Mitigating the Negative Piezoelectricity in Organic/Inorganic Hybrid Materials for High-performance Piezoelectric Nanogenerators

Huiling Guo, Liang Li, Fang Wang, Sang-Woo Kim, and Huajun Sun*

ABSTRACT: The conversion of ecofriendly waste energy into useable electrical energy is of significant interest for energy harvesting technologies. Piezoelectric nanogenerators based on organic/inorganic hybrid materials are a key promising technology for harvesting mechanical energy due to their high piezoelectric coefficient and good mechanical flexibility. However, the negative piezoelectric effect of the polymer component in composite devices severely undermines its overall piezoelectricity, compromising the output performance of PVDF-based piezoelectric hybrid nanogenerators. Here, to conquer this, we report a two-step poling schedule to orient the dipoles of organic and inorganic components in the same direction. The optimized nanogenerator delivers a combination of high piezoelectric coefficient, great output performance, and remarkable stability. The isotropic piezoelectricity in the composite device collaborates to output a maximum voltage of 110 V and a power density of 7.8 μW cm⁻². This strategy is also applied to elevate the piezoelectricity of other organic/inorganic-hybrid-based nanogenerators, substantiating its universal applicability for composite piezoelectric nanogenerators. This study presents a feasible strategy for enhancing the effective output capability of composite nanogenerator technologies.

KEYWORDS: energy harvesting, piezoelectric nanogenerator, composite film, negative piezoelectricity, polarizing

INTRODUCTION

Energy harvesting technologies are promising solutions toward overcoming the issues of energy consumption, which collect wasted energy from the surrounding environment, convert them into electrical energy, and then power the Internet of Things. Among them, piezoelectric nanogenerators are the dominant choice to harvest mechanical vibration energy owing to their simple configuration and high conversion efficiency. A series of piezoelectric nanogenerator devices have been demonstrated, including piezo-photodetectors, sensors, and self-powered wearable devices. A wide range of piezoelectric materials and their applicability have been elucidated over the past decades, out of which BaTiO₃ has attracted enormous attention due to its superior piezoelectric property and environmental friendliness in comparison to other inorganic piezoelectric materials. However, the challenges for BaTiO₃ associated with rigidity, brittleness, toxicity, low voltage coefficient, and lack of mechanical flexibility limit its commercial application to some extent. Therefore, significant efforts in recent years focus on developing composite piezoelectric nanogenerators based on organic and inorganic hybrid materials. Because piezoelectric polymers, such as PVDF and its copolymers P(VDF–TrFE), feature high piezoelectric coefficients, mechanical flexibility, and biocompatibility, they sever as better alternatives to inorganic piezoelectric materials.

A piezoelectric material converting a mechanical deformation into electrical energy is based on the piezoelectric effect of crystalline structure, which is associated with the noncentrosymmetric nature of crystals. Nearly all materials exhibit no piezoelectric effect because of the irregular arrangement of the electric dipoles within materials. In order to obtain piezoelectricity, applying an electric field is critical to move the molecules freely and to align the electric dipoles in one orientation, commonly at a temperature near the Curie temperature. After the temperature drops to room temperature and the applied electric field is removed, the configuration of aligned dipoles is permanently locked, thereby exhibiting piezoelectric behavior. For BaTiO₃, when the electric field is parallel to the polarization direction, the lattice expands because the positive ions move along the electric field, whereas negative ions move in the opposite direction. This indicates that BaTiO₃ has a positive piezoelectric coefficient (Figure 1a). In contrast, PVDF and P(VDF–TrFE) lattices contract in the direction of the applied electric field, owing to the shrinkage of the intermolecular van der Waals gap under...
the electric field, leading to the negative piezoelectricity.\textsuperscript{30,31} The negative piezoelectric effect within the composite piezoelectric nanogenerators undermines the overall piezoelectric coefficient and, consequently, the output performance. Therefore, mitigating the negative piezoelectricity in organic/inorganic hybrid materials is critical for developing high-performance piezoelectric nanogenerators.

In this sense, we here propose a two-step poling schedule to eliminate the negative piezoelectricity and improve the output performance of composite piezoelectric nanogenerators. As illustrated in Figure 1b, BaTiO\textsubscript{3} is initially polarized through high-temperature annealing under an electric field, and then, P(VDF–TrFE) is polarized under the opposite electric field to acquire an equidirectional piezoelectric coefficient. Different from the conventional structural design and composition regulation,\textsuperscript{32–35} the mitigation of the negative piezoelectricity can effectively improve the piezoelectric property and reduce the energy loss, which enables the high-efficiency output of organic/inorganic composite nanogenerators. Correspondingly, the composite nanogenerator after two-step poling delivers excellent performance in terms of output voltage and power density, together simultaneously with excellent stability.

**EXPERIMENTAL SECTION**

**Fabrication of Piezoelectric Composite Films.** The transparent and homogeneous solution of P(VDF–TrFE) dissolved in N,N-dimethylformamide was prepared by magnetic stirring for 24 h at room temperature. Then, BaTiO\textsubscript{3} nanoparticles with the weight ratios of 5, 10, 15, 20, 25, and 30 wt % were added to the P(VDF–TrFE) solution to prepare the hybrid solution. After stirring for 24 h, the hybrid solution was coated on a smooth glass substrate, followed by heating at 60 °C for 4 h in a drying oven to obtain the composite films. The composite films were then annealed at 135 °C for 24 h to increase the content of the \(\beta\)-phase of P(VDF–TrFE). The prepared films were cut into a required dimension of \(2 \times 2\) cm\textsuperscript{2} to fabricate the nanogenerator device.

**Fabrication of Piezoelectric Nanogenerator Devices.** The top and bottom of composite films were sputtered with Pt electrodes by an ion sputterer to prepare the sandwiched structural devices. Also, two copper wires were fixed by silver paste on the top and bottom sides of the films. Then, a flexible PET substrate with a size larger than the active area was attached to the bottom side by PDMS and dried at 80 °C for 30 min in a drying oven. To obtain a permanent electric dipole orientation, the simultaneous and two-step poling schedules were applied under an external electric field.

**Polarization Steps.** The simultaneous poling procedure was that the piezoelectric nanogenerator was polarized at 40 kV mm\textsuperscript{-1} for 4 h at 50 °C and then cooled to room temperature with the electric field held. The two-step poling procedure was that first the composite film was polarized at 40 kV mm\textsuperscript{-1} for 4 h at 125 °C, and cooled to 110 °C under the electric field. Then, the composite film was further cooled to room temperature without an electric field to only polarize the BaTiO\textsubscript{3} component. Second, this composite film was polarized at 40 kV mm\textsuperscript{-1} for 4 h at 50 °C under an opposite electric field to polarize the P(VDF–TrFE) component to obtain equidirectional piezoelectricity to BaTiO\textsubscript{3}.

**Measurements and Characterization.** The X-ray diffraction (XRD) patterns of the composite films were determined by using a diffractometer (Bruker D8 ADVANCE) with a Cu K\(\alpha\) radiation source. Fourier transform infrared spectrometry (FT-IR) was used to obtain the chemical structure of the composite films and the \(\beta\)-phase proportion of P(VDF–TrFE). Scanning electron microscopy (SEM) was used to characterize the microstructure of composite films after different poling schedules. The differential scanning calorimetry (DSC) was used to measure the melting behavior of composite films, which was carried out from room temperature to 200 °C under a nitrogen atmosphere with a heating/cooling rate of 10 °C/min. The piezoelectric response of samples after different poling schedules was examined by using piezoresistive force microscopy (PFM). An electrometer (Keithley 6514) was used to collect the voltage and current generated by the piezoelectric energy harvester. The applied amplitude is defined as the displacement of test equipment during the bending process, which increases from 1 to 5 mm. A software platform based on LabVIEW was used to realize real-time data acquisition. A digital meter was used for measuring the current values of the composite films. COMSOL Multiphysics 5.4 simulation software was applied to simulate the electric potential distribution for simultaneous and two-step polarized devices. Specifically, the piezoelectric block with a size of \(85 \mu\)m × \(85 \mu\)m × \(60 \mu\)m is modeled as the representative of the three-dimensional device, and BaTiO\textsubscript{3} is evenly dispersed in P(VDF–TrFE) bulk. For the simultaneous polarized device, the device is polarized along the \(z\)-axis. For the two-step polarized device, the BaTiO\textsubscript{3} is polarized along the \(z\)-axis while P(VDF–TrFE) is polarized along the \(−z\)-axis. With the lower electrode set to ground (0 V), a pressure of 0.1 MPa was loaded on the upper surface to simulate the potential distribution.

**RESULTS AND DISCUSSION**

In this study, the raw piezoelectric composite nanogenerator (NG) is prepared by the solution casting method without follow-up polarizing. The baseline composite nanogenerator (S-NG) is polarized under 50 °C, and an electric field is applied to simultaneously acquire piezoelectricity in BaTiO\textsubscript{3} and P(VDF–TrFE). Also, the two-step polarized composite nanogenerator (TS-NG) initially gets the piezoelectricity in BaTiO\textsubscript{3} under an electric field and at a temperature above 125 °C, which is higher than the Curie temperatures of BaTiO\textsubscript{3} (120 °C) and P(VDF–TrFE) (106 °C). This indicates that both BaTiO\textsubscript{3} and P(VDF–TrFE) can be polarized. However, during the cooling process, from 110 °C to room temperature, the absence of the electric field leads to the depolarization of P(VDF–TrFE). Then, the opposite electric field is applied to the composite film to polarize P(VDF–TrFE), ensuring its polarization direction same as BaTiO\textsubscript{3}. The XRD patterns shown in Figure 2a show that all three composite films consist of crystalline P(VDF–TrFE) and BaTiO\textsubscript{3}.\textsuperscript{36,37} Specifically, the
intensities of all diffraction peaks of composite films increase after simultaneous polarizing and two-step polarizing, implying that P(VDF-TrFE) and BaTiO$_3$ crystallize from the nonpolar phase to the polar phase, respectively. For TS-NG, the peaks at 40 and 46° corresponding to the β-phase of P(VDF-TrFE) show an increase in the relative peak intensity after two-step polarizing, indicating the enhancement of the β-phase of P(VDF-TrFE) in the composite film. The β-phase proportion is assessed to be 85.8, 91.8, and 89.7% for NG, S-NG, and TS-NG, respectively, based on FT-IR (Figure 2b) and eq S1 (Supporting Information). This result indicates that the two-step polarizing has little impact on the stabilization of the β-phase of P(VDF-TrFE). This contributes to the piezoelectricity and the associated output electric power of the energy harvesting material. The increases in the crystallinity and polarity are confirmed by the melting behavior of composite films. As shown in Figure 2c, the Curie temperature of P(VDF-TrFE) in TS-NG is observed at 110 °C upon cooling, which is the highest as compared to pristine NG (107 °C) and S-NG (108 °C). The high Curie temperature for TS-NG is attributed to the enhanced dipole moment within P(VDF-TrFE) arising from a more regular and well-aligned chain packing. In addition, the melting and crystallization temperatures for TS-NG shift to higher temperatures compared to the other two films, indicating the formation of a polar polymer with higher polarity in the composite film after two-step poling.

The piezoelectricity of material is closely relative to the magnitude of dipole moment, which can be directly evaluated by the value of the piezoelectric coefficient ($d_{33}$) of the polar material. As shown in Figure 2d, the NG film shows a very small $d_{33}$ value (3.9 pC N$^{-1}$) because of the counterbalance of dipoles lying in irregular directions in BaTiO$_3$ and P(VDF-TrFE). The piezoelectric coefficient is strongly dependent on the BaTiO$_3$ content in the composite film (Figure S1a, Supporting Information). As for S-NG film, the piezoelectric coefficients are all negative (set the direction of the applied electric field to positive) after simultaneous polarizing and reach up to the maximum value of $-29.2$ pC N$^{-1}$ with 15 wt % BaTiO$_3$. This result suggests that P(VDF-TrFE) contributes the large majority of the piezoelectricity but offsets the piezoelectric effect of BaTiO$_3$ because of the negative piezoelectricity of P(VDF-TrFE) originating from their reduced lattice dimensionality associated with highly aniso-

Figure 2. (a) XRD patterns of NG, S-NG, and TS-NG. “B” and “P” represent the diffraction peaks of BaTiO$_3$ and P(VDF-TrFE), respectively. (b) FT-IR spectrum of NG, S-NG, and TS-NG composite films. (c) DSC curves for NG, S-NG, and TS-NG. “Tc”, “Tm”, and “Tcry” represent the Curie transition temperature, melting temperature, and crystallization temperature, respectively. (d) Comparison of $d_{33}$ values of NG, S-NG, and TS-NG films. (e) $d_{33}$ values of our TS-NG film and previously reported nanogenerators. (f) Change of $d_{33}$ value after two-step poling for representative organic/inorganic hybrid films. 

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tropic bonding arrangement. In contrast, the TS-NG film displays positive piezoelectric coefficients after both first-step and second-step polarizing (Figure S1b, Supporting Information), indicating that the only BaTiO$_3$ is polarized initially and then P(VDF–TrFE) is polarized to dedicate to the overall piezoelectricity. P(VDF–TrFE) dominates the piezoelectric performance because almost more than 90% of piezoelectricity is from P(VDF–TrFE) (Table S1, Supporting Information). Indeed, as the content of BaTiO$_3$ particles increases, excessive weight ratios of BaTiO$_3$ particles may bring about particle agglomeration in the polymer matrix, which lower the contribution to piezoelectricity of composite film. Therefore, TS-NG film with 15 wt % BaTiO$_3$ exhibits a larger piezoelectric coefficient of 38.8 pC N$^{-1}$, which is higher than that of S-NG film by 9.6 pC N$^{-1}$. Also, this value is larger than most $d_{33}$ values of organic/inorganic hybrid materials reported in the previous literature (Figure 2f and Table S2, Supporting Information), which contributes to the improvement in the output performance of piezoelectric nanogenerator. In addition, as shown in Figure 2e, this two-step poling strategy was also conducted for other organic/inorganic hybrid-based piezoelectric nanogenerators (Table S3, Supporting Information), such as PVDF/Ba$_{0.85}$Ca$_{0.15}$Zr$_{0.1}$Ti$_{0.9}$O$_3$ (BCZT), PVDF/PbTiO$_3$–PbZrO$_3$ (PZT), PVDF/Pb(Mg$_{1/3}$Nb$_{2/3}$)$_3$O$_5$–PbTiO$_3$ (PMN-PT), and P(VDF–TrFE)/Na$_{0.5}$K$_{0.5}$NbO$_3$ (KNN). These results indicate that all $d_{33}$ values after two-step poling are larger than those after simultaneous poling, demonstrating the broad applicability of this approach for composite nanogenerators.

The microstructure of composite films after different poling schedules is examined by using SEM. As presented in Figure S2 (Supporting Information), NG film displays a highly densified structure before poling. Also, BaTiO$_3$ has uniformly dispersed across the P(VDF–TrFE) matrix without particle agglomeration, which is consistent with the elemental mapping scan (Figure S3, Supporting Information). However, after poling, both S-NG and TS-NG films exhibit a great number of pores owing to the orientation of P(VDF–TrFE) during the poling process. The porosity calculated from BET analysis further confirms that the specific surface area of NG increases from 135.2 to 253.9 m$^2$ g$^{-1}$ for S-NG and to 260.5 m$^2$ g$^{-1}$ for TS-NG (Figure S4, Supporting Information). Moreover, the porosity and pore diameter of TS-NG are close to S-NG, demonstrating that the two-step poling schedule has little impact on the microstructure and only improves the crystallinity of P(VDF–TrFE). Therefore, the influence of the triboelectric effect on the output behavior of composite film is not taken into account. Also, we used a plasma sputter to deposit platinum electrodes to obtain a tight attachment between the electrode and the surface of the film. The enlarged SEM image shows that there is no obvious gap between electrode and surface (Figure S5, Supporting Information), indicating that no triboelectricity is produced at the surface of films.

**Figure 3.** (a) PFM phase images and corresponding (b) phase histograms of NG, S-NG, and TS-NG. (c) Piezoelectric potential distribution of S-NG and TS-NG. (d) Stress distribution of composite devices.
To investigate the local piezoelectric response of composite films, the PFM was used to clarify the piezoelectricity direction and piezoresponse phase signals. As shown in Figure 3a, the dark region of the film represents the domains that are oriented upward, indicating the same polar direction as the applied electric field. The unpoled film presents many randomly distributed domains, and the corresponding phase histogram in Figure 3b exhibits an almost equal amount of the out-of-phase domain. This result indicates that NG film will not show the piezoelectric effect. When the composite film is under a bias, P(VDF-TrFE) and BaTiO$_3$ display different domain orientations which are 180° out of phase because of the opposite polarization. Therefore, the phase histogram of S-NG comprises domains with positive orientation stemming from the polarized P(VDF-TrFE) with an opposite piezoelectric coefficient. Also, the other domains still exhibit negative orientation due to the high alignment of BaTiO$_3$ in composite films after simultaneous poling. A theoretical investigation of the piezoelectric potential distribution of composite films after simultaneous and two-step polarizing was performed using COMSOL Multiphysics software. As shown in Figure 3c, after a force is applied to the upper surface of films, S-NG film generates a negative piezoelectric potential because the piezoelectric effect of BaTiO$_3$ is offset by P(VDF-TrFE) with negative piezoelectricity. However, the TS-NG film shows a positive potential under the same compressive force, due to the synclastic piezoelectricity of BaTiO$_3$ and P(VDF-TrFE). Moreover, the generated piezoelectric potential of TS-NG (8.94 V) is higher than that of the S-NG (5.68 V) device, which is consistent with the following output voltage. In addition, when loading the pressure, the strain is concentrated in the interface between BaTiO$_3$ and P(VDF-TrFE) for devices after both simultaneous and two-step poling (Figure 3d).

Due to the mitigation of negative piezoelectricity of P(VDF-TrFE), superior output performance can be acquired for a composite nanogenerator. As shown in Figure 4, using the P(VDF-TrFE) copolymer as the bulk of piezoelectric material, BaTiO$_3$ particles with different contents are incorporated to measure the output voltage and current. Both output voltage and current of nanogenerators after simultaneous and two-step polarizing are initially increased and then decreased with the increasing BaTiO$_3$ content. Specifi-
Figure 5. Output voltage of (a) S-NG and (b) TS-NG under variable loading resistances. Evolution of voltage and current of (c) S-NG and (d) TS-NG as a function of loading resistances. The power density of (e) S-NG and (f) TS-NG under variable loading resistances.

Figure 6. Photographs of (a) wrist bending and (d) mouse clicking. The generated voltage signal by (b,c) wrist bending and releasing and by (e,f) slow and fast finger tapping the TS-NG device. (g) Mechanical motion of original and bending stages of the device to drive light-emitting diodes. (h) Current stability test analysis of the TS-NG device.
current of the TS-NG film with the same content of BaTiO₃ are 110 V and 2.8 μA, respectively, which are nearly triple those of S-NG. This result indicates that the polarization direction of BaTiO₃ is the same as that of P(VDF-TrFE) after two-step polarizing and the output performance of the composite nanogenerator can be significantly improved. Moreover, as the applied amplitude increases from 1 to 5 mm (Figure S6, Supporting Information), the output voltage and current both gradually increase for S-NG and TS-NG. Also, the increasing trend of output current is a little behind that of output voltage (Figure S7, Supporting Information), indicating that the output voltage is sensitive to the applied strain, compared with the output current. Also, TS-NG always exhibits higher values of voltages and currents than S-NG, no matter what amplitude is applied. It is noteworthy that the piezoelectric voltage and current signals exhibit an alternative output behavior with alternating voltage and current (Figure 4g) because of the alternating input nature of the external mechanical force.⁴⁴ Moreover, the piezoelectric potential is created by the change of applied mechanical stress. As shown in Figure 4h, during the bending process, the nanogenerator is subjected to compressive stress, thus generating a positive piezoelectric potential by its contraction. However, when the arc structure started to bounce back after releasing the force, the nanogenerator undergoes tensile stress.⁴⁵ Therefore, the device displays a negative piezoelectric output voltage. At the same time, the electrons return to maintain the charge balance, leading to the negative electrons flowing through the collector.

The output power of the composite film is tested with different load resistances at 5 mm amplitudes bending force with a frequency of 1 Hz (Figure 5). The measured circuit for output voltage and current under different load resistances is shown in Figure S8 (Supporting Information). The voltage increases along with the increase of load resistance for both S-NG and TS-NG, reaching up to 32 and 105 V under a load resistance of 1.5 GΩ, respectively, further confirming the high output performance of the two-step polarized film. However, the current steadily decreases as the load resistance increases from 0.9 to 0.03 μA for S-NG, and from 2.05 to 0.1 μA for TS-NG. Therefore, the areal power density calculated based on eq S2 (Supporting Information) is shown in Figure 5e.⁴⁵,⁴⁶ Obviously, TS-NG achieves a maximum power density of 7.8 μW cm⁻² at a load resistance of 100 MΩ (RMS is 3.7 μW cm⁻²), which is 10 times higher than that of S-NG (0.6 μW cm⁻², RMS is 0.37 μW cm⁻²). The RMS voltage and power density are displayed in Figure S9 (Supporting Information). Indeed, the power density of the piezoelectric nanogenerator is highly dependent on the orientation of dipoles. After two-step poling, the piezoelectric dipoles of BaTiO₃ and P(VDF-TrFE) are oriented in the same direction. Thus, the TS-NG nanogenerator delivers a high output power owing to the collaboration between BaTiO₃ and P(VDF-TrFE).

To demonstrate the high-power property of TS-NG in practical application, we utilize a TS-NG nanogenerator to harvest human motions such as wrist bending and mouse clicking. As shown in Figure 6a–f, the TS-NG nanogenerator can generate a voltage of ≈1.5 V during wrist bending and releasing. Moreover, when the finger is tapping on the mouse, the output voltage of the TS-NG nanogenerator reaches up to 3.4 V with slow tapping and 3.7 V with fast tapping. The improvement of output may be attributed to the large contact area and force between TS-NG film and finger. Moreover, the output voltage of the assembled sensor with an area of 2 × 2 cm² reaches as high as 90 V, which is sufficient to light up more than 60 LEDs when the film is subjected to bending force (Figure 6g and Video S1, Supporting Information). These results further demonstrate the practicability of this device based on the two-step polarizing strategy. Moreover, the output performance of the TS-NG nanogenerator exhibits little decay even after 6000 s (at a frequency of 1 Hz), especially for the output current, indicating excellent stability and remarkable flexibility (Figures 6h and S10, Supporting Information). Moreover, there is no formation of cracks and defects on the TS-NG film after 6000 times of bending-releasing cycling (Figure S11, Supporting Information), demonstrating that the durability of the device in terms of cycle stability is outstanding.

### CONCLUSIONS

In summary, we report a universal strategy to enhance the output performance of organic/inorganic hybrid piezo-nanogenerators through a two-step poling schedule, which can effectively orient the direction of piezoelectricity of organic components, same as the inorganic component. This method enables excellent output capability in the P(VDF-TrFE)/BaTiO₃ composite device with an output voltage of 110 V, power density of 7.8 μW cm⁻², and superior flexibility. Finally, the homemade nanogenerator device is effective for harvesting energy from wrist bending and finger pressing. Moreover, the composite device after two-step poling exhibits high stability after 6000 cycles without obvious debilitation, which can still drive 60 commercial LEDs. This work provides a feasible approach for improving the output performance of composite piezoelectric nanogenerators for highly effective energy conversion.

### ASSOCIATED CONTENT

#### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsami.2c08162.

Piezoelectric coefficient of the composite film with different BaTiO₃ contents; table of piezoelectric coefficients in the literature; table of piezoelectric coefficients of different composite films; SEM images; EDS mapping images; nitrogen adsorption and desorption isotherms; and pore diameter distribution (PDF)

Lighting up of 60 LEDs driven by the piezoelectric nanogenerator based on the two-step polarizing strategy (MP4)

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Notes
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