A comparative study of ZnO nanogenerator based piezo in series and parallel as green energy harvester

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Abstract. This paper reports the performance of the flexible nanogenerator as a green energy harvester depends on connection variation. The ZnO nanogenerator was successfully fabricated on Al/PET. The ZnO Nanorods were synthesized using a hydrothermal method at 95°C. The ZnO Nanorods were characterized using x-ray diffraction (XRD), scanning electron microscopy (SEM), fourier transforms infrared spectroscopy (FTIR), and UV-Vis spectrophotometer. The performance of the nanogenerator was performed using Osciloscop and electrometer in series and parallel connection. Based on the XRD pattern, ZnO sample has a hexagonal (wurtzite) crystal structure in which the lattice parameters of $a = b$, and $c = 3.258$ Å, and $5.218$ Å, respectively. The morphology of ZnO is a hexagonal form with diameters of 81.06 nm to 467.69 nm and rod length around 595 nm to 1331 nm. Based on the FTIR spectra, there are Zn-O bonds at 435–445 cm⁻¹, 535 cm⁻¹ and, 541 cm⁻¹. Moreover, ZnO nanorods have an energy gap band of 3.2 eV. The performance of the ZnO nanogenerator in series form has the highest voltage with a value of 10.36 volts AC, and the voltage value in dual-source is more than ten times larger compared to a single source.

1. Introduction
Piezoelectric Nanogenerators, first introduced in 2006, is a self-powered source of electrical energy from mechanical energy. Piezoelectric nanogenerators are very effective in use because they can generate electrical power from various mechanical energy sources in daily life in any form, such as pressing and bending from body movements. Piezoelectric nanogenerators have good portability, and it is one of the green technology solutions to reduce fossil fuel consumption [1]. On the other hand, piezoelectric nanogenerators are available in any size, easy to fabricate, and have high performance and low fabrication costs [2]. Therefore, piezoelectric nanogenerators have the potential to replace both batteries and accumulators in widespread use.

Various modifications have been conducted to improve the performance of the nanogenerator. In general, the improvement of nanogenerators can be made in a couple of ways, the first is modifying the materials, and the second one is combining it with other nanogenerators or devices. Material modifications are referred to the addition of doping to the materials, morphology modification, surface...
treatment, etc. [3,4]. The piezoelectric nanogenerator can be combined with solar cells, biofuel cells, and other nanogenerators to enhance power output [5–7].

Ren et al. reported that a nanogenerator integrated with organic solar cells on the sleeve shirt surface has performed of 80 V/ 3 μA [8]. Furthermore, Unsal and Bedeloglu had reported the study of two piezoelectric nanogenerators combined by arranging the layers; there were electrode, piezoelectric material, and an electrode generating 500 V / 5.3 mA [9]. In comparison, Alluri used two nanogenerators of different types, piezoelectric, and triboelectricity. They combined based on three layers, such as an electrode, piezoelectric-triboelectric material. The electrode has generated an even higher 530 V / 1.288 mA [10]. Those studies used parallel connections, and there has been no study that explains in detail the influence of variations of connections. The application of different connections affects the performance of the nanogenerator. It is influenced by the strong electric voltage by each electric source (nanogenerator) which previous studies have not revealed the performance issued in detail. Knowing the influence of series and parallel connections between piezoelectric nanogenerator will be a big step for the widespread use of green energy harvesters.

This study focuses on flexible piezo nanogenerators that depend on connection variations both in series and parallel. The treatment is done to determine the voltage of the nanogenerator output. Piezoelectric materials used are ZnO Nanorods. ZnO is a type-n semiconductor with a wide band gap (3.37 eV) [11]. This causes ZnO to have several advantages, such as good transparency and high electron mobility. Based on these advantages, ZnO is widely applied to piezoelectric nanogenerators. The commonly used ZnO structure in nanogenerator is the ZnO nanorod. The interesting thing about ZnO nanorod is its ability to convert mechanical power into electrical energy directly through the piezoelectric properties of the semiconductor material. Some research data prove that ZnO nanorods have better output than other ZnO nanostructures such as nanosheets of 400 mV/-A [12], nanowire 50 V/750 μA [13], nanofiber 60 mV V/ 0.03 μA [14] at 6.8 V/ 1.45 μA [15]. Then, the nanogenerator is tested Voltage one by one, in series, and parallel so that the output data is obtained in the form of voltage (V). This study was conducted to find out the detailed differences for the voltage output so that it becomes a reference in the use of nanogenerators as multi-source on green harvesting devices.

2. Method

2.1. Preparation of Al/PET Substrate
Aluminum-coated polyethylene (Al/PET) with 4 x 3 cm and 1 x 3 cm Kapton coated Al/PET (Kapton/Al/PET) were sonicated with acetone solution at 50 ºC and DI water at room temperature alternately for 15 minutes each. The substrate was then dried at room temperature to remove DI water on the surface.

2.2. Synthesis ZnO Nanoparticle Solution
The synthesis of ZnO nanoparticle solution begins with dissolving zinc acetate powder (Zn(CH$_3$CO$_2$)$_2$), and 20 ml of ethanol (C$_2$H$_5$OH) then stirred with a magnetic stirrer on a hot plate with a temperature of 70 ºC for 45 minutes at 300 rpm. Then added 0.2 M Monoethanolamine (MEA) and stirred again using a magnetic stirrer on a hot plate at 70 ºC for 2 hours with 300 rpm peed so that a clear solution was obtained.

2.3. ZnO Seed Layer Procedure
ZnO nanoparticle solution was deposited on the Al/PET substrates using the spin coating method with 3000 rpm speed for 30 seconds. Then the film was heated at 100 ºC for 3 hours.

2.4. Synthesis Film ZnO Nanorods
ZnO nanorods are grown on top of seed layers using Zinc Nitrate tetrahydrate (Zn(NO$_3$)$_2$·4H$_2$O), and Hexamethylenetetramine (C$_6$H$_2$N$_4$) dissolved DI Water. The hydrothermal method refers to Kammel’s method to grow these ZnO nanorods [16]. The solution was stirred with a magnetic stirrer
for 45 minutes at room temperature with a speed of 300 rpm. Furthermore, ZnO film was dipped in the solution for 6 hours at 95 °C. Then the sample was rinsed using DI water and heated for 3 hours at a temperature of 100 °C in the oven. The fabrication of each layer of the piezoelectric nanogenerator can be seen in figure 1.

**Figure 1.** Layers of Piezoelectric Nanogenerator

2.5. Characterization

To determine crystal structure, ZnO nanorods were characterized by x-ray diffraction (XRD) with PANalytical X’Pert PRO type. Scanning electron microscopy (SEM) with FEI Inspect-S50 type was used to determine the morphology properties of the ZnO nanorods. Fourier transforms infrared spectroscopy (FTIR) with Shimadzu IR Prestige 21 type was used to identify chemical bonds of the ZnO nanorods, and UV-Vis spectrophotometers with Analytic Jena Specord 200 Plus type was used to determine the bandgap of ZnO nanorods.

2.6. Voltage Measurement

Oscilloscope InfiniiVision DSO-X 2012A was used to measure the alternating current (AC) voltage of electrical nanogenerators (NG). The pressure used at 12.3 kN/m² was given to piezo nanogenerator samples. The performance of this nanogenerator device (active area 3 x 3 cm²) was measured by pressing the nanogenerator using an automatic suppressor so that electrical energy could be harvested from mechanical energy while the models used were two different material systems that are interconnected, i.e., polyimide layer on one substrate and ZnO nanorods in the other substrate. In addition, spacers (barriers between electrodes) were applied to nanogenerators with spacer distances of 3.0 mm (sample A) and 1.5 mm (sample B). Open circuit (Voc) voltage was measured in the absence of external voltage applied to the sample. This measurement referred to Huang modeling [17]. The Voc of A and B samples were measured as a single source piezo nanogenerator. Second, The samples were measured with series connections, AB and BA as dual-source connected series piezo nanogenerators. Finally, it was connected in parallel and measured the Voc as a parallel-connected dual-source piezo nanogenerator.

3. Result and Discussion

3.1. ZnO Nanorods Crystal Structure

The x-ray diffraction pattern produced by ZnO nanorods on the Al/PET substrate using a hydrothermal method at 95 °C as shown in figure 2. Investigation of lattice parameters and crystal size of ZnO nanorods is conducted based on XRD data by Rietveld refinement method using Rietica program. Based on the visible pattern, the ZnO nanorods sample has a hexagonal crystal structure (wurtzite), as evidenced by the presence of diffraction peaks in the hkl (002) plane that is oriented towards the c axis. This is validated by research conducted by Nikam, and Yao, also in accordance with JCPDS 36 – 1451 [18,19]. The sample lattice parameters of ZnO nanorods a = b and c are 3.258 Å and 5.218 Å, respectively. This lattice parameter indicates that crystals formed with single-phase accordance to
research conducted by Mufti [20]. Crystal size is obtained through calculations based on Scherrer equations.

\[ D = \frac{0.9 \lambda}{B \cos \Theta} \]

The value of 0.9 is the constant of Scherrer on the crystals of the crystals [21]; \(\lambda\) X-ray wavelengths from Cu K\(\alpha\) radiation, i.e. 0.154056 nm; \(B\) is the Full Width Half Maximum (FWHM) value which is the highest half full width value of the peak; and \(\Theta\) is the peak diffraction in rad. The sample crystal size of ZnO nanorods that are FWHM at peak (002) is 49.09 nm.

![Figure 2](image)

**Figure 2.** X-Ray diffraction pattern of ZnO nanorods

Figure 2 shows the ZnO nanorods peaks that are formed are at angles of 31.89° (d100), 34.27° (d002), 36.07° (d101), 47.41° (d102), 56.45° (d110), and 62.95° (d103). On the other hand, there are other peaks found such as PET (25.77° and 53.05°), Al (111) 38.39°, Al (200) 44.63°, Al (202) 65.03°, and Al (311) 78.17°. The intensity of the ZnO peaks is lower than the substrate peaks due to the heating temperature of the film being 100 °C so that the crystallinity of ZnO nanorods has not been perfectly formed. It was also reported that the best crystallinity of ZnO nanorods is produced by a temperature of 400 °C in the film heating process [22].

### 3.2. Morphology of ZnO Nanorods

The morphological tracing of crystals owned by ZnO nanorods is shown in figure 3 a) – c), which results from surface and cross-sectional magnification using an electron microscope. The results showed that seed layers have successfully produced ZnO that grows vertically parallel, with the length and diameter of ZnO nanorods represented successively by thickness and diameter. In figure 3 a) – b) visible morphology of the surface of ZnO nanorods are enlarged 20k times and 50k times, respectively. The image shows that the ZnO formed is hexagonal, and nanorods are scattered throughout the surface of the substrate. However, the nanorods in the sample were not evenly dispersed. It is because of the lack of thickness of the ZnO seed layers. According to Solis-Pomar’s research, the thicker ZnO seed layers, the better the spread of ZnO nanorods will be [23].

| Elements | Wt% | At% |
|----------|-----|-----|
| OK       | 15.00 | 41.89 |
| ZnK      | 85.00 | 58.11 |
| Matrix   | Correction | ZAF |

**Table 1.** EDX of ZnO Nanorods.
The diameters of the nanorods varied from the smallest to the largest, i.e., 81.061 nm to 467.69 nm with an average distribution size of nanorods diameter are 318.85 ± 2.05 nm shown by figure 3 d). Further morphological tracing is seen in figure 3 c), which shows the morphology of cross-sectional ZnO nanorods by SEM. It appears that ZnO nanorods bind well to the substrate. The measurable thickness of ZnO nanorods is a variety of smallest to largest sizes of 595.27 nm to 1331.84 nm with an average distribution thickness of ZnO nanorods of 983.23 ± 112.94 nm (see figure 3 e)). The results were confirmed by research by Wahid that observed ZnO nanorods depending on the growth times and heating of the substrate, with the result being ZnO nanorods that through heating temperatures of 100 °C for 6 hours can reach a maximum thickness of 1500 nm [24]. Furthermore, the sample content is obtained from the characterization of EDX, as seen in table 1. It is seen in table 1 that the content of oxygen and zinc is 41.89 % atoms and 58.11 % atoms.

![Image 1](image1.jpg)

**Figure 3.** Scanning Electron Microscopy image of ZnO nanorods image a) with magnification of 50k times on the surface b) the cross section of ZnO nanorod c) histogram diameter size distribution, and c) histogram thickness size distribution.

### 3.3. Chemical Bond of ZnO Nanorods

Fourier transform infrared spectroscopy (FTIR) spectra is used to identify chemical bonds in the ZnO nanorods. Figure 4 shows the spectrum produced from the ZnO nanorods sample. The presence of Zn-O bonds is commonly found in areas around 400-500 cm⁻¹ [25]. The absorbance of Zn-O is found at 435-445 cm⁻¹ [26]. A small span was observed at about 535 cm⁻¹ indicating the bending vibration of Zn – O [27]. Absorption of Zn-O tendril vibration characteristics was observed at 541 cm⁻¹ [28]. On the other hand, some chemical bonds are still present in the sample (other than Zn-O) as at the 1330 cm⁻¹,
1365 cm\(^{-1}\), 1670 cm\(^{-1}\), 2370 cm\(^{-1}\), 3483 cm\(^{-1}\) and 3574 cm\(^{-1}\) (see figure 4.). The FTIR analysis result also confirms the ZnO successfully has been formed.

Figure 4. FTIR Spectra of ZnO Nanorods

Figure 5. a) Absorbance Spectrum of ZnO Nanorods Sample. b) Graph of the Relationship between \((ahv)^2\) and \(hv\).

3.4. Energy Band Gap Film ZnO
The absorbance of UV-Vis Spectra at wavelengths of 350 - 1100 nm from ZnO is shown in figure 5 a). The presence of peak absorption is observed in the wavelength range of 360 - 380 nm, which is the typical signal of pure ZnO. It appears that ZnO wavelength absorption is less than 400 nm indicates the intrinsic gap of ZnO [29]. The energy gap value (Eg) is obtained using tauc method from the characterization of UV-Visible Spectroscopy (absorbance) as shown in figure 5 b). The intersection of the linear equations obtains the energy bandgap (Eg) of ZnO 3.2 eV. This value of the energy band gap matches some ZnO research that has been done from previous research [30].

3.5. Piezo Nanogenerator Output Performance
3.5.1. Piezo Nanogenerator as a Single Source. In figure 6, a) – b) visible output performance of sample A and B nanogenerators. Voltage Measurement is done by Open circuit (Voc). Samples A and B are
two samples synthesized by the same method. Before the output measurement was performed, both samples measured the resistance. The resistance values of sample A and sample B are 5.83 MΩ and 1.16 MΩ, respectively.

![Graphs of Voc](image)

**Figure 6.** A measurable graph of the nanogenerator as a single source of time. a) Voc Sample A. b) Voc Sample B.

When the nanogenerator is pressed, the voltage and current will appear, indicating a difference in potential and load flow. This is due to the spontaneous polarization of both layers due to the outermost atom being unable to preserve its position. This is a result of the noncentrosymmetric nature of ZnO. The symmetry group of ZnO P63mc causes the anisotropic piezoelectric properties. Thus, an electric dipole appears inside the crystal and can be modulated by the application of mechanical voltage (direct piezoelectric effect). In this situation, the dipole moment will appear throughout the crystal and cause a tendency to be negatively charged in the ZnO system and positively charged in opposite material systems so that there is a potential difference that triggers the transfer of cargo from one material to another. In this state, the nanogenerator reaches its maximum potential. After releasing the pressure from the nanogenerator, the crystalline dipole moment will return to zero so that there is no potential difference and no electron displacement in both materials [31]. This is indicated by the shrinking of the measured output on the oscilloscope and electrometer. In this time, the nanogenerator reaches its minimum potential.

From figure 6, it was later seen that Voc sample A was larger than Sample B. Voc A had the highest value of 0.85 Volts while B was 0.60 Volts. It is due to the difference in spacer thickness in the other samples. Spacers work as "payload dividers" to control piezoelectric charge generation points during the press-release process [32]. When the Al electrode is pressed, the spacer leaves several microseconds to accumulate a piezoelectric charge on ZnO nanorods. In the case of piezoelectric charge generation, mechanical deformation of the dipole occurs only when the Al electrode comes into contact with ZnO nanorods. The presence of spacers is useful to allow piezoelectric to be sequential so that piezoelectric is more effective.
3.5.2. Piezo Nanogenerator as Dual Source Connected Series. In figure 7 a) – b) visible output performance of the series-connected nanogenerator A and B samples. AB's description indicates that both the positive probe of the oscilloscope is connected to the sample A nanogenerator while the negative probe is connected to the sample nanogenerator B. Voltage measurements are performed in the same way as the Open circuit (Voc). With the series-connected, the resistance value is 6.99 MΩ.

![Figure 7](image)

**Figure 7.** A measurable graph of the nanogenerator as a serially connected dual source to time. a) Voc Sample AB. b) Voc Sample BA.

It appears in figure 8 that there is a difference in output results when samples A and B are connected back and forth. AB sample output performance has the smallest output even from sample A (when used as a single source), whereas BA samples have the highest output compared to sample B (when used as a single source). Given that the nanogenerator is an AC power source, the signal emitted by each nanogenerator during the press-release process will combine. As shown in Figure 8. a) – b), each sample has a different output for voltage so that when nanogenerators A and B are paired back and forth in series, it will certainly produce constructive or destructively output (see figure 8) [33]. The Voc BA has a greater value of 10.36 Volts at the highest measured voltage, while the Voc AB is 1.12 Volts. It proves that both measured voltages are gaining voltage (constructive). The problem is caused by a very small time difference in the press-off process so that the resulting phase difference has not reached the maximum value that both nanogenerators can achieve.

![Figure 8](image)

**Figure 8.** a) wave weakening (destructive) events. b) wave strengthening events (constructive)
Figure 9. A measurable graph of the Voc nanogenerator as a dual source that connects parallel to time

Figure 9 shows output performance A and B samples connected in parallel. Voltage measurements are done in the same way as Open circuits (Voc). With the parallel connection, the resistance value is 0.967 MΩ. When two or more sources of AC (in this study were nanogenerators) are operated in parallel, currents can circulate between them. This current circulation occurs if the internal voltage generated by each nanogenerator is different. Currents produced from any nanogenerator can flow into one or more other nanogenerators [34]. In that case, it will lead to the meeting of the two waves produced by the nanogenerator and lead to the weakening and strengthening voltage. Unlike series connections, when connected in parallel, we will find significant changes to the electric current.

On the other hand, the output voltage (see figure 9) shows a value of 6.59 Volts. This time, a (constructive) strengthening event occurred; however, no higher than the nanogenerator paired in series. This is in accordance with research conducted several years ago that when the voltage source is connected serially, it will only cause a significant increase in its electric current.

4. Conclusion
In this study, piezo nanogenerator-based ZnO nanorods were successfully synthesized through hydrothermal methods. X-ray diffraction pattern of ZnO nanorods shows that the crystal structure it has is hexagonal (wurtzite), as evidenced by the high diffraction peak in the hkl (002) plane that is oriented towards the c axis. The morphology of ZnO in hexagonal form was shown by Scanning Electron Microscopy (SEM). Furthermore, the characterization of SEM Crosssection confirms that the ZnO hexagonal extends vertically and binds well to the substrate. Based on the Fourier spectrum of transform infrared spectroscopy (FTIR), the sample has a Zn-O bond. ZnO Nanorods has an energy gap band of 3.2 eV. According to the characterization of the voltage in nanogenerators as single and multi-source parallel-series, it is seen that the series nanogenerator has the most significant voltage with a value of 10.36 volts AC. Interestingly, when connected as a dual source, it will produce a voltage more than ten times rather than a single source. This is due to the strengthening output phase of nanogenerators.

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