from Tokyo Chemical Industries. PVA is a polymer with the chemical formula $(\text{C}_2\text{H}_4\text{O})_n$.

3.2 ZnO Powder synthesis

The hydrothermal (or solvothermal) approach is a valuable technique for synthesising one-dimensional nanostructures, including nanowires, nano needles, flowers, and nanorods. In this process, a wet chemical reaction occurs when the precursors are dissolved in water or organic solvents at temperatures ranging from 50°C to 250°C. This approach is very effective in synthesising ZnO nanostructures, resulting in an increased presence of crystalline defects. These flaws, in turn, lead to a higher number of oxygen vacancies, ultimately enhancing the optical and electrical characteristics of the ZnO nanostructures. The hydrothermal approach is recognised for its ability to provide a consistent dispersion via co-precipitation. In addition, the hydrothermal approach occurs at moderate temperatures, preventing phase transition in

comparison to other procedures. The precursor for the ZnO solution was obtained by using 50 mM of Zinc nitrate hexahydrate salt. The substance was dissolved in 60 ml of deionized (DI) water and agitated vigorously for 30 minutes until a clear and uniform solution was achieved.

While stirring, HMTA was introduced to maintain a pH of 7 by raising the concentration of OH⁻ ions and causing the precipitation of Zn²⁺ ions. For doping purposes, SnCl₂ flakes were added to the Zn precursor salt at various concentrations of Sn (2.5%, 5%, 7.5%, and 10%). Subsequently, the addition of HMTA during the stirring process resulted in the transformation of the doped solution into a milky color. Following agitation, the solution was then placed in a high-temperature oven for duration of 6 hours at a temperature of 95 degrees Celsius. The precipitate that was generated was purified by many rounds of centrifugation using deionized water. The accumulated precipitation was subjected to a drying process at a temperature of 90 degrees Celsius for a few hours. Figure 1 depicts the schematic diagram illustrating the process of material synthesis and gadget construction.

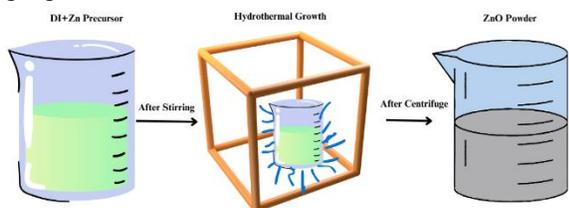


Fig 1: ZnO Powder synthesis and film deposition

3.3 Designing of paper based PENG

A 2.5 cm X 2.5 cm photographic paper sheet was employed as a substrate for the piezoelectric device. The carbon conductive ink was applied to the device as both the bottom and top electrodes using the bristle coating technique. The electrode was coated in four layers to guarantee the quality and consistency of the film. Following the application of a coating, the electrode was subjected to ambient conditions for drying. Simultaneously, 5 grammes of PVA salt were dissolved in 100 millilitres of deionized water and vigorously agitated for 2 hours at a temperature of 95 degrees Celsius. The result of this process is a clear and thick solution, with no breakdown of the polymer due to heat. The ZnO/ZnO+Sn powder, as it was originally synthesised, was combined with a viscous PVA solution at a ratio of 10:1, with the powder

measured in milligrammes and the solution measured in millilitres. The mixture was deposited by drop casting to create the film and then subjected to a curing process at a temperature of 90 degrees Celsius for duration of one hour. The connections were made by employing 0.1 mm copper wire with silver paste, which was obtained from the top and bottom electrode. The piezoelectricity of ZnO nanoarray is a result of the disruption of the initially harmonious crystalline structure of each ZnO nanorod, which has an initial neutral overall charge. While a stress or strain is applied to the wurtzite structure of a ZnO nanorod, each individual crystal unit gets deformed and generates an electrical dipole, resulting in an electrical potential difference across the nanorod.

Due to the alignment of all nanorods along the electrode-electrode direction of this device, opposing charges will collect on the electrodes, resulting in an overall electrical potential throughout the whole device. An assessment is conducted to evaluate the piezoelectric performance of the device. More precisely, a traditional impact test is used to measure the voltage output of the device as it is made, based on the amount and frequency of the applied force. A shaker is used to provide a controlled average force on a device placed on a vertical wall, with the amplitude and frequency of the force being regulated. Subsequently, the gadget was enveloped with anti-static tape to avert the occurrence of electrostatic discharge. Figure 2 displays the schematic of the developed PENG gadget.

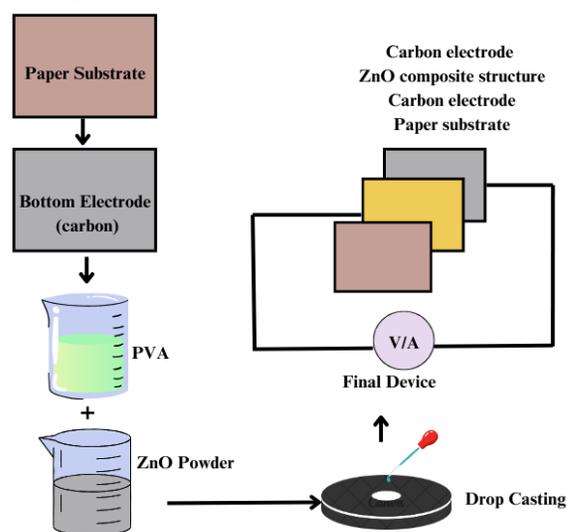


Fig 2: Developed PENG gadget

3.4 Characterization

The field emission scanning electron microscope (FESEM, Carl Zeiss Sigma series supra-55) was used to analyse the surface morphology and energy dispersive x-ray spectroscopy (EDS) of both doped and virgin powders. The crystal structure and phase pattern were assessed using powder X-ray diffraction (XRD) analysis, specifically with 2 theta values ranging from 30 degrees to 70 degrees (measured using the Rigaku Smart Lab system). The Raman spectra were used to investigate the lattice defects and vibrational modes of both doped and clean materials within the wavenumber range of 300 to 800 cm^{-1} , using an excitation wavelength of 514 nm (LabRAM HR Evolution; Horiba, Japan). The piezo output efficiency and I-V characteristics were assessed using a Keithley 2612B source metre.

IV. DISCUSSION

The concentration of Sn doping was tuned with respect to the piezoelectric performance. We have shown that the piezoelectric response improves up to a Sn doping concentration of 2.5%, after which performance loss occurs for Sn doping concentrations more than 2.5%. Thus, the impact of increased doping concentrations of PENG devices has not been addressed in this work.

4.1 Morphology of surface

Figure 3 displays the FESEM pictures and Energy Dispersive X-ray Spectroscopy (EDS) graphs of both Sn doped and pure ZnO nanostructures. The presence of Zn and Sn was verified using EDS characterization. The produced ZnO nanoparticles were found to have achieved a hexagonal nanorod form. However, the presence of Sn ions in ZnO resulted in a noticeable change in the structure of the material, namely in the form of hexagonal nanorods. The nanorods' diameter has augmented according to the rise in dopant concentration. The enlargement in diameter of the ZnO nanorods may be attributed to the infiltration of Sn^{4+} ions into the vacant areas inside the crystal structure of ZnO. In addition, the research from the literature [63 and references therein] indicated that there is a rise in the interspaces as the doping level increases. The ImageJ program was used to determine the diameter of nanorods. The pure ZnO had a diameter of 185 nm, whereas the 2.5% and 5% Sn doped ZnO had diameters of 230 nm and 250 nm, respectively.

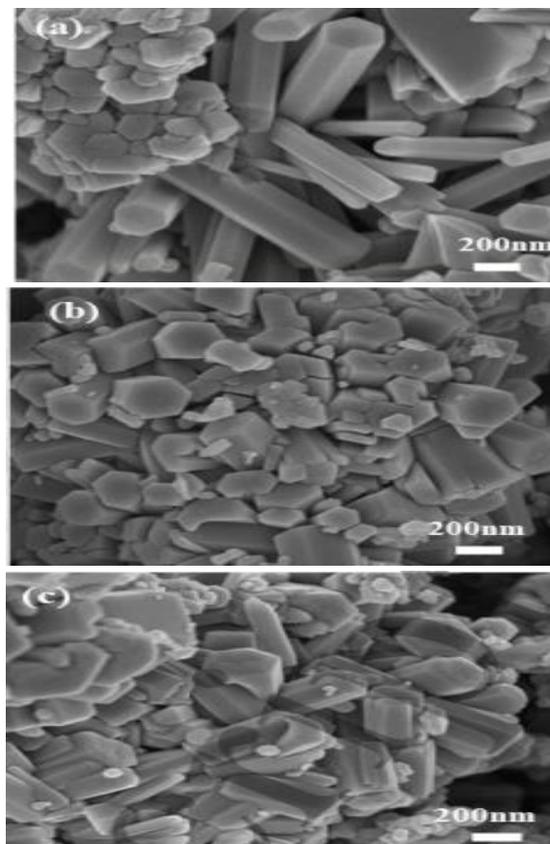


Fig 3: The FESEM images

4.1 Output response of Piezoelectric

Figure 4 shows the output response of Piezoelectric. To produce the desired sound, the musical drums must be struck with specified patterns at varying intensities. In this case, the device has been subjected to unequal hand stamping with a weight ranging from 4 N to 22 N. A load resistance of 940 mega ohms was used in the circuit to determine the voltage when no current is flowing. The irregular hand-stamping applies compressive stress on the device. Stress causes a distortion in the symmetry of the ZnO lattice, resulting in the creation of a dipole. The presence of a non-net zero dipole moment results in the generation of potential across the device. The lack of balance in the charge center allows the electron to go from the surface top electrode to the bottom electrode, resulting in the generation of a positive potential peak during measurement. When the external tension is removed from the gadget, the dipole moment disappears. Subsequently, the electron returns to its original location, resulting in the generation of a negative potential peak. The primary positive and negative maximum potential.

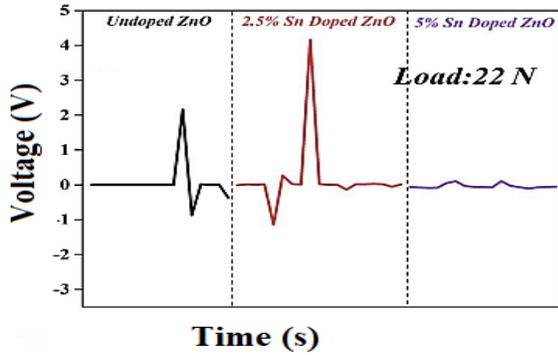


Fig 4: Piezoelectric output response

4.2 Power based on piezoelectric

The piezoelectric nanogenerator, which operates on paper, has been extensively used as a sensor or self-powering system in MEMS applications. The suggested application is designed to effectively operate under various hand stamping conditions to collect piezoelectric power using a paper PENG device, similar to other devices used for power production. Greater force results in increased lattice distortion via the strain phenomenon, which in turn provides greater power. Figure 5 displays the highest achievable voltage and current values at various levels of Sn doping concentration, corresponding to a maximum force of 22 N. The device with 2.5% tin-doped zinc oxide (Sn-doped ZnO) has achieved the highest voltage (4.15 V) and current (36 nA) compared to other devices with Sn-doped ZnO. The increased output voltage seen in the 2.5% Sn doped ZnO device might be attributed to the decrease in electrical conductivity or the increase in resistivity. Put simply, more resistivity results in a greater potential drop. The decrease in conductivity for electricity may be attributed to the heightened strain in the microscopic structure of ZnO, which subsequently diminishes the speed of electrons.

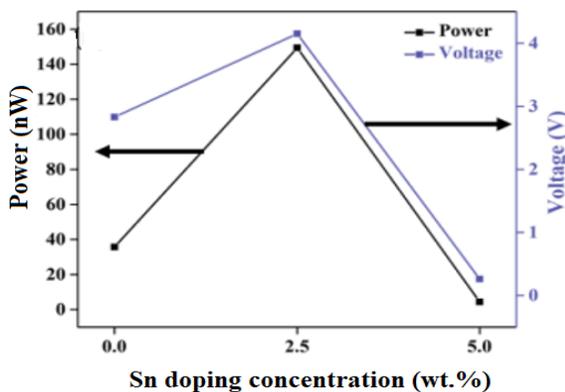


Fig 5: Piezoelectric output response

Figure 6 plots the various force impacts. The PEDOT:PSS/CuSCN/ZnO architecture's adaptability is shown by its ability to be generated on Ag-plated polyester, a different conductive textile, therefore creating a p-n junction-based ZnO textile nanogenerator architecture. The application of the ZnO seeding layer and CuSCN coat is influenced by the various surface characteristics of nylon and the way of fiber weaving. Therefore, multiple optimal accumulation methodologies were used to create CuSCN/ZnO on Ag-plated nylon material.

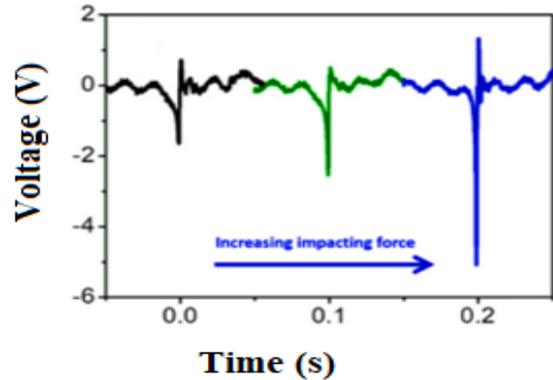


Fig 6: Various force impacts

V. CONCLUSION

Ultimately, wearable nanogenerators made from textile-based PEDOT:PSS/CuSCN/ZnO have been effectively created by producing ZnO nanorods on conductive textiles. We conducted a thorough investigation on the impact of varying lengths of ZnO nanorods on the performance of nanogenerators. Our findings indicate that longer ZnO nanorods have the ability to generate a greater piezoelectric potential, resulting in enhanced energy harvesting capabilities. The device, which incorporates the benefits of CuSCN and PEDOT: PSS to minimize the screening effect, demonstrates the capability to harness energy from various types of mechanical stimuli such as shaking at different frequencies, impacts with varying intensities, flicking, moderate finger bending, and tapping. The nanogenerator has the capability to produce an output voltage of around 5V, an average power density of 0.67 $\mu\text{W}/\text{cm}^2$, and it demonstrates consistent performance even after undergoing different cycles of testing. An LCD panel was powered using the output power of the nanogenerator to showcase its practical use. Furthermore, the adaptability of the PEDOT:PSS/CuSCN/ZnO structure was shown by the

creation of a device on Ag-plated nylon textile, which likewise exhibits the capability to collect energy. The current study showcases a textile-based energy harvester that is both adaptable and robust. The manufacturing process described is straightforward and can be easily scaled up. This technology has great promise for future use in self-powered portable and wearable electronic devices.

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