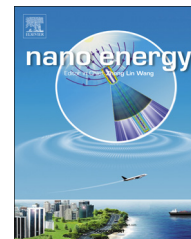




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RAPID COMMUNICATION

High-performance hybrid cell based on an organic photovoltaic device and a direct current piezoelectric nanogenerator



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Abstract

The search for harvesting both the mechanical and solar energies from a single hybrid system is of significant value and represents a new trend in energy harvesting technologies. This single hybrid system can utilize both the energy sources easily available from nature and most importantly it is clean and sustainable. It is a novel technique involving completely different physical principles utilized for scavenging different types of energies. This report presents studies of a hybrid power generator made a direct-current piezoelectric nanogenerator based on ZnO nanosheets and a bulk heterojunction organic solar cell based on an inverted structure. The device shows much larger electric power output compared to its two individual power output components, which facilitates more effective multi-type energies harvesting and clarifies a mechanism for realizing multi-functional energy devices.

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Introduction

New energy harvesting devices have developed considerably in recent decades, due to the continuous growth in the demand for renewable energy sources, the increasing need

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to reduce global warming and the continued depletion of fossil fuels in modern society [1-5]. Harvesting mechanical, thermal, magnetic, chemical, and light energies from living environments is one of the most urgent challenges for the sustainable development of human civilization and has critically importance for powering small scale portable electronics, self-powered body-implanted devices, and self-powered sensors [6-9]. Recently, intensive research has focused on developing various types of photovoltaic devices for harvesting solar light energy; of these, organic materials-based photovoltaics as energy harvesting devices have proven to be important since they are flexible, eco-friendly, and easy to process. Further very effective power conversion of organic solar cells (OSCs) under indoor illumination compared to other solar cells should be noted [10-12].

In addition, many types of mechanical energy scavenging devices such as a piezoelectric nanogenerator (PNG) have attracted considerable attention for self-powering small scale devices including sensors and wearable electronics [13-19]. Among the various piezoelectric materials utilized for fabricating PNGs, the ZnO nanostructures have been regarded as the most popular building blocks owing to their semiconducting and piezoelectric coupling properties [20,21]. Furthermore, harvesting both mechanical and solar energies from a single hybrid system is currently highly desirable and represents a new trend of all-in-one multiple energy harvesting technologies [22]. Moreover, because of the completely different physical principles utilized for scavenging different types of energies, each type of corresponding conversion device involves an independent unit. Therefore, innovative approaches can be developed for the conjunctural harvesting of multiple types of energies using an integrated structure so that the energy resources can be

effectively and complementarily utilized whenever and wherever one or all of the energy resources are available.

Recently, although harvesting both solar and mechanical energies at the same time from a single hybrid cell based on both a one-dimensional (1D) ZnO nanorod-based PNG and a solar cell has been demonstrated in the previous researches [23-26], the effect on the total performance of the hybrid cell due to the different nature of the piezoelectric output signals from PNG has not been clearly investigated and discussed. In the reported hybrid systems, due to the alternative current (AC) piezoelectric output from PNGs and the direct current (DC) output electric signal from the solar cell, the total output from the hybrid cell is degraded significantly during rectification. Nevertheless, the output current generated from PNG in the hybrid system is quite low compared to that in the solar cell, thus restricting the wide application of the hybrid cell for powering small scale electronic devices.

In the present article, we report the fabrication of a hybrid system consisting of a highly efficient DC type PNG (DC-PNG) based on 2D ZnO nanosheets and an OSC based on poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C₆₁-butyric acid methyl ester (PC₆₀BM) for harvesting multiple type energies, i.e. solar energy and mechanical energy simultaneously/individually. We also demonstrate the high DC output current from the PNG cell under vertical compressive force/pressure and high output performance from an OSC separately under light illumination. Our approach relies on the connection of the anode of the OSC with the cathode of PNG to harvest solar and mechanical energy under external mechanical force and light illumination. The power-generating performance of the serially integrated hybrid cell (s-HC) is synergistically enhanced by the contribution of a PNG, compared with the output power

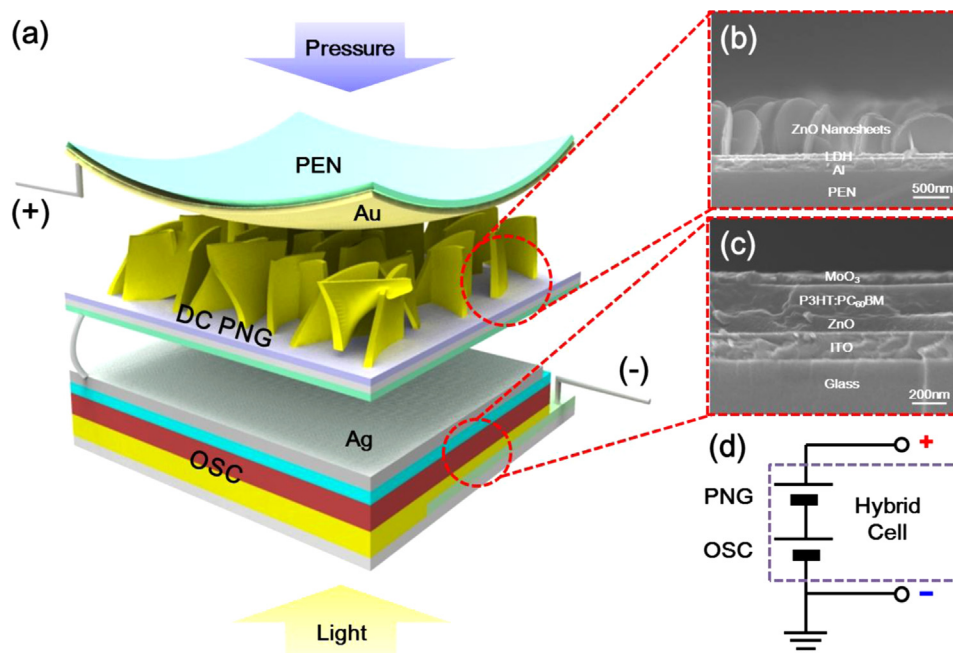


Figure 1 Design of a s-HC for simultaneous harvesting of mechanical and solar energies under external mechanical force and light illumination. (a) 3D schematic representation of a s-HC. (b) Cross-sectional FE-SEM image of ZnO nanosheets grown on an Al film. (c) Cross-sectional FE-SEM image of the inverted OSC structure. (d) A schematic showing equivalent circuit of the s-HC based on OSC and DC-PNG.

generated independently from the solar cell component under illumination. The mechanisms of power generation from our fabricated s-HC and corresponding synergetic effect are discussed in detail in light of band bending and piezoelectric polarization. Our work provides a promising approach for harvesting multi-type energies more effectively and a mechanism for realizing multi-functional energy devices.

Experimental

The inverted structure OSC is fabricated on $1.5 \times 1.5 \text{ cm}^2$ indium tin oxide (ITO)-coated glass. The substrates are cleaned with acetone and methanol. A ZnO precursor is made by dissolving 0.91 g of zinc acetate and 0.447 g of ethanolamine in 9.216 g of 2-methoxyethanol, at 80°C for 1 h, then sonicated for 30 min. The ZnO film is spin-coated on the ITO/glass substrate at 2500 rpm for 20 s and annealed at 160°C for 30 min. These ZnO coated substrates are placed into a glove box with a nitrogen environment. A polymer blend is composed of P3HT and PC₆₀BM in chlorobenzene under a 1:1 component ratio of P3HT:PC₆₀BM with a concentration of 20 mg/mL. The active layer is also spin-coated at a speed of 1000 rpm for 60 s on ZnO film and heated at 140°C for 30 min. Then, the MoO₃ layer is evaporated on the top of the active layer and Ag is deposited using a shadow mask by thermal evaporation in a vacuum of about 3×10^{-6} Torr. The active area of OSC is 0.06 cm^2 .

For the PNG, $2 \times 2.5 \text{ cm}^2$ Al tape is attached to a polyethylene naphthalate (PEN) substrate and cleaned using acetone to remove impurities. In order to grow the ZnO nanosheets on the Al substrate, zinc nitrate hexahydrate [$\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$] (0.025 M) and hexamethylenetetramine (0.025 M) are mixed with de-ionized water (250 mL). The Al substrate is immersed in this solution at 95°C for 3 h. To fabricate the anode substrate which is placed on ZnO nanosheets, Au is deposited on the PEN substrate using a thermal evaporator of 200 nm thickness.

Results and discussion

Figure 1 shows a schematic diagram and cross-sectional field-emission scanning electron microscopy (FE-SEM) images of an s-HC based on an OSC and a DC-PNG. The flexible DC-PNG is based on the ZnO nanosheets of about $0.5 \mu\text{m}$ height as a piezoelectric active layer and the Zn:Al layered double hydroxide (LDH) layer of about 20 nm thickness on the Al/PEN substrate (Figure 1a and b). The bottom side of the PNG device, the Al layer, serves not only as an electrode, but also as a catalytic layer for the formation of the ZnO nanosheets via the unintentionally self-assembled LDH layer. The OSC is fabricated with an inverted structure based on Ag (anode, 80 nm)/MoO₃ (electrode blocking layer, 20 nm)/P3HT:PC₆₀BM (active layer, 200 nm)/ZnO (electron transport layer, 50 nm)/ITO (cathode, 200 nm) (Figure 1c). An equivalent circuit of the s-HC composed of OSC and PNG is shown in Figure 1d. The performance of s-HC was measured in this study with the transparent OSC side injecting the sun-light source and the PNG side pushing the top PEN side as shown in Figure 1a.

The performance of the s-HC is characterized by measuring the open-circuit voltage (V_{oc}) and short-circuit current density (J_{sc}). The output parameters are presented as V_{OSC} and J_{OSC} for OSC, V_{PNG} and J_{PNG} for PNG, and V_{HC} and J_{HC} for the s-HC. Figure 2a shows the current density vs voltage (J -

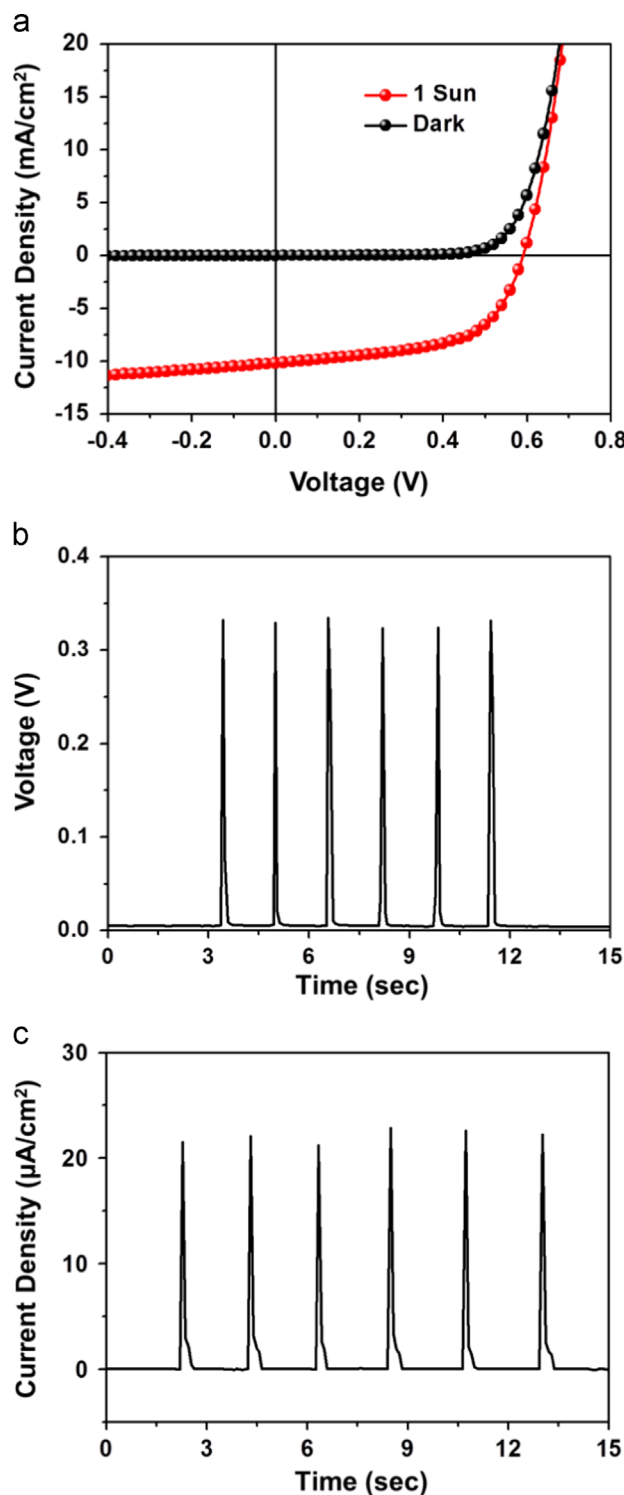


Figure 2 Typical output signals of the OSC and PNG. (a) J - V curve of the OSC resulting from conversion of solar energy in P3HT:PC60BM into electricity. (b, c) DC-type VPNG and JPNG graph from PNG under vertical compressive force, respectively.

V) curves of OSC used in s-HC under air mass 1.5 global irradiation and in the dark [27]. The OSC showed distinct diode behavior in the dark condition and the J_{OSC} and V_{OSC} with the values of 10.17 mA/cm^2 and 0.59 V were obtained under a light condition. The time-dependence V_{PNG} and J_{PNG} are shown in Figure 2b and c. When a vertical compressive pressure is applied periodically to the PNG in the dark condition, very stable and DC-type outputs with V_{PNG} of 0.378 V and J_{PNG} of very high value up to $22.06 \mu\text{A/cm}^2$ are generated.

It is worth noting that a negative piezoelectric output, which is observed in the AC-type outputs of common PNGs, is not observed after the removal of pressure in the present case. The DC-type output signal from the proposed PNG is strongly related to the Zn:Al LDH layer formed between the ZnO nanosheets and Al electrode. When ZnO nanosheets undergo an external vertical compressive mechanical force, the piezoelectric potential is created by the buckling of the ZnO nanosheets [28]. Further, the LDH layer is an anion exchange and weak conducting layer, which contains positively charged

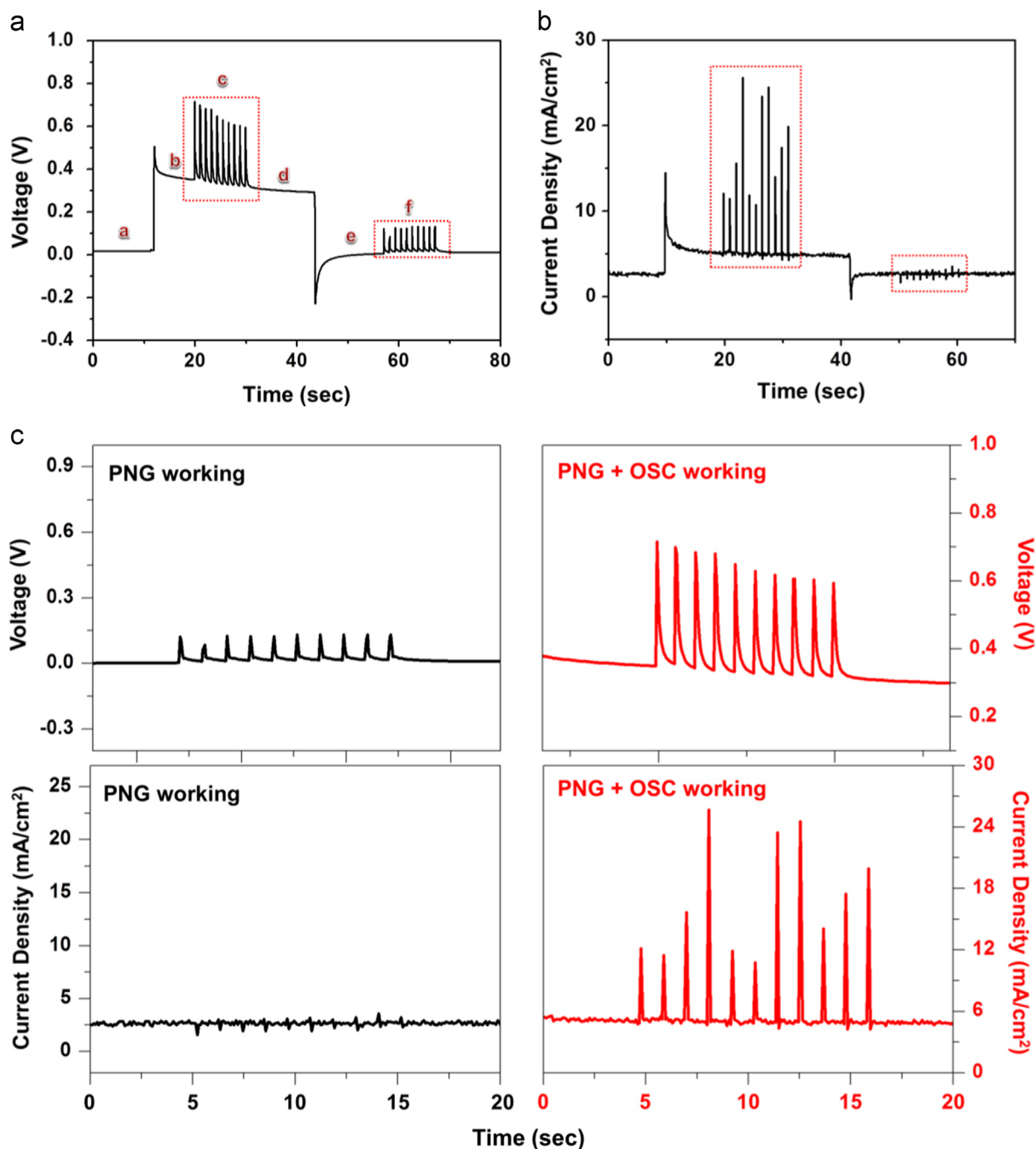


Figure 3 (a) V_{HC} and (b) J_{HC} of the s-HC. The six stages labelled “a”, “b”, “c”, “d”, “e”, and “f” are measured under controlled application of applied light and mechanical compressive force. (c) Enlarged plots of the corresponding outputs showing the difference in the V_{HC} and J_{HC} between the cases, when only PNG is working (left panel) and when both OSC and PNG are working (right panel).

layers as well as charge balancing anions (NO_3^-) located in the interlayer region. The positive charged layers in LDH, facing the ZnO nanosheet, are compensated by the electrons from the ZnO nanosheets, resulting in an accumulation of large negative charges at the ZnO/LDH interface layer [28]; a high potential difference is also created, resulting in a DC-type V_{PNG} and J_{PNG} observed under mechanical stress as shown in Figure 2b and c. These DC outputs have significant advantage in facilitating the integrated hybrid device of highly efficient power output without using any additional rectification diode, which is generally required to convert AC to DC.

Figure 3 shows the measured V_{HC} and J_{HC} from s-HC under controlled applications of dark/light illumination and mechanical force. As can be seen from Figure 3a, no V_{HC} (base-line ~ 0 V) from s-HC is observed when the s-HC is measured under dark condition and in absence of external pressure (labelled “a” in Figure 3a). When lit up, V_{HC} rapidly increases and is maintained at about 0.4 V due to the photovoltaic characteristic of OSC (labelled “b” in Figure 3a). When the mechanical pressure is applied periodically at an interval of 1.0 s to the s-HC, V_{HC} increases up to a maximum of 0.71 V with DC output pulse (labelled “c” in Figure 3a). As the light turns off and the mechanical force is removed, V_{HC} rapidly decreases to 0 V (labelled “e” in Figure 3a). Again, when the pressure is applied periodically, steep V_{HC} peaks are also measured, even though OSC is not functioning (labelled “f” in Figure 3a). Similar trends are also observed for the J_{HC} curve as shown in Figure 3b. By integrating the OSC and PNG, V_{HC} of s-HC seems to be close to the sum of the output voltages of OSC and PNG. In other words, the s-HC yields an overall enhancement of output voltage (“c” in Figure 3a) in comparison to the output of only OSC (“b” in Figure 3a) or that of only PNG (“f” in Figure 3a) in s-HC. Very interestingly, it should be noted that the measured values of V_{HC} and V_{PNG} from s-HC differ before and after the hybridization. Figure 3c shows the enlarged plots corresponding to the values of V_{HC} and V_{PNG} of Figure 3a. It is evident that a difference of output voltage occurs up to 0.15 V as shown in Figure 3c. The significant difference in J_{HC} is also manifested more clearly and the details are described in the next section. Therefore, Figure 3c indicates that the enhancement in hybrid systems is not only a simple sum of the outputs from the two devices, but is also due to the synergy effect between the devices having different operation mechanisms, as will be discussed further in Figure 6.

Generally, the AC output of PNG needs a rectification process to obtain a DC signal. The s-HC produces a high output with DC signal without any rectification, avoiding any extra rectification circuit in the proposed s-HC will definitely provide further advantages in the miniaturization sector. Furthermore, to better understand the charge generation and working mechanism of the power enhancement from s-HC, two more hybrid cells were fabricated. The first hybrid cell was fabricated using similar OSCs and with different piezoelectric materials such as poly(vinylidene fluoride-co-trifluoroethylene) [P(VDF-TrFE)] polymer. The V_{HC} of the fabricated s-HC based on P(VDF-TrFE) is measured under controlled application of dark, light illumination, and compressive force. In the case of s-HC with P(VDF-TrFE)-based PNG (electrode/P(VDF-TrFE) thin film/electrode/PEN), the output voltage due only to the piezoelectric P(VDF-TrFE)-based PNG (labelled “b” in Figure 4a) was generated under compressive force from s-HC; however, the total output V_{HC} is not obtained (labelled as “a” in Figure 4a)

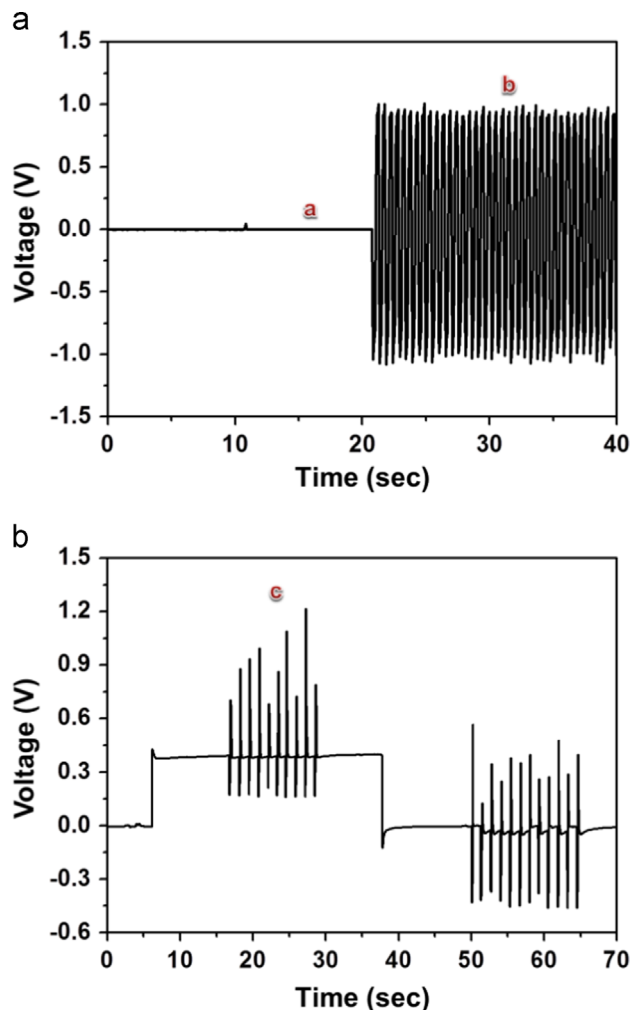


Figure 4 V_{HC} of the s-HC using various PNGs. (a) The s-HC using an OSC and a P(VDF-TrFE)-based PNG (PEN/ITO/P(VDF-TrFE)/ITO). (b) The s-HC using the OSC and a ZnO thin film based PNG (the structure is PEN/ITO/ZnO/MoO₃/Ag).

due to the electrically insulating property of P(VDF-TrFE) and therefore the output current generated in the OSC cannot pass through the P(VDF-TrFE) thin film.

Secondly, another s-HC is fabricated using the same OSC and a piezoelectric ZnO thin film instead of nanostructures. The V_{HC} of the fabricated s-HC was also measured under controlled application of dark, light illumination, and compressive force. The V_{HC} measured under vertical compressive force is labelled “c” in Figure 4b. As shown in Figure 4b, the piezoelectric AC signal is produced from PNG, while a DC type signal is observed from the OSC under light illumination; therefore, although in this case, both solar and mechanical energies are harvested simultaneously, due to the AC-type output from PNG, the s-HC requires a rectifier circuit using diodes to convert AC to DC to power the electronics. Therefore, the proposed s-HC using ZnO nanosheets is not only able to harvest both solar and mechanical energy together, but can also offer high DC output without any further rectification process. Hence, the proposed hybrid system can work as a full-time energy harvesting system when OSC and PNG are connected in series, where PNG effectively can work as a piezoelectric energy harvester using mechanical sources such as wind, sound, and human activity. The OSC also

plays the role of energy harvester using light source, forming a composite energy harvesting system for multiple simultaneous energy generation.

Figure 5 shows the V_{HC} and from s-HC, under vertical compressive pressure applied and held for a long period of time. The nature of the output results in terms of hybridization shows a similar trend as that shown in Figure 3a. When the pressure is applied periodically at 1.0 s intervals for an extended period of 3.0 s, V_{HC} increases and remains constant while pressure is held on s-HC. The V_{HC} graph shown in Figure 5a indicates a magnified pulse of V_{HC} . The results obtained can be explained using the energy band diagram (Figure 5b-d). When the OSC is connected with a PNG in series, in such a way that the Au electrode is connected to the cathode of the OSC and the Al electrode is connected to the anode of the OSC, the Schottky barrier height (ϕ_1) is created at the end of the conduction band (CB) and valence band (VB) of the ZnO nanosheets and Au interface (Figure 5b) [28]. When light illuminate the s-HC in the absence of any pressure, the quasi-Fermi levels (E_F) of Au is shifted ($\Delta\phi$) resulting in a V_{OSC} generated by the OSC. Flowing of the photo-generated electrons from the OSC is inhibited through the PNG due to the formation of the Schottky barrier between the ZnO nanosheets

and the Au electrode. This phenomenon derives the increase of the resistance of PNG and therefore V_{HC} decreases in the circuit (original V_{OSC} is 0.59 V shown in Figure 2a).

When the ZnO nanosheets are subjected to mechanical pressure from the top, large negative charges (V^- formation) at the ZnO nanosheet/LDH interface build up, while corresponding positive charges (V^+ formation) are accumulated at the Au electrode side (Figure 5d), which results in band structure change in the ZnO nanosheets according to our previous report [28]. The former leads to the piezoresistance effect, which causes the electrons from the OSC to start flowing more significantly through the PNG. Consequently, during this time when the pressure is applied, the V_{HC} increases as also reflected in Figures 3a and 5a. Further, piezoelectric potential remain in the ZnO nanosheets for an extended period of time without being fully screened by the free carriers as long as the pressure is preserved [29,30]. As a result, the CB of ZnO nanosheets near the Al electrode are elevated by the remnant polarization towards the Al electrode, and the electrons flow from the Au electrode to the Al electrode through the PNG. Alternatively, as the compressive pressure on the PNG is released, the CB and VB of the ZnO nanosheets revert back to the initial state,

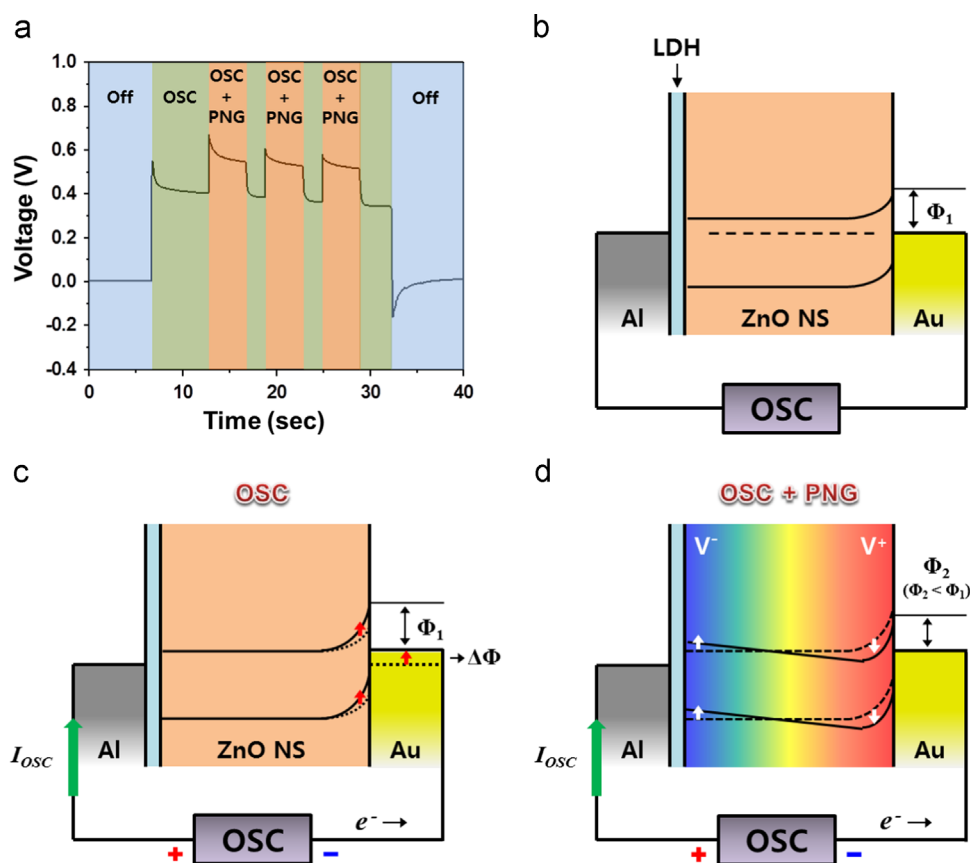


Figure 5 Energy band diagram of the s-HC corresponding to the “OSC” and “OSC+PNG” processes labelled in Figure 5a. (a) V_{HC} of the s-HC when the pressure is applied periodically at an interval of 3.0 s for an extended period of 1.0 s. (b) Energy band diagram of PNG in absence of any pressure having Schottky contact with an Au electrode and an OSC. The dashed line shows the Fermi level of the ZnO nanosheets; upper and lower solid lines refer to the conduction and valence band of ZnO, respectively. (c) Energy band diagram when an OSC is working; dashed and solid lines represent the band diagrams before and after applying the bias provided by an OSC, respectively, with a relative shift of the Schottky barrier height (ϕ_1) at the end of the Au electrode. (d) When pressure is applied to the PNG, ZnO nanosheets exhibited a new band diagram; dashed and solid lines represent band diagrams before and after applying the pressure on PNG. ϕ_2 refers to the changed Schottky barrier height.

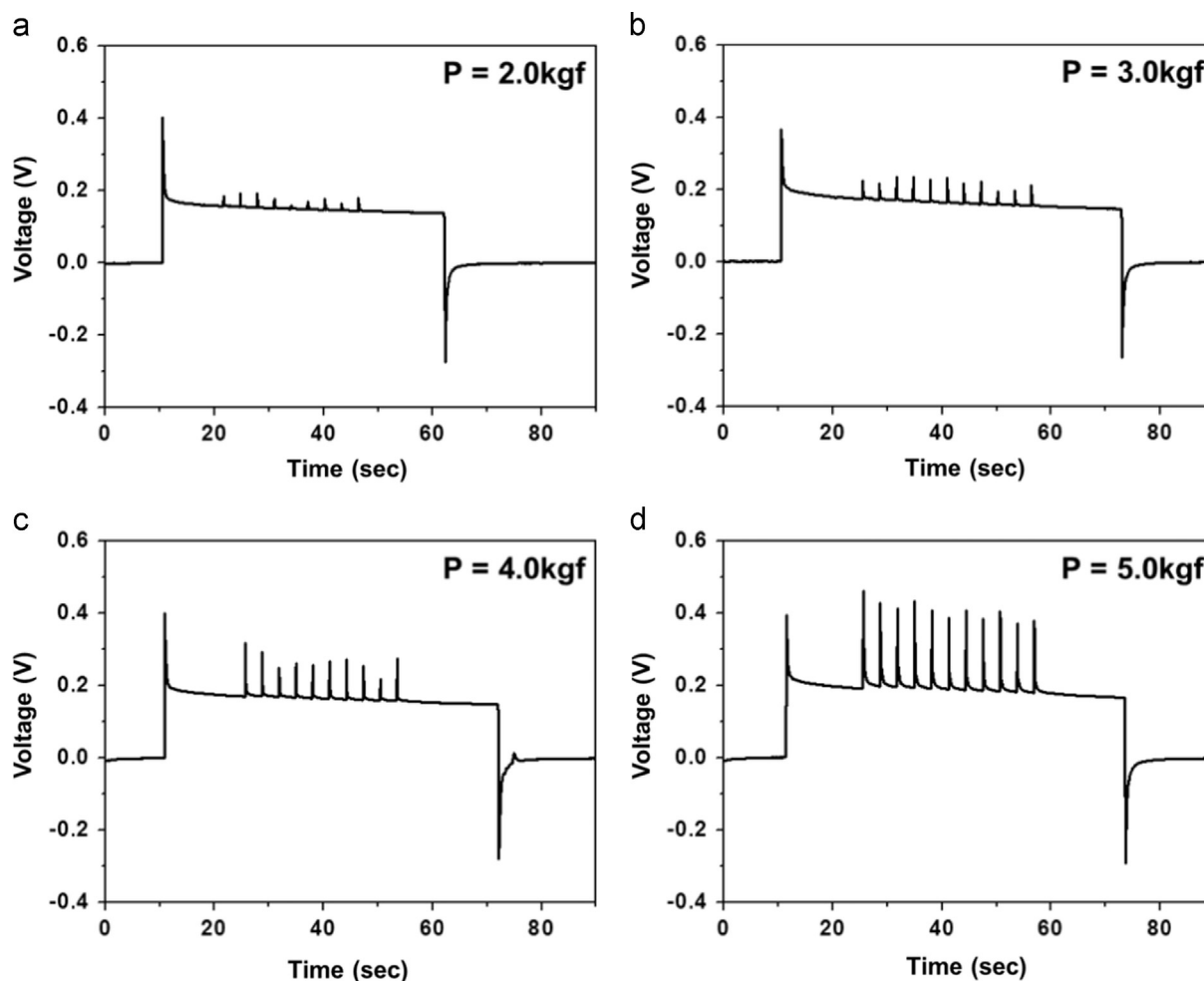


Figure 6 (a-d) V_{HC} graphs of s-HC under different pressures. V_{HC} response of the s-HC to increasing pressure.

where only OSC is working (Figure 5c). With the immediate removal of the light again, the modified Schottky barrier height and the band energy level return to the original state (Figure 5b).

We measured the voltage of s-HC under application of different compressive pressures in order to verify the enhancement of piezoelectric potential from s-HC. An increase in the pressure on the ZnO nanosheets leads to an increase of piezoelectric potential producing high output voltage in the circuit. The variation of V_{HC} from s-HC with the increase of pressure is shown in Figure 6a-d. As the pressure increases in steps, i.e. from 2.0 kgf to 5.0 kgf, the V_{HC} linearly increases. Therefore, the V_{OSC} of OSC is influenced more as the piezoelectric potential increases in the ZnO nanosheet. Hence, the proposed hybrid energy harvesting device and its output results present a new path for photovoltaic power enhancement. It is well-known that the efficiency of organic solar cells is too low for commercialization and several studies on a hybrid structure are still in progress by various research groups. In the present study, the integration of a novel DC type PNG with OSC is successfully achieved not only for harvesting multiple energies simultaneously but also for providing a way to enhance the open-circuit voltage and short-circuit current of an organic solar cell. Furthermore, sustaining high output piezopotential during the holding of pressure offers considerable advantage for

the continuous and high output supply to power small scale nanodevice systems by s-HC.

Conclusion

In summary, we presented a serially integrated hybrid cell consisting of an organic solar cell and a piezoelectric nanogenerator, which can be used in an energy harvesting system for converting solar and mechanical energy into electricity simultaneously. When periodic vertical compressive pressure is applied to the PNG, the open-circuit voltage of OSC increases due to the piezoelectric potential of the ZnO nanosheets. The energy band diagram in the ZnO nanosheet is tuned under the controlled application of force and light. The photo generated electrons from the OSC can effectively flow through the PNG, due to the lowered Schottky barrier height between the ZnO nanosheets and the Au electrode. Consequentially, JSC from s-HC also increases under application of compressive pressure. These DC outputs from the hybrid cell offer a high efficient power output without using any additional rectification diode. Therefore, the present study will definitely provide a window for various photovoltaic systems to improve the output performance and it will offer a promising approach

for effectively harvesting multi-type energies for realizing multi-functional energy devices.

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wires, as well as applications of nanowires in energy sciences, electronics, optoelectronics and biological science. His discovery and breakthroughs in developing nanogenerators established the principle and technological road map for harvesting mechanical energy from environment and biological systems for powering a personal electronics. His research on self-powered nanosystems has inspired the worldwide effort in academia and industry for studying energy for micro-nano-systems, which is now a distinct disciplinary in energy research and future sensor networks.



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