[Experimental Methods]

The fabrication and characterization of nanogenerators: Flexible substrate (Kapton polyimide film, Model: HN500, DuPont) with the thickness of ~50.8 um was utilized as a substrate for high performance nanogenerator fabrication. First, the flexible substrate was rinsed with acetone, methanol and DI water in series. ITO transparent conducting layer and thin ZnO seed layer were deposited via RF sputtering method. Parts of ITO layer were remained for the direct contact electrode using a shadow mask during ZnO deposition. After then, hydrothermal method was used for the growth of vertically aligned NW array on Kapton substrate using previously reported methods [1]. To grow the nanowires, a solution in DI water was mixed with hexamethylenetetramine (HMTA) and zinc nitrate hexahydrate ($ZnNO_3 \cdot 6(H_2O)$, resulting in ~5 mM solution. The substrate was placed on the top of the solution upside downward and heated up to 80 °C for 16 hours in isothermal oven to grow vertical NW array. The NW grown substrate was rinsed with DI water and blown by nitrogen gas. After then, polymethyl-methacrylate (PMMA) was spin-coated onto the nanowires to enhance the stability and mechanical robustness of the entire structure. Oxygen plasma etching was performed, leaving behind fresh and clean one-side ends on the nanowires. Afterward, the bottom electrode was fabricated on another flexible substrate. Thin Au film was deposited via a RF Sputter method with a shadow mask. Finally, NW array and Au film substrates were assembled and contacted firmly. All characterization of NGs was conducted after connecting the ITO electrode on the substrate with the NWs and the bottom Au film. A mechanical vibration system (Labworks Inc.) served to apply periodic deformation to NGs at desired frequency and speed. Short-circuit current and open-circuit voltage were measured via current and voltage preamplifier, respectively.

To store electric energy from NGs, we first integrated ten NGs in series to achieve enough voltage output, so that we could use a rectification circuit to make it direct current (DC)-like-signal. And supercapacitor/capacitors were integrated in the circuit as storage parts. After actuating NGs with mechanical vibration, the electrochemical properties of the supercapacitors/capacitors were characterized using a potentiostat/galvanostat (Princeton Applied Research VersaStat 3F).

The Number of Effective NWs in Nanogenerators based on Solid or Flexible Substrates

Figure S1 shows schematic diagram depicting two different nanogenerators fabricated on solid substrates or flexible substrates systems. To generate potential difference along ZnO NW, it should be under mechanical stress in a proper way. In this regard, the number of contacted NW between two electrodes will be a key parameter to determine microscopic signal output in nanogenerator. Figure S1a shows previously reported nanogenerator on solid substrates. In case of moderately aligned NW array, only few NWs can make contacts with electrode (Left in Figure S1a) resulting in only few source of electric field between two electrodes (Right in Figure S1a). On the other hand, the number of effective NWs can be enhanced in flexible substrates as it shown in Figure 1Sb. Even NW array has only moderate alignments, top and bottom flexible substrates can be bent to follow the topological end of NWs (Left in Figure 1Sb). Thus, it can enhance the number of contacts between NWs and electrode which makes more source of electric field between two electrodes. As a proof of concept, we observed much enhanced output signal in NG based on flexible substrates. Previous NG fabricated on solid substrate with the same structure showed only voltage of ~96 mV and current of ~9 nA/cm^2, while one on flexible substrates showed voltage of ~300 mV and current of ~100 nA (in Figure 2).

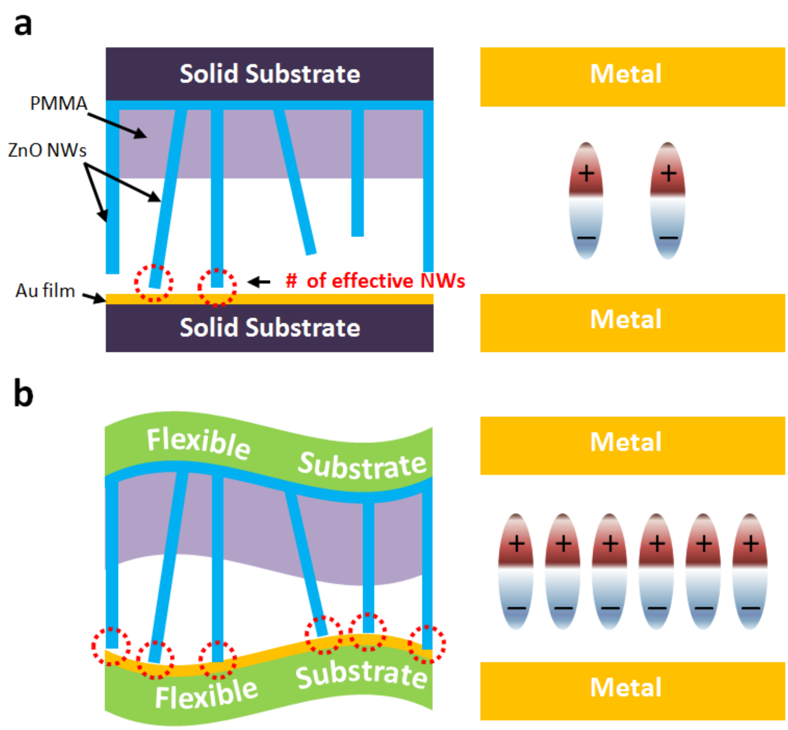

Figure S1

Switching Polarity Test

We have conducted a switching polarity test to rule out any artifact such as coupling with measurement system. The sample with asymmetric voltage output was chosen intentionally for this experiment. First, we measured ten-layer integrated NGs with a voltmeter (Figure S2a), which showed peak-to-peak voltage output from +3.1 V to -1.3 V (Figure S2b). Afterward, the direction of connection of voltmeter and NGs was changed as shown in Figure S2c. The measurement of voltage was conducted again. Peak-to-peak voltage output was observed from +1.4 V to -3.1 V, which is almost exact amount of voltage output except its polarity. This testing method rules out the artifacts such as any capacitance coupling with measurement system because they won't reverse their polarity even we switch the connection mode.

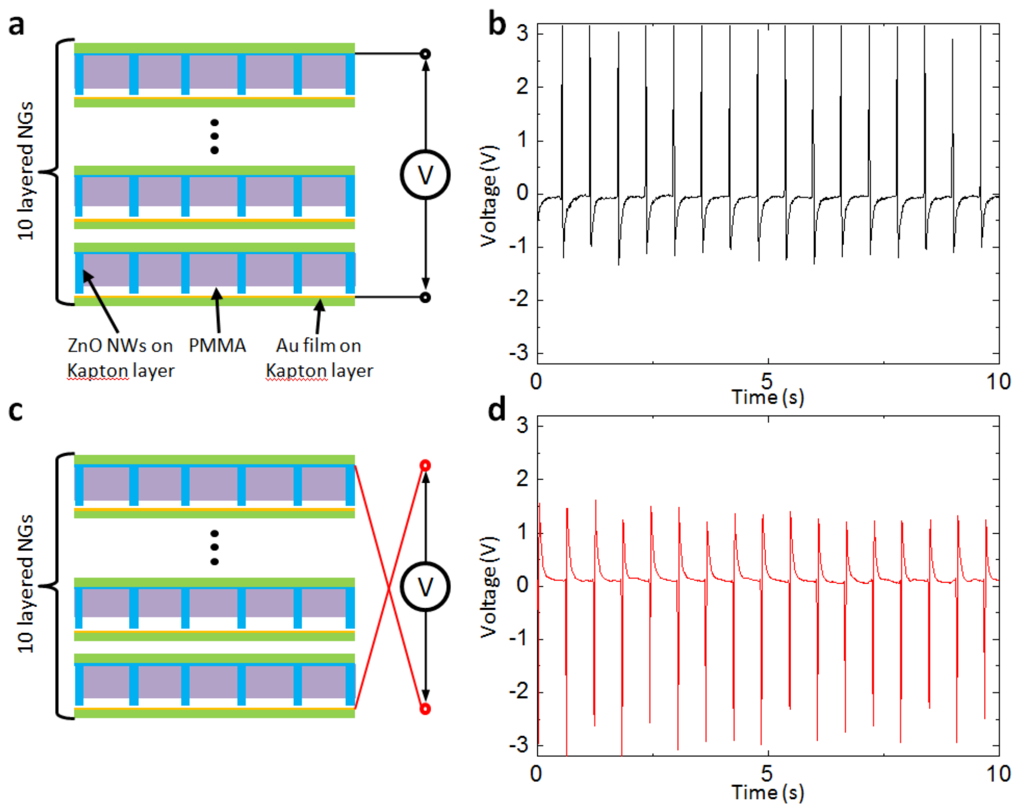

Figure S2

Comparison of Output Current Density:
from nanogenerators based on flexible or solid substrates

Current density outputs from NGs with the employment of different substrates were investigated. Figure S3a and 3b show close-circuit current density outputs from nanogenerators based on solid substrates [ref.1] and flexible substrates, respectively. In the case of a nanogenerator based on solid substrates, a rigid silicon wafer served as substrates for both growth of ZnO NW array and Au electrode (Fig. S3a inset). On the other hand, flexible Kapton layers were employed as substrates in this work (Fig. S3b inset). Figure Aside from flexibility of substrates, both samples were in very similar structure. Solid and flexible substrate-based nanogenerators showed current density outputs of ~8.9 $nAcm^{-2}$ and ~125 $nAcm^{-2}$, respectively. A Plausible explanation is proposed in Figure S1 and the main text as well.

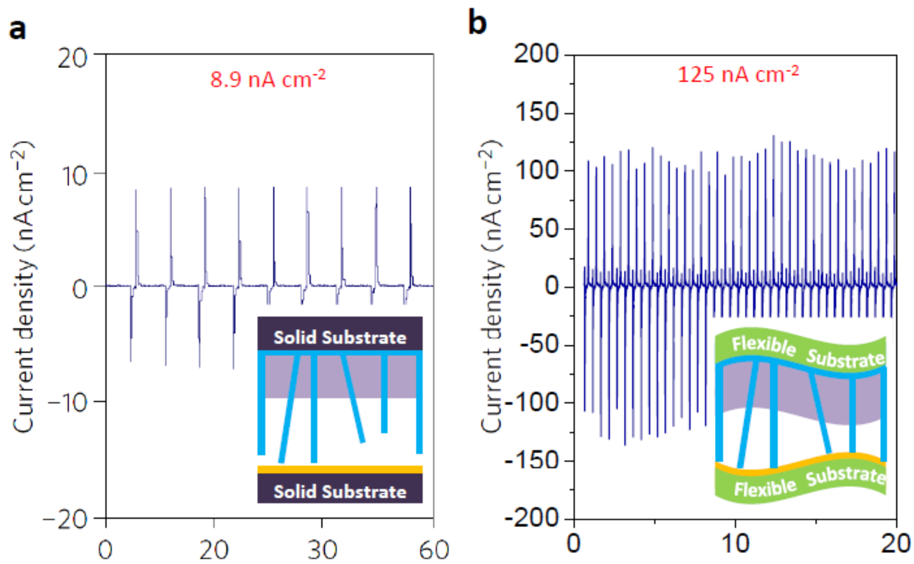

Figure S3. Image (a) adapted from Ref. [1]

Reference

[1] S. Xu, Y. Qin, C. Xu, Y. Wei, R. Yang and Z. L. Wang, *Nat. Nanotechnol.* 2010, **5**, 366.

Electronic Supplementary Material (ESI) for Energy and Environmental Science

Stability of the Au Film on Flexible Substrates

Quality of Au film on a Kapton substrate was investigated to verify stability of flexible nanogenerator. In our fabrication, stability of Au film on flexible substrate can be a crucial because protective PMMA layer was only coated on ZnO NW array. To explore the stability, we applied mechanical vibration to a nanogenerator for ~10 h. After then, the nanogenerator was dissembled and uniformity of Au film on Kapton substrate was observed via an optical microscope. Figure S4 showed a photographic image and optical microscope image of Au film after energy harvesting process. Because of applied mechanical vibration, photographic image showed relatively the uniform color of Au film. On the other hand, in optical microscope image, it showed few cracking areas. Although Au film on flexible substrate is able to serve as conducting film for a proper time, it may need an alternative to achieve a fully sustainable nanogenerator on flexible substrates in a future.

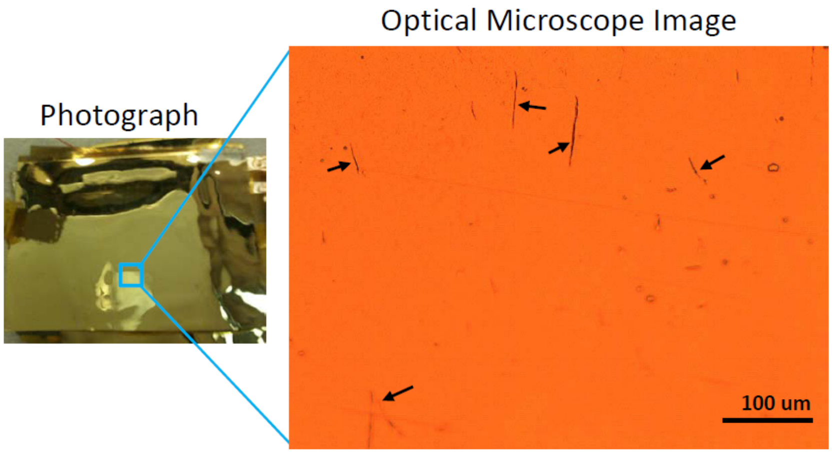

Figure S4