

A Method for Quantitatively Separating the Piezoelectric Component from the as-received "Piezoelectric" Signal

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Article

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Abstract

Polymer-based piezoelectric devices are promising for developing future wearable force sensors, nanogenerators, and implantable electronics etc. The electric signals generated by them are often assumed as solely coming from piezoelectric effect. However, triboelectric signals originated from contact electrification between the piezoelectric devices and the contacted objects can produce non-negligible interfacial electron transfer, which is often combined with the piezoelectric signal to give a triboelectric-piezoelectric hybrid output, leading to an exaggerated measured “piezoelectric” signal. Herein, a simple and effective method is proposed for quantitatively identifying and extracting the piezoelectric charge from the hybrid signal. The triboelectric and piezoelectric parts in the hybrid signal generated by a poly(vinylidene fluoride)-based device are clearly differentiated, and their force and charge characteristics in the time domain are identified. This work presents an effective method to elucidate the true piezoelectric performance in practical measurement, which is crucial for evaluating piezoelectric materials fairly and correctly.

Background

Mechanical energy sources are almost everywhere in the surrounding environment, *e.g.* wind, wave, and human motion. Harvesting these energies from the environment, so as to provide electricity to smart terminals and other equipment, has become an attractive and competitive way-out in the era of Internet-of-Things¹⁻⁶. In recent years, piezoelectric nanogenerators (PENGs) and triboelectric nanogenerators (TEGs) as new kinds of energy harvesters have attracted much attention, especially in the field of wearable devices⁷⁻¹³. The polymer-based PENGs and TENGs are even more attractive because they are flexible, light-weighted, and easy-to-process, which can be used to scavenge mechanical energy at multi-scales especially from human motion¹⁴⁻¹⁹.

Contact electrification (CE) as a universal electron transfer phenomenon that can occur between any material in different physical states (solid, liquid, and gas)²⁰. It is often coupled with electrostatic induction to generate electric signals in the process of repeated contact and separation of two different materials, which is the working mechanism of TENGs. Recently, Sutka *et al.* confirmed that CE can take place in piezoelectric devices during a variety of motions, including interfacial shear friction, nanofiller slippage, pre-existing static charge and contact friction²¹. Simultaneously, the existence of triboelectric signals generated by the contacted objects and piezoelectric devices can be strong and shouldn't be ignored, especially under repeated pressing. However, some previous works ignored this kind of triboelectric signal and regarded the output from piezoelectric devices as the sole piezoelectric signal, leading to exaggerated performance. The lack of a reliable method to differentiate piezoelectric signals and triboelectric signals obstructs us reveal the influence of triboelectric signals and evaluate the true contribution of the piezoelectric effect, which results in incorrect evaluation on piezoelectric performance. More specifically, because TENG and PENG have the same output including voltage, current, and charge, traditional analysis on electric signals cannot identify their respective part in a hybrid output. Therefore,

developing a method to identify and extract the piezoelectric component from a “piezoelectric” signal is particularly important for quantitatively evaluating the performance of piezoelectric materials.

In the present work, we analyzed the CE process that happened between loading objects and the encapsulating materials, and developed an effective method to identify the piezoelectric part from a hybrid triboelectric-piezoelectric output in a sandwich-structured poly(vinylidene fluoride) (PVDF)-based piezoelectric device system. Based on our experimental observation, the triboelectric signals can take a large portion, which should not be ignored. Here we investigated the force-time curve of the piezoelectric process as a strong supplement rather than solely analyzing the electric signals, which is the source of the electric signals. By comparing these two curves simultaneously, it can be found that before and after the device-object contact, the recorded electric signals belong to the triboelectric contribution, and the electric signals acquired when they are contacted can be ascribed to the piezoelectric effect. Then the piezoelectric charge transfer was quantitatively extracted from the hybrid output and the piezoelectric coefficient (d_{33}) was calculated, such results were consistent with those measured by other methods, which are elucidated as follows. In such way, we can conveniently separate the combined signals clearly, and the accuracy is only limited by both force-time and charge-time resolutions.

Results

1. Piezoelectric-triboelectric hybrid output in piezoelectric devices

Figure 1a shows the schematic of the PVDF-based piezoelectric device in pressure sensing. CE can occur between the human finger and the piezoelectric device—the human finger would have positive charges and the protective layer of the device would have negative charges. As a result, a single-electrode triboelectric nanogenerator (SE-TENG) will form when the human finger is approaching the piezoelectric device again. Its working mechanism is illustrated in Fig.1b, including four typical stages, such as contacted, separating, separated, and contacting. Among these stages, the triboelectric signals are generated in the stages of contacting and separating. On the other hand, the PENG will be formed when the human finger is pressing the piezoelectric devices (right in Fig.1a), and its working mechanism is clearly illustrated in Fig.1c. The piezoelectric signals would be generated in the stages of pressing and releasing. Therefore, the SE-TENG and PENG exist during the practical application of piezoelectric devices, resulting in the triboelectric-piezoelectric hybrid output.

2. Verification of the existence of the SE-TENG system in piezoelectric devices

In order to verify the existence of the SE-TENG system during the measurement of piezoelectric devices, here we replaced the PVDF film with a non-piezoelectric material polyimide (PI) for investigation. This means that the interference from piezoelectric signals was eliminated and thus only triboelectric signals will exist during the measurement. The testing platform is illustrated in the experimental section and Supplementary Fig. 1. During the measurement, two different wiring methods were adapted including forward connection and reverse connection so as to verify whether the electric signals were generated by

the testing system itself. More specifically, if there were not any interference from other signals, the electric signals generated by the device would symmetrically flip with constant amplitude when the connection direction was switched^{22, 23}.

Figure 2a shows a schematic diagram of measuring electric signals between the front electrode and back electrode of the PI-based device. The open-circuit voltage (Fig. 2d) and transferred charge (Fig. 2g) curves show significant electric signals in the forward connection, while the electric signals obtained in the reverse connection are very low. Thus the electric signals in the forward and reverse connection are not symmetrical along the x-axis, suggesting they are not generated by the testing system composed of the front and back electrode. This is because the Kapton layer with negative charges can induce several times more positive charges in the front electrode than the back electrode. In the forward connection, the Al plate, Kapton tape layer, and the front electrode form a SE-TENG, then triboelectric signals can be detected when connecting the signal probe with the front electrode (Supplementary Fig. 2a). By contrast, positive charges in the front electrode would flow to the ground in the reverse connection and thus weaker electric signals can be detected in the back electrode (Supplementary Fig. 2b).

To verify that CE did occur between the Al plate and the Kapton layer, two kinds of contact-separation triboelectric nanogenerators (CE-TENGs) were tested (CS-TENG-1 in Fig. 2b and CS-TENG-2 in Fig. 2c). For CS-TENG-1, it is composed of Al plate, Kapton layer, and front electrode. The voltage signals (Fig. 2e) generated by CS-TENG-1 in the forward connection and in the reverse connection are symmetrical along the x-axis. More specifically, the signs of voltage signals obtained in two connections are opposite and the amplitudes of the voltage signals are constant. The charge signals (Fig. 2h) of CS-TENG-1 have the same characteristics with voltage signals. For CS-TENG-2, it is composed of Al plate, Kapton layer, front electrode, PI film, and back electrode. Similarly, the voltage signals (Fig. 2f) of CS-TENG-2 obtained in the forward connection and in the reverse connection are symmetrical along the x-axis; the charge signals (Fig. 2i) of CS-TENG-2 obtained in the forward connection and in the reverse connection are symmetrical along the x-axis too. The above results indicate that the electric signals are generated by CS-TENGs where the Al plate and the Kapton layer constitute the triboelectric pairs, and their working mechanism is illustrated in Supplementary Fig. 3. Therefore, the SE-TENG is confirmed by the experiments, which is consistent with the theoretical analysis.

It is worth noting that the peak voltage (~ 1.5 V) and charge transfer (~ 1.1 nC) generated by SE-TENG are too large compared with polymer-based piezoelectric materials (such as PVDF, the peak voltage of them usually is several volts^{24, 25}). Simultaneously, using a finger to tap the SE-TENG can generate larger electric signals (Supplementary Fig. 4, up to ~ 7 V and ~ 4 nC). As a result, these triboelectric charges can significantly influence the measurement of piezoelectric signals, which shouldn't be ignored.

3. Involving force signals to differentiate the triboelectric signals and piezoelectric signals

It is essential to differentiate the triboelectric signals and piezoelectric signals before quantitatively obtaining the piezoelectric charge transfer and evaluating the piezoelectric performance. Here we developed an effective method that can find the sources of the resulting electric signals by analyzing the

force loading process of piezoelectric devices. And this simple method was demonstrated by successfully identifying the sole triboelectric signals and piezoelectric signals.

The sole triboelectric signals are generated by the PI-based device (Fig. 3a). The sole piezoelectric signals are generated by the PVDF-based device with a conductive shielding layer (Fig. 3b). In addition, the role of shielding layer is illustrated in Supplementary Fig. 5; the PI-based device with a shielding layer (Supplementary Fig. 5a) cannot generate any electric signals (Supplementary Figs. 5c, e), while the PVDF-based device with a shielding layer (Supplementary Fig. 5b) can generate symmetrical pure piezoelectric signals in the forward and reverse directions very well (Supplementary Figs. 5d, f).

According to the force signal (Fig. 3i), the sole triboelectric signal generation process can be divided into three stages including contacting, contacted, and separating. In the contacting stage, the Al plate is contacting with the PI-based device. At the same time, the short-circuit current I_{SC} (Fig. 3c) and charge curve (Fig. 3g) fall gradually, while the open-circuit voltage V_{OC} signal (Fig. 3e) rises slowly. When Al plate and the PI-based device are contacted with each other, the electrostatic balance between them is formed; the I_{SC} signal backs to the baseline quickly, while the V_{OC} and transferred charge signals remain unchanged. In the separating stage, the Al plate is separating from the PI-based device, which breaks the electrostatic balance that existed before, resulting in the signal rise of the I_{SC} . Besides, the V_{OC} and transferred charge signals go back to the baseline gradually. Differently, the I_{SC} can reach to peak when the Al plate and the PI-based device are separated, and then falls to the baseline. Overall, there is a phase difference between the sole triboelectric signal and force signal.

By contrast, according to the force signal (Fig. 3j), the sole piezoelectric signal only occurs in the contacted stage (light green area) and its generation process can be divided into three stages including compressing, compressed, and releasing. In the compressing stage, the PVDF film experiences a repaid elastic deformation and thus the I_{SC} (Fig. 3d) and charge signals (Fig. 3h) reach to peak dramatically, while the V_{OC} (Fig. 3f) falls to the lowest point. In the compressed stage, the elastic deformation of PVDF film reaches the maximum value, indicating that the piezoelectric effect would not be activated further. As a result, the I_{SC} cannot increase further and falls to the baseline; the V_{OC} and charge signal remain unchanged. In the releasing stage, the elastic deformation of PVDF film is releasing rapidly. The V_{OC} and charge signals go back to the baseline, while the I_{SC} decreases to the lowest point when the elastic deformation is released and then rises back to the baseline. More importantly, the V_{OC} and the charge signals have the same trend with the force curve, and there are no phase differences existed. Therefore, analyzing the force signal of devices is an effective method for differentiating triboelectric signals and piezoelectric signals.

4. Identifying the piezoelectric signal in “piezoelectric” output

Above results and discussion demonstrated that the effectiveness of differentiating sole triboelectric signals and sole piezoelectric signals by analyzing the force loading curve. Now we can identify the piezoelectric signals from the “piezoelectric” output. Two measurements were carried out by taking into account the direction difference between triboelectric signals and piezoelectric signals. This is because,

the direction of triboelectric signals is determined by the triboelectric series and keeps constant²⁶, and that of piezoelectric signals depends on the polarization direction of signal acquisition side.

Figure 4a is a schematic of measuring electric signals from the negative polarization side of the PVDF film. The signal generation process of the hybrid output can be divided into six parts, including contacting, contacted, compressing, releasing, released, and separating (Fig. 4b). Figure 4c shows the sole triboelectric signals and corresponding force signal of the PI-based device; Figure 4d shows the sole piezoelectric signals obtained in the negative polarization side of PVDF-based device with a shielding layer and its corresponding force signal; Fig. 4e shows the triboelectric-piezoelectric hybrid signals generated by the PVDF-based device. By manipulating the direction of the piezoelectric signals to be opposite with the triboelectric signals, the turning points (⊠ and ⊞) of electric signals can be observed in Fig. 4e. The existence of turning points not only differentiates the piezoelectric part (light green area) and triboelectric part (light yellow area) in the hybrid output, but also proves the difference between piezoelectric signals and triboelectric signals in the time domain which is discussed before. Therefore, the hybrid output can simply be regarded as the sum of the sole triboelectric signals and the piezoelectric signals.

However, if flipping the piezoelectric device and collect the hybrid output from the positive polarization side of the PVDF film (Supplementary Fig. 6a), things are different (Supplementary Fig. 6b). The hybrid output (Supplementary Fig. 6e) can still be regarded as the sum of sole triboelectric signals (Supplementary Fig. 6c) and sole piezoelectric signals (Supplementary Fig. 6d), while there are no turning points in the stages of contacted and released due to the direction of piezoelectric signals is the same as triboelectric signals.

Overall, by investigating the force loading signal of the devices, it's very convenient to determine whether the resulting voltage signal is the triboelectric signal, the piezoelectric signal, or hybrid signal from the perspective of the time domain, while traditional voltage signal analytical methods cannot do.

Extracting the piezoelectric charge transfer from “piezoelectric” output for piezoelectric performance evaluation

For separating piezoelectric charge, a compressed balance analysis (CBA) method is used to extract piezoelectric charge transfer from the hybrid output. Specifically, as shown in Fig. 5a (⊠), there is an electrostatic balance when the piezoelectric device is at the compressed stage. Q₃ is the transferred charge in the Al plate, -Q₂ is the transferred charge in the Kapton layer, q is the total charge transfer in the electrode which contains both piezoelectric and triboelectric parts (Fig. 5b), Q₁ is the induced piezoelectric charge. Their relationship is:

$$Q_3 - Q_2 + q - Q_1 = 0 \quad (1)$$

After flipping the device (⊞ in Fig. 5a), the electrostatic balance becomes:

$$Q_3 - Q_2 - q' + Q_1 = 0 \quad (2)$$

Where $-q'$ is the total charge transfer in the electrode (Fig. 5c). By making Eq. (1) – Eq. (2):

$$Q_1 = (q + q')/2 \quad (3)$$

As shown in Eq (3), the total induced piezoelectric charge Q_1 can be calculated by substituting the values of q and q' into expression without considering the interference from triboelectric charges. Furthermore, the true piezoelectric coefficient d_{33} of PVDF film can be obtained by Eq (4), where F represents the applied force.

$$d_{33} = Q_1/F \quad (4)$$

The transferred charge curves measured in the positive and negative polarization direction of the PVDF-based device under different forces are shown in Supplementary Figs. 7a, b, respectively. Additionally, the piezoelectric charge transfer can be extracted directly from the hybrid output by calculating the relative value in the stage of compressing (Supplementary Figs. 7c, d, and Note 1), which is called directly extracting (DE) method. In order to verify the effectiveness of above two methods, an outside shielding (OS) method is introduced to obtain the sole piezoelectric charge transfer (details are shown in Supplementary Fig. 8 and Note 2), which can be regarded as a benchmark because the triboelectric part is eliminated. Figure 5d shows the charge-force curves obtained by the above three methods. Importantly, the calculated d_{33} of CBA is 33.74 pC/N and that of DE is 32.64 pC/N, which are nearly consistent with the shielded device (33.20 pC/N). Simultaneously, the transferred piezoelectric charges calculated by these three methods in different force loading are almost equal (Fig. 5e). Furthermore, a commercial piezoelectric meter is also introduced to measure the d_{33} of the PVDF film (Supplementary Fig. 9). The average d_{33} measured by this instrument is 33.05 pC/N, which is consistent with the aforementioned results, indicating that these three methods are both effective. By contrast, the fake piezoelectric performance would be always obtained when just using the apex or bottom of electric signals that contain the triboelectric part and piezoelectric part as the evaluation index. Therefore, our method CBA can be used for separating the piezoelectric component from the triboelectric-piezoelectric hybrid output, and evaluating piezoelectric performance without influenced by triboelectric signals.

Discussion

In summary, this work elucidates the output signal composition of conventional piezoelectric devices, and develops an effective method to quantitatively extract the piezoelectric part from a hybrid output regardless of the interference from triboelectric signals caused by CE. For a typical sandwich-structured PVDF device, an SE-TENG and a PENG both exist during the compressing test, resulting in the triboelectric-piezoelectric hybrid output. Our method that analyzes the force signals of PVDF-based devices can differentiate the triboelectric part and piezoelectric part from the hybrid output in the time domain. More specifically, the triboelectric part occurs before and after the contact between the object and device, while the piezoelectric part only exists when the object and device are contacted. The

piezoelectric charge transfer can be extracted from hybrid output using CBA method that analyzes the electrostatic balance among the Al plate, Kapton layer, electrodes, and PVDF film. Besides, the d_{33} calculated by CBA is 33.74 pC/N, which is consistent with the results of OS (33.20 pC/N) and DE (32.64 pC/N). It is also verified by a commercial instrument, indicating that both of these three methods are effective. This research gives a deep insight on the output composition of piezoelectric devices in practical measurement and provides a simple method to separate the piezoelectric component from a hybrid output. It would help future researchers to evaluate true piezoelectric performance of flexible piezoelectric devices, which is beneficial for developing next-generation high-performance piezoelectric materials and promoting their application in wearable electronics.

Methods

Fabrication of PVDF-based piezoelectric devices: A commercial poled PVDF film with silver electrode (110 μm , Measurement Specialties Inc.) was encapsulated by the Kapton tape (50 μm). The surface area of PVDF-based device was $3\times 3\text{ cm}^2$.

Fabrication of PI-based non-piezoelectric devices: A polyimide (PI) film with a thickness of 125 μm was coated by silver paste on both two sides. Then, the Ag electrodes were obtained after sintering at 160 $^{\circ}\text{C}$ for 10 mins. Finally, it was encapsulated using a Kapton tape (50 μm) for a PI-based non-piezoelectric device ($3\times 3\text{ cm}^2$).

Measurements: The piezoelectric and non-piezoelectric devices were compressed repeatedly by a linear motor (LinMot E1100) to generate electric signals. An aluminum (Al) plate ($2\times 2\text{ cm}^2$) fixed on the linear motor was used as an indenter to compress the PVDF-based and PI-based devices. At the same time, the short-circuit current, open-circuit voltage and transferred charge were measured by an electrometer (6514, Keithley); the force data were obtained by using a force gauge (ZTS-DPU 100N, IMADA) and the sampling rate was 2000 data points per second. The piezoelectric coefficient d_{33} of the PVDF film was measured by a commercial instrument (ZMpiezo-P) provided by Foshan Zilm Technology Co. Ltd.

Declarations

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Author contributions

C. C. and C. Y. conceived the idea and designed the experiments. C. C. and S. Z. carried out the experiments and analyzed the results. C. C. wrote the paper. All authors reviewed, discussed the edited the manuscript. C. Y. and Z. W. supervised the project.

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Figures

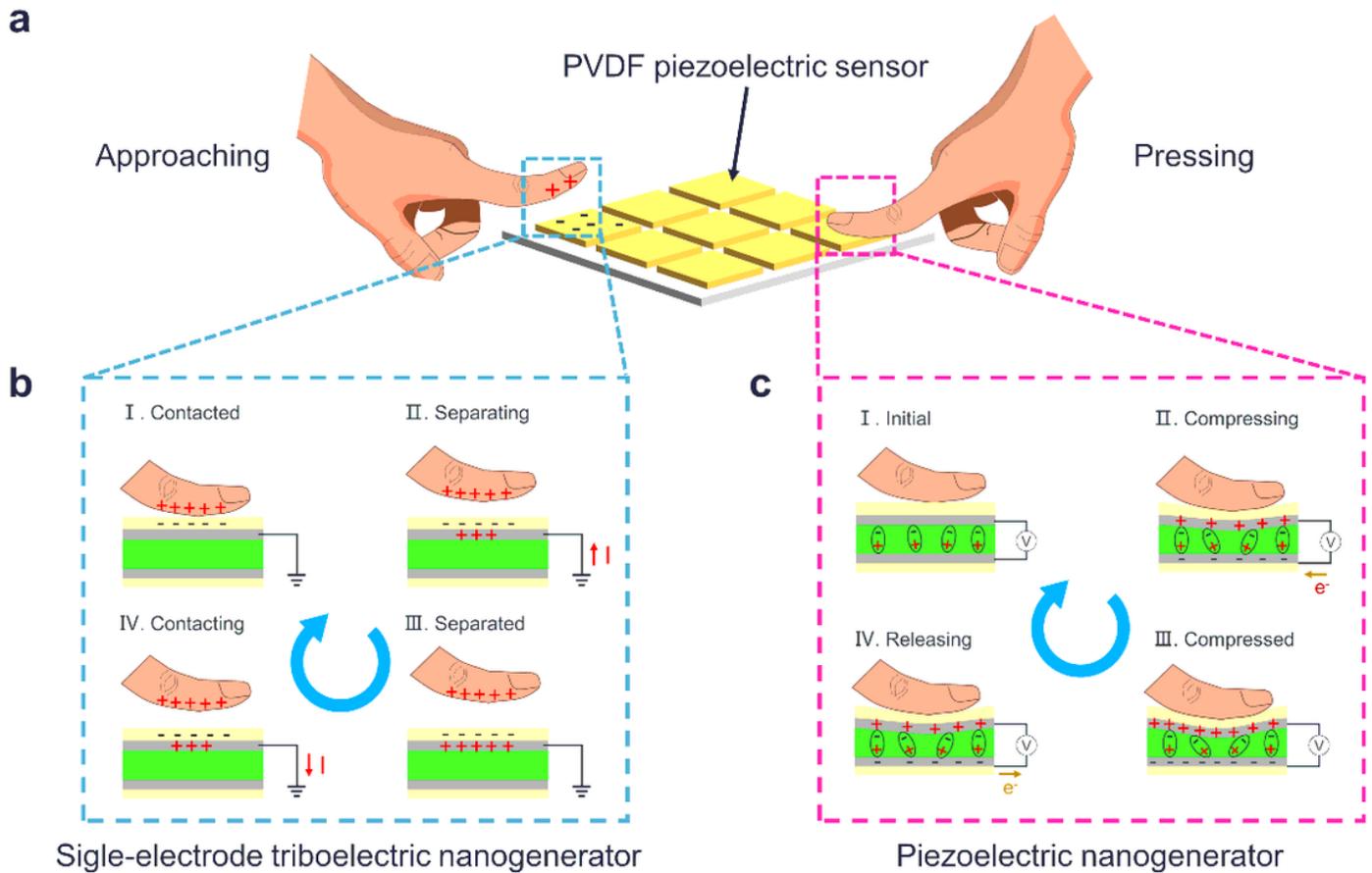


Figure 1

Schematic of the existence of both SE-TENG and PENG during practical application of piezoelectric devices, but the time at which the signal is generated has distinct differences. a The human finger has positive charges and the surface of a piezoelectric pixel has negative charges (left). The piezoelectric material would experience elastic deformation when human finger is pressing it and a piezoelectric effect would occur (right). b An SE-TENG is formed in the practical application of piezoelectric devices when using the positive finger touches the negative piezoelectric pixel again. The working mechanism of SE-TENG contains four stages including contacted, separating, separated, and contacting, the triboelectric signals will be generated in the stages of separating and contacting. c The working mechanism of PENG contains four stages including initial, compressing, compressed, and releasing. Piezoelectric signals will be generated in the stages of pressing and releasing.

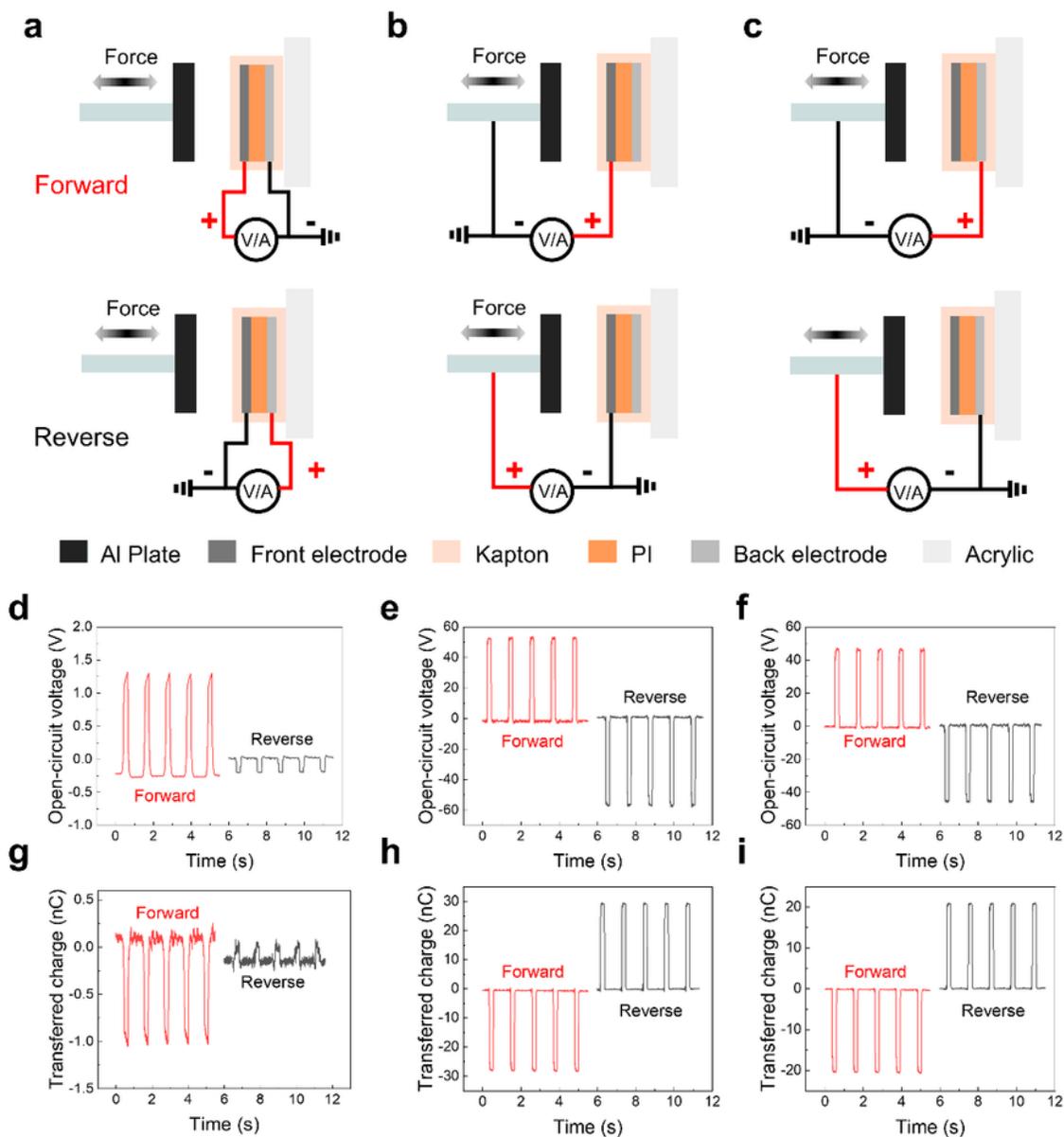


Figure 2

Verifying the existence of SE-TENG in a compression test. a Schematic diagrams of the forward connection and reverse connection when measuring the electric signals between the front electrode and the back electrode of the PI-based device. The corresponding open-circuit voltage and transferred charge results are shown in d and g, respectively. b Schematic diagrams of the forward connection and reverse connection when measuring the electric signals between the front electrode and the Al plate. The

corresponding open-circuit voltage and transferred charge results are shown in e and h, respectively. c Schematic diagrams of the forward connection and reverse connection when measuring the electric signals between the back electrode and the Al plate. The corresponding open-circuit voltage and transferred charge results are shown in f and i, respectively.

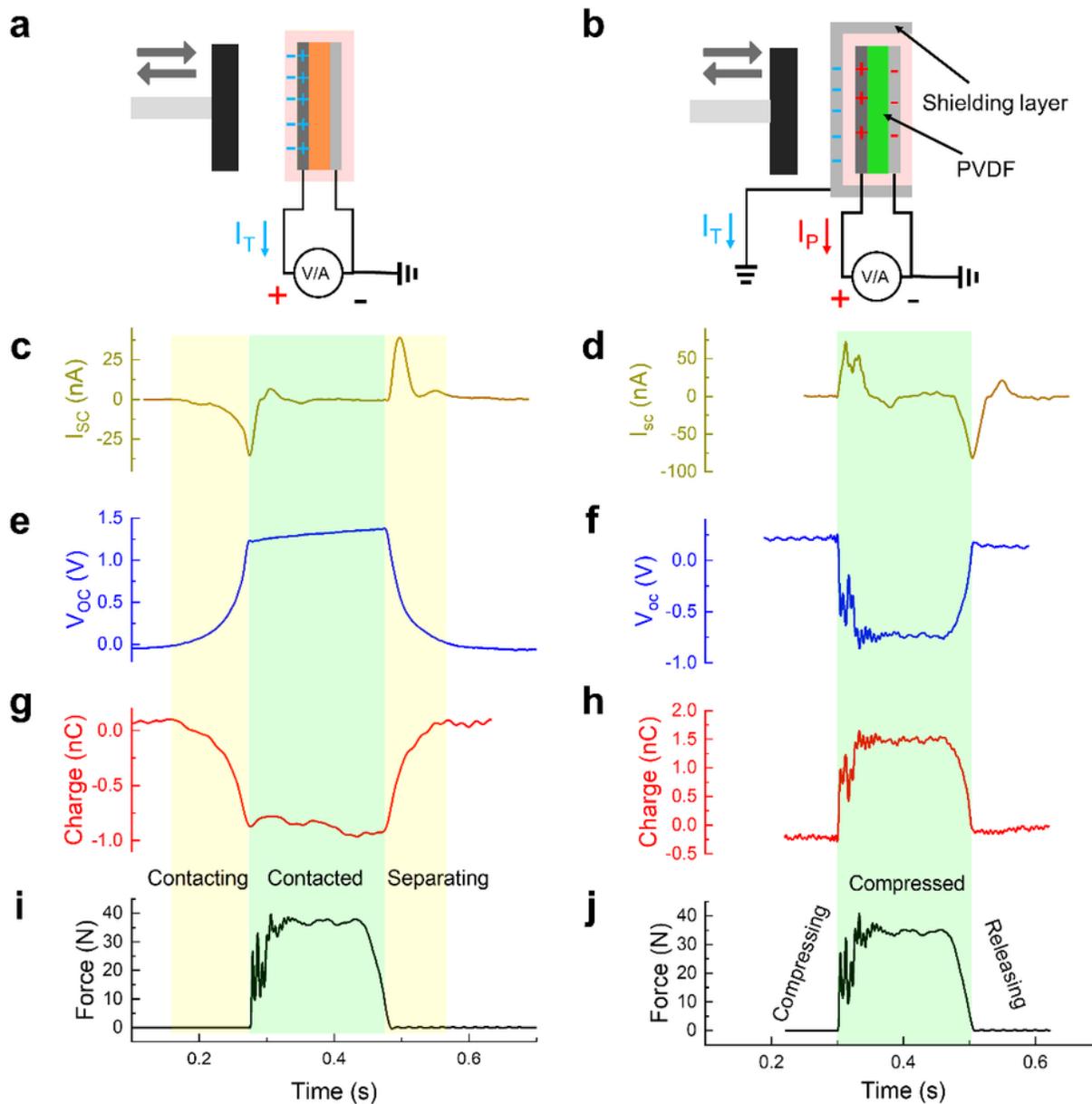


Figure 3

Using loading force signal to differentiate the sole triboelectric signals and the sole piezoelectric signals.

a The sole triboelectric signals (IT) are obtained in the front electrode of the PI-based device without a shielding layer. The corresponding short-circuit current c , open-circuit voltage e , and transferred charge g are generated in the stages of contacting and separating, according to the force signal i .

b The sole piezoelectric signals are obtained in the front electrode of PVDF-based device with a shielding layer. The conductive shielding layer is attached on the device's surface, which can guide the induced triboelectric charges (IT) flow to the ground and only piezoelectric signals (IP) can be obtained in the front electrode. The corresponding short-circuit current d , open-circuit voltage f , and transferred charge h are generated in the stages of compressing and releasing, according to the force signal j .

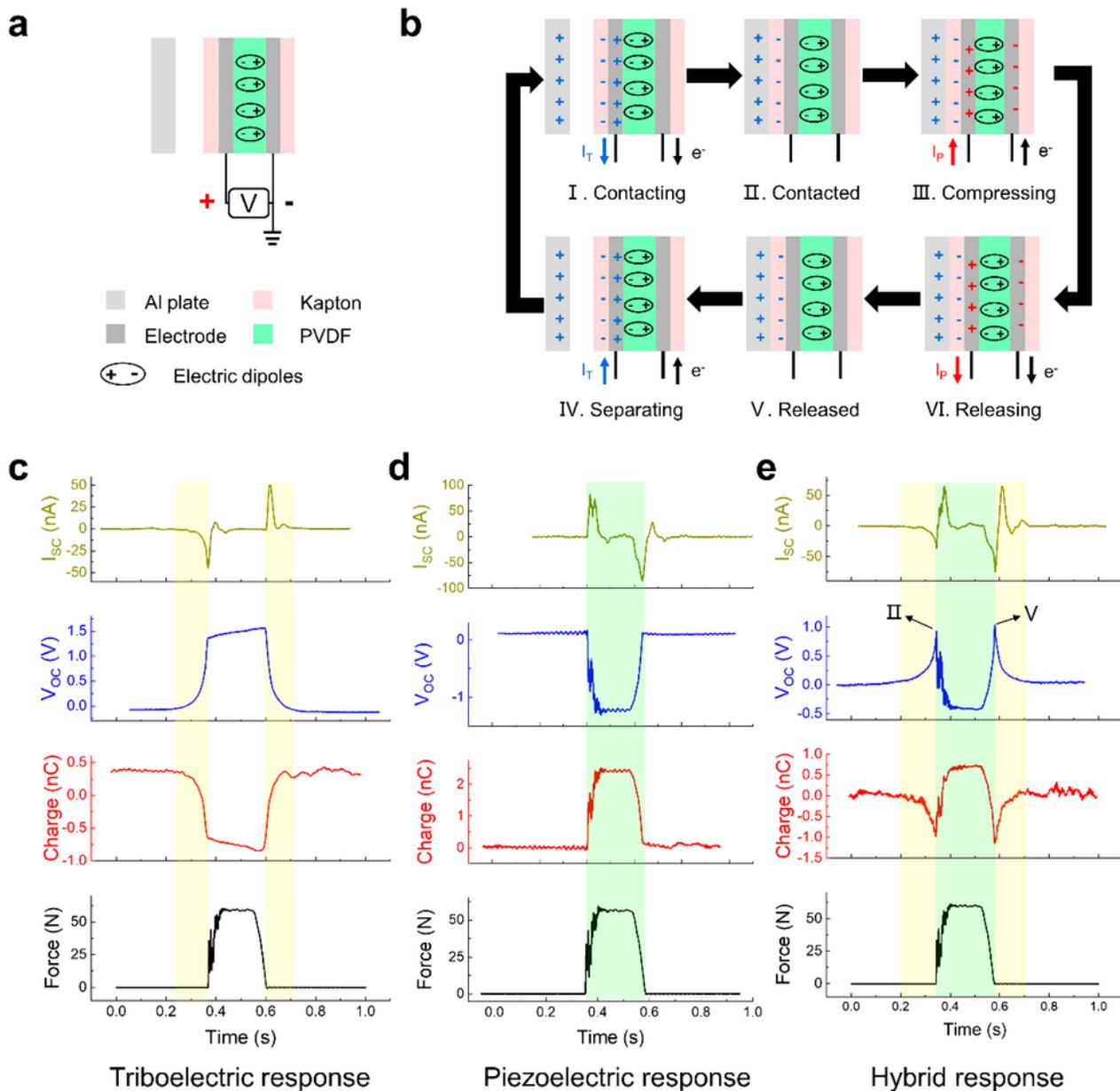


Figure 4

The triboelectric-piezoelectric hybrid output generated by the PVDF-based device with the negative polarization direction. a Schematic of measuring the triboelectric-piezoelectric hybrid output in the negative polarization direction of the PVDF film. b The signal generation process of hybrid output contains six stages including contacting, contacted, compressing, releasing, released, and separating stages. c The sole triboelectric signals (I_{sc} , V_{oc} , and charge) are generated by the PI-based device, and

their corresponding force loading signal. Sole triboelectric signals are generated in the stages of contacting and separating (light yellow area). d The sole piezoelectric signals are generated by the PVDF-based device with shielding layer, and their corresponding force signal. Sole piezoelectric signals are generated in the stages of compressing and releasing (light green area). e The triboelectric-piezoelectric hybrid signals (ISC, VOC, and charge) are generated by the PVDF-based device, and the corresponding force loading signal. The hybrid signals exist in the stages of contacting, compressing, releasing, and separating.

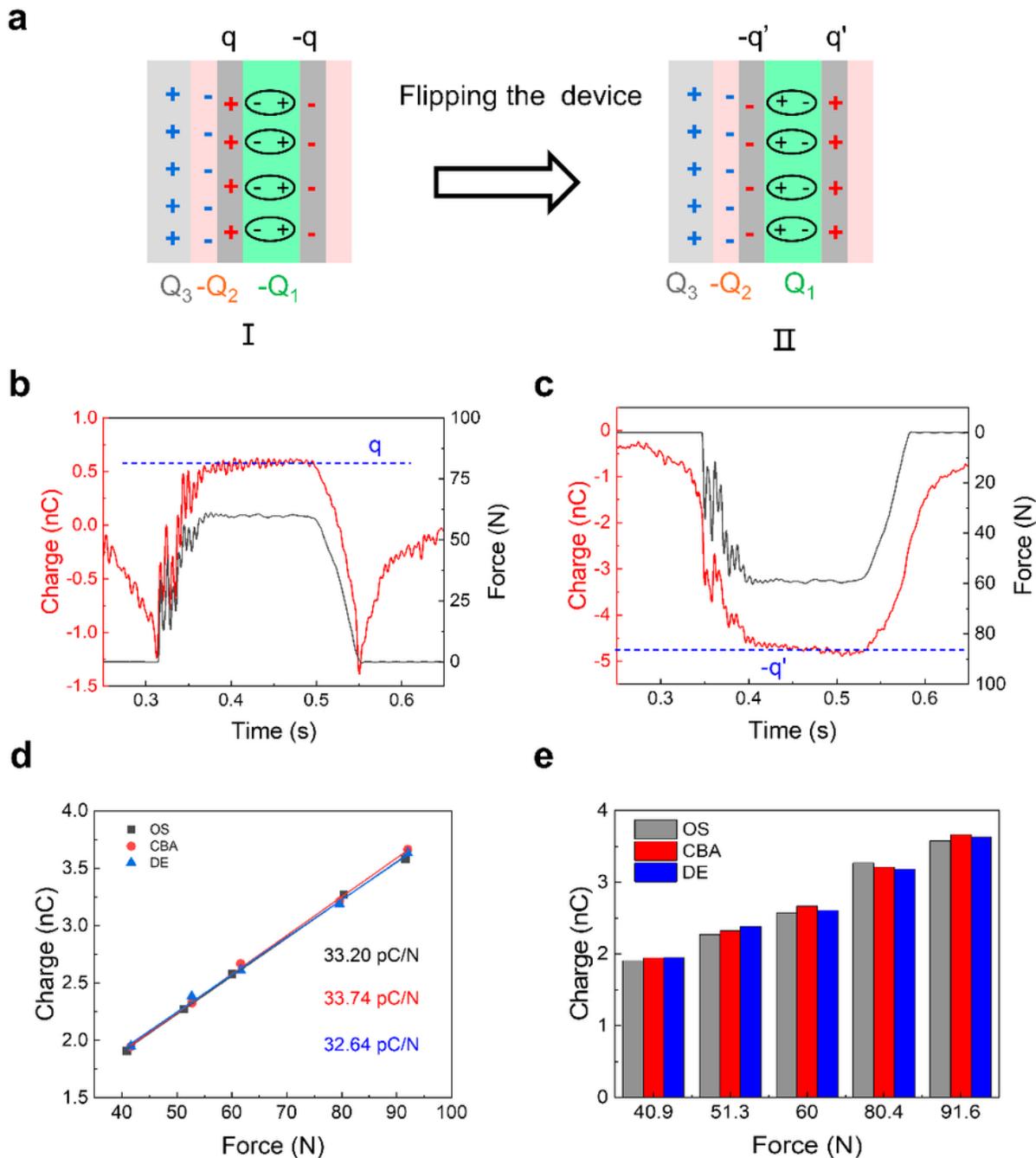


Figure 5

Evaluating the piezoelectric performance of PVDF film by extracting the piezoelectric charge transfer from the hybrid output. a The electrostatic balance is achieved when the Al plate and the PVDF-based device are contacted. Q_3 represents the positive polarization, $-Q_2$ represents the negative polarization, Q_3 is the total charge in the Al plate, $-Q_2$ is the total charge in the Kapton layer, q is the charge transfer between electrodes in the positive polarization, Q_1 is the induced piezoelectric charge, $-q'$ is the charge transfer between electrodes in the negative polarization. The transferred charges between electrodes are measured from the b positive and c negative polarization sides of PVDF-based device, and the applied force is 60 N. d The charge-force curves of PVDF-based device are obtained by three methods including outside shielding (OS), compressed balance analysis (CBA), and directly extracting (DE). The d_{33} of OS is 33.20 pC/N, that of CBA is 33.74 pC/N, and that of DE is 32.64 pC/N. e The piezoelectric charge transfer under different forces is obtained by three methods including OS, CBA, and DE. They are almost equal.

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