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## Worm structure piezoelectric energy harvester using ionotropic gelation of barium titanate-calcium alginate composite

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### ABSTRACT

A laterally aligned flexible composite linear worm-based piezoelectric energy harvester made up of piezoelectric barium titanate nanoparticles and a three dimensional gel network of calcium alginate biopolymer was aimed to harness the low frequency mechanical energy. It is highly desirable to fabricate innovative micro/nanostructures for high performance energy harvesting beyond the conventional thin films, and small scale fabrication of nanowires (or rods). The open circuit voltage of a single composite worm-based energy harvester (diameter  $\approx 550 \mu\text{m}$ , length  $\approx 2.5 \text{ cm}$ ) increases up to 5 times by increasing the frequency of mechanical load (11 N) from 3 to 20 Hz. Similarly, 1.5 times voltage increment was observed by increasing the length of the composite worm from 1.5 to 3.5 cm upon the bio-mechanical hand force. The energy harvester can function as an efficient portable/wearable self-powered device due to its good flexibility, and multiple lengths of composite linear worms can be utilized to drive low-power electronic devices. In this work, the composite worms were prepared by an ionotropic gelation approach, which is eco-friendly, non-toxic, having low processing temperature/time, and potential for cost-effective, large-scale fabrication, making it suitable for low frequency based self-powered devices.

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### 1. Introduction

Portable and wearable electronics are versatile technologies that may enable future generations to scavenge energy from our daily life environment such as the human body motions, fluidics, and mechanical vibrations. Recently, many energy harvesting (EH) technologies have evolved such as thermoelectric generator (THG), hydroelectric generator (HEG), triboelectric nanogenerator (TNG) and piezoelectric nanogenerator (PNG) devices for portable/wearable applications. Studies show that, a glass fabric based THG developed using the screen-printing technique and it generates high power density [1]. An efficiency of 1.53% was generated using the THG with eight pairs of segmented legs and the corresponding materials were synthesized by spark plasma sintering [2]. A rational

design of multi-unit HEG was fabricated to harvest the water related energy in the environment [3]. Over past decade, rapid and continuous development of PNG and TNG based devices took place, which can convert mechanical energy to electrical energy. Wang et al. [4] developed the basic conventional TNG device structures and it generates high electric power based on four different working modes such as vertical contact-separation mode, lateral sliding mode, single electrode mode and free-standing triboelectric-layer mode. Arunkumar et al. [5] reported the lightweight, cost-effective single-electrode based smart seat TNG device for harvesting energy from the living environment. But these devices suffer from the complicated device design; interference, stability and output performance depend on the growth of various nanostructures (NS) on active triboelectric layers [6]. On the other side, PNG devices were developed using 1D-NS (one-dimensional nanostructures), nanowall, micro belts and thin films, which are useful to harness low frequency mechanical energy. Zhu et al. [7] studied flexible PNG device using sweeping-printing method and

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the active layer used for energy generation is horizontally aligned zinc oxide (ZnO) nanowires (NWs). Wang et al. [8] reported the conversion efficiency of the NWs based PNG device is estimated to be 17%–30%. Yang et al. [9] showed the possibility of energy generation from the single lead zirconate titanate ( $\text{Pb}(\text{ZrTi})\text{O}_3$  designated as PZT) wire and its self-powered temperature sensor application. Kumar et al. [10] developed the biocompatible composite PNG for self-powered ultraviolet (UV) photosensor application using the ZnO NWs and the reduced graphene oxide (rGO). It reveals that, the fabricated PNG can generate an output voltage and current of 5.5 V and 0.63  $\mu\text{A}$ , during the foot-stamp operation. They also synthesized the ZnO nanowall for energy generation using the cost-effective hydrothermal method [11]. Wu et al. [12] firstly demonstrated the single-zinc stannate ( $\text{ZnSnO}_3$ ) micro belt nanogenerator and it has an energy conversion efficiency of 4.2–6.6% based on the strain 0.8–1% respectively. All these conventional PNGs use various types of ZnO NS and generate the small output power (nW to  $\mu\text{W}$ ). This can be useful to drive the low power light emitting diodes (LEDs) and monochrome liquid crystal displays (LCDs). Later, the fabrication of PNG devices were extended using the flexible organic polymers, inorganic perovskite NS and organic-inorganic composite films. Pi et al. [13] studied the organic polymer based PNG device and generates a current density  $\approx 0.56 \mu\text{Acm}^{-2}$ . Kim et al. [14] fabricated inorganic nanoparticles based nanocomposite thin films using layer-by-layer approach for PNG device. Ni et al. [15] reported the single barium titanate ( $\text{BaTiO}_3$  designated as BTO) nanowire based PNG device and the electrical output is proportional to the strain and strain rate. Kim et al. [16] studied the ferroelectric properties of the organic/inorganic composite films with many number of nanolayers and suggest the suitability for resistive switching memory and energy generation applications. Alluri et al. [17] developed the zirconium-doped barium titanate (BTZO) nanocubes/poly (vinylidene fluoride) (PVDF) composite films and the generated output voltage  $\approx 11.95$  and current  $\approx 1.35 \mu\text{A}$  respectively. The above mentioned polymer and composite PNGs generates the output power density from  $\mu\text{W}$  to  $\text{mW}$  range. Over the last decade, many researchers demonstrated that the PNG devices can function as a standalone power source and also have a capability to drive various sensors such as UV photo detector output controlled with different wavelengths of light source [10], fluid velocity sensor to measure the different water speeds [17], active gas sensor to detect oxygen, water vapor and hydrogen sulfide ( $\text{H}_2\text{S}$ ) gases [18], pH sensor to measure different pH levels of buffer solution [19], composite worm structure output controlled by different pH values of buffer solution [20], and glucose sensor to measure different glucose concentrations of human saliva [21]. These self-powered sensors do not require any external power source (battery) and an additional sensory circuit. Among these EH technologies, PNG device is the most reliable and prominent technology. Other EH technologies may have greater energy conversion efficiencies but suffer from the inherent limitations such as leakage currents, difficulties in maintenance, aging effects (performance degradation), complex working mechanisms, pressure and humidity effects, respectively [6]. In addition, many existing portable devices use batteries as electrical driving source, which consists of complex, sensitive materials with limited life time [22].

The potential use of PNG device is not only limited to the conversion of regular mechanical vibration energy, but also extended to convert the ocean waves, wind, rain drop, vehicle suspension motion and tap water motions to electrical energy. Viet et al. [23] developed floating energy harvester to harness the energy from intermediate and deep water waves. Wu et al. [24] suggested the piezoelectric patches on cantilever and a proof of mass is enough to harvest the wind energy. Alluri et al. [17] explore the feasibility of

flexible PNG using the zirconium-doped barium titanate (BTZO) nanocubes/poly (vinylidene fluoride) (PVDF) film can utilize the tap water motion to harvest the electric power. This device can also have the functionality to sense or generate energy with various speeds of water flow ON/OFF conditions. Ilyas et al. [25] developed an alternating approach for harvesting energy with piezoelectric materials by utilizing rain drop impacts. They thought that the efficiency of the device can be improved by modifying the droplet impact mechanism with the harvester surface by exploring new surface materials to maximize inelastic collision. Xie et al. [26] designed an efficient dual-mass piezoelectric bar harvester, which can convert ambient vibrations of a vehicle suspension system subjected to roughness of road surfaces to electrical energy. They stated the construction and installation of multi-unit piezoelectric bar harvesters in vehicle can generate more energy and have significant impact on automobile industry. Azizi et al. [27] studied the dynamics of a piezoelectric bimorph cantilever energy harvesting device with respect to the harmonic base excitations. They stated that the high output power can be possible to generate at the base excitation frequency coalesced with the first natural frequency of the device. Furthermore studies show that the piezoelectric EH devices are highly efficient to utilize and convert the regular natural contractile and relaxation motions of the heart, lung and diaphragm. In addition, many studies demonstrate the output of PNG devices is also useful to drive pressure sensor and motion sensor, respectively. Chun et al. [28] reported ZnO embossed hollow hemispheres thin film for highly responsive pressure sensors and PNGs. They stated for one piece of hemi-sphere layer is stacked over another to form a layer-by-layer matched architecture and the PNG output increases up to 2 times. They also suggested that the stretchable composite films with hemisphere structure based PNG device useful to sense the directional motion of the human body parts [29].

The piezoelectric coefficient ( $d_{33}$  or  $d_{31}$ ), electro-mechanical coefficient ( $k$ ) and relative permittivity ( $\epsilon_r$ ) of NS will play a key role for efficient energy conversion. Initially, conventional PNG devices fabricated by lower  $d_{33}$  of ZnO NS can generate small output power. Further, the PNG fabrication and output power improved by high  $d_{33}$  nanomaterials such as PZT, BTO sodium potassium niobate ( $\text{KNaNbO}_3$  designated as KNN), and flexible PVDF polymer, respectively. PZT based materials and devices produce high performance [30] but facing global restriction due to lead toxicity and environmental pollution. In this case, KNN and BTO are alternative biocompatible materials with reasonable energy conversion efficiency. To date, the progress on KNN based PNG devices generates medium range outputs [31] and have serious drawbacks such as densification, manufacturing cost [32]. Hong et al. [33] suggest that the KNN material has poor piezoelectric properties at room temperature, which is another demerit for KNN. Shrout et al. [34] demonstrated that the KNN properties are strongly temperature dependent, while degradation occurs through thermal cycling between two ferroelectric states. Certainly, BTO and doped BTO based devices are eco-friendly, cost-effective and generate higher or equal to the PZT device output performance. These PNG devices are suitable for self-powered biosensors and implantable devices in health monitoring applications. The main disadvantage of pure BTO nanomaterials is brittle nature and the devices cannot withstand at high mechanical forces, which can be solved by the composite technology [35]. Recent studies show the composite PNG devices have greater energy conversion efficiency, reducing internal leakage currents and can sustain at high input mechanical forces. These PNG devices are made up of inorganic piezoelectric NS as a filler material along with the organic polymer matrix. Another serious factor for approaching the composite technique is manufacturing cost, which depends on the fabrication of active

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