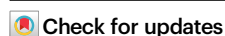


Direct current generation in triboelectric nanogenerators through ionic dynamics and electrode polarization effects

Received: 14 January 2025

Accepted: 17 September 2025

Published online: 29 October 2025



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Efficient conversion of mechanical energy into direct current remains a significant challenge for current energy-harvesting devices. In ionic tribomaterials, the displacement and polarization of mobile ions can dynamically control both the magnitude and direction of generated charges. This process shares similarities with semiconductor-based tribovoltaic systems but differs from conventional dielectric triboelectric devices, where electrical conductivity and charge retention are often in competition. While ionic tribomaterials are receiving increasing attention, their ability to generate direct current directly from mechanical motion has not been fully investigated. Here, we show that incorporating ionic components, such as plasticizers, into a common dielectric polymer (polyvinyl chloride) transforms the output from alternating to direct current. This process is driven by contact electrification combined with electrode polarization, enabling stable direct current generation across contact–separation, sliding, and rotary motions—modes that are typically difficult to unify in a single design. The resulting devices maintain stable output under extended operation and varying environmental conditions, demonstrating a robust and versatile route for mechanical-to-electrical energy conversion. This approach bridges the performance gap between polymer-based triboelectric devices and tribovoltaic systems, offering a broadly applicable strategy for sustainable energy harvesting technologies.

The emergence of internet of things (IoT) devices¹, sensors², and wearables³ has fuelled the demand for compact energy sources. In response, researchers have engaged the scope of small-scale energy harvesting, leading to the development of innovative technologies such as triboelectric nanogenerators (TENGs)⁴ and piezoelectric nanogenerators (PENGs)⁵ to meet this need. These devices leverage the

intrinsic properties of materials to capture energy from mechanical motion. TENGs exemplify this shift towards sustainable energy solutions⁶. Operating on the principle of contact electrification and electrostatic induction, TENGs utilize material pairs to facilitate the transfer of electrons, ions, and material⁷, to produce electrical energy. TENGs are promising for small-scale energy harvesting⁸, but their AC

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output requires rectifiers for DC conversion, which leads to complexity and energy losses, highlighting the need for more efficient DC generation methods.

Efforts to address this challenge have led to the exploration of strategic approaches such as electrostatic breakdown⁹, phase control^{10,11}, mechanical switching methods¹², which use dielectric materials, and the tribovoltaic effect, which uses semiconductor-based materials¹³ to design TENGs which can produce DC. However, TENGs utilizing dielectric materials often require complex structures and exhibit high impedance, resulting in energy losses⁹. Electrostatic breakdown necessitates high surface charge density and generates output only during unidirectional sliding^{9,14}, while phase control and mechanical switching methods introduce complexity that can lead to wear in components and still rely on rectifiers in some cases, contributing to further power losses^{13,15}. The tribovoltaic effect directly generates DC electricity using semiconductor materials by creating electron-hole pairs from mechanical sliding, resembling the photo-voltaic effect. However, understanding of this effect is limited and often relies on complex, costly materials^{16,17}. Despite the capability for high-current DC generation, the output suffers from low voltage, and increasing the area does not directly increase the power generated.

Recent studies have demonstrated DC generation in TENGs using conductive materials, though with notable limitations. Kunming Shi et al. introduced a DC-TENG in contact-separation mode with an adhesive-conductive interface, where mechanoion generation via covalent bond cleavage drove rectification-free DC output¹⁸. However, this design was limited to contact-separation mode, relying on engineered adhesive and conductive materials, and offered only a basic understanding of the DC generation mechanism, limiting its adaptability to other modes. Similarly, Xuemei Zhang et al. achieved high-voltage DC output through the quasi-tribovoltaic effect using conductive insulators and high-dielectric electrets¹⁹. However, the device's sliding-mode operation involved a complex electrode structure and required a higher load resistance (100 MΩ, compared to 20 MΩ in our design), which may limit its scalability and efficiency for high-power applications. All these methods have enabled the production of DC from TENGs without rectification or complex structures. Yet, hurdles remain, such as generating DC from multiple mechanical loading modes, using inexpensive materials, without complex structures or external rectifiers, while achieving high output, and extending the applicability of DC TENGs to other device structures for maximizing energy harvesting. Therefore, further studies into novel methods and deeper insights into materials are needed to address these challenges efficiently.

Here, we introduce an approach to DC generation through a simple ionic-based direct current TENG (iDC-TENG). By leveraging the properties of partially conducting high-dielectric polymers within a conductor-dielectric structure, we demonstrate DC power generation without the need for an intricate design structure. We empirically show how material modification and ionic material behaviour can tune the output form (AC/DC) and magnitude of TENGs using different polymers with ionic content. This approach merges the advantages of tribovoltaic nanogenerators (TVNGs) and dielectric TENGs by overcoming the traditional tradeoff between conductivity and triboelectric charge, allowing for dynamic modulation of charge magnitude and direction through the ionic effect. Poly (vinyl chloride) (PVC) is tuned by adjusting its electrical properties to satisfy the conditions needed to achieve stable DC output. Electrode polarization (EP) is identified as a key phenomenon for the occurrence of DC in this method, and its relation to DC generation in TENGs is studied. With an understanding of the basic principle behind the occurrence of DC using the contact-separation (CS) mode, we further show improvement strategies and explore other oscillation dependencies for stable DC output. Finally, we explore how this mechanism can be extended into sliding-based TENGs and even, rotation-based TENGs (RTENGs).

We achieved a uniform DC output of (92 V and 9.60 mA/m²) for CS mode, (50 V and 3.94 mA/m²) for sliding, and (200 V and 727 μA/m²) (crest factor = 1.02) for a miniature RTENG design. In all configurations, the iDC-TENGs demonstrated excellent long-term stability, with sustained or continuous DC output over tens to hundreds of thousands of cycles under mode-specific operating conditions. Our findings not only showcase the viability of DC energy generation but also highlight the importance of identifying key parameters and the driving force of the mechanism which creates a solid foundation for optimizing DC-TENG devices and paving the way for industrializing efficient energy harvesting systems.

Results and discussion

The structure and working mechanism of the iDC-TENG

Figure 1a presents the schematic of the iDC-TENG, highlighting its device structure, which employs a conventional conductor-dielectric configuration operable in multiple mechanical modes—sliding, contact-separation, and rotary. The device consists of substrates, two electrodes, and a polymer film. Notably, the top electrode (Au) functions as both a triboactive layer and an electrode. Details of the measurement setup, including the mechanical pusher and force sensor for the contact-separation mode, are depicted in the photograph and schematic of Supplementary Fig. 1.

Considering the critical interplay between triboelectric charge generation and electrical conductivity in tribolayer materials, enhancing conductivity can improve charge transport but often leads to charge leakage, reducing triboelectric efficiency. Polymers containing ionic species exhibit multiple polarization mechanisms, which dominate under specific mass percentages and frequencies. These include dielectric polarization, interfacial (electrode) polarization (EP), and others. EP is particularly significant as it increases triboelectric charge generation while constraining conductivity to mitigate charge leakage, as illustrated in Fig. 1b, c. In conventional dielectric films, higher conductivity typically results in significant charge leakage, limiting their triboelectric performance. In contrast, in polymers with ionic species, EP arises from the movement and distribution of ionic species within the film, creating an internal electric field across the material, when they are submerged between two metal electrodes with a voltage across the film. While this principle is well-known in ionic actuators, it is rarely considered in TENGs utilizing ionic polymers. Here, the internal electric field is shown to regulate triboelectric charge generation, direction, and magnitude, enabling dielectric materials with ionic species to achieve stable DC output without added complexity. Using this principle, the limitations of tribovoltaic nanogenerators (TVNGs)—such as the rigidity and cost of semiconductor materials—and those of dielectric TENGs, primarily their AC output, can be addressed. This creates a pathway for developing superior TENGs that combine the functional advantages of common dielectric materials with the simplicity and DC output characteristics of TVNGs. To control the properties of PVC, we blend PVC with a compatible plasticizer. Pristine PVC is a highly resistant dielectric material widely used in all modes of TENGs. Primarily, PVC is a hard, rigid thermoplastic polymer, and the plasticizer is added to increase its plasticity, making it bendable for various applications²⁰. Implementing PVC into the conductor-dielectric contact separation mode results in AC power generation through the creation of alternating current from opposite charges between materials with different electron affinities (e.g., PVC and Au) during contact and separation across a load (Supplementary Fig. 2). Contrastingly, as shown in Fig. 1d, when the plasticizer content in PVC is increased from a ratio of 1:0 (PVC: plasticizer) gradually to 1:1, there is a gradual transition of output form from AC to DC. The output transition is separated into two parts: The first part (1:0–1:0.4) shows decreasing overall output peaks of the AC behaviour as a result of charge loss by leakage path (increased percolation)²¹, and the second part (1:0.4–1:1) shows the complete increase in unidirectional peaks

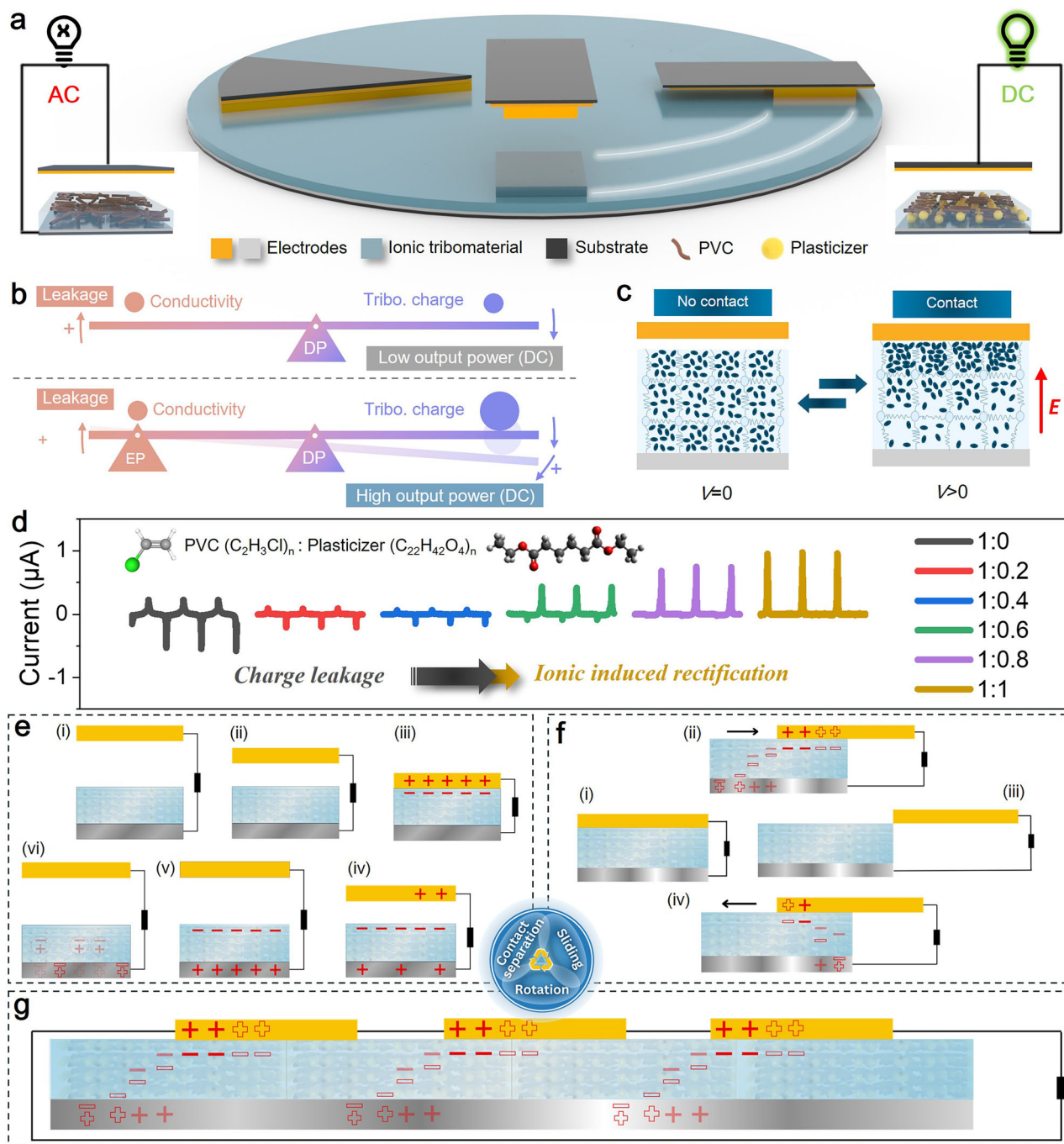


Fig. 1 | Schematic device structure, working mechanism, and output form of the iDC-TENG. a Schematic illustration of the iDC-TENG using a PVC (polyvinyl chloride) gel layer in rotary, contact-separation, and sliding modes with direct current output **(b)** Lever system showing relation of conductivity and power output in TENGs with ionic based TENG showing displacement of ions under an electric

field **(c)** Schematic of polarization of ionic species in the film under contact-induced electric field **(d)** Short-circuit current output of PVC (polyvinyl chloride) films containing different ratios of DOA (bis(2-ethylhexyl) adipate) plasticizer, showing the transition from AC to DC, Working mechanism of **e** Contact Separation iDC-TENG, **(f)** Sliding iDC-TENG, and **g** iDC-RTENG.

transforming into full DC output, which is primarily a result of interfacial polarization.

This behaviour is primarily realized because the intrinsic electrical properties of a PVC film can be widely changed from its original state of high resistance to partially conducting by the inclusion of a plasticizer. With an increase in plasticizer, a softer, slightly conductive polymer emerges due to the PVC-plasticizer interaction forming free volume by the effect of the plasticization²². The PVC also shows a higher tendency to gain electrons with the inclusion of the plasticizer²³. It is further identified that plasticized PVC having a conductivity above ~ 2 nS,

shows DC characteristics in output. The conductivity, dielectric loss, along with the voltage output and transferred charge are shown in Supplementary Fig. 3. To better visualize the output of the iDC-TENG based on the PVC gel we have shown the full cycle of the open-circuit voltage (V_{OC}), short-circuit current (I_{SC}) and transferred charge (Q) of an increased ratio (PVC: plasticizer, 1: 5) in Supplementary Fig. 4 and Supplementary Movie 1. The output clearly shows DC features, with a monotonic increase and unidirectional output. The V_{OC} , I_{SC} and Q could reach 92 V, 6 μA , and 50 nC respectively, where each spike of output is seen per cycle, at the separation stage of the operating

iDC-TENG. The detailed analysis of the output form upon contact shows that, the V_{OC} is maintained at -0 V , along with the I_{SC} , which is unlike the case for conventional TENG in the conductor-dielectric mode shown previously in Supplementary Fig. 2.

The proposed mechanism (Fig. 1e) of this iDC-TENG is described as follows for contact separation mode: (i) At the initial stage, the two electrodes are separated, and (ii) even in the process of contacting, charges are barely transferred because both contact surfaces exhibit conductive behaviour leading to almost complete depletion of static charges on their surfaces. (iii) Upon intimate contact, charges are exchanged between the Au and the plasticized PVC surfaces due to contact electrification by effective areal contact. The top electrode which is a metal has a more positive triboelectric tendency than the plasticized PVC film, therefore upon contact, the triboelectric effect would occur, and positive and negative charges are assumed to be generated on the top electrode surface and the PVC gel surface, respectively. Charges that would have remained on the dielectric material in normal cases (when the dielectric is not conducting) quickly dissipate upon separation (iv) due to the high mobility and ionic nature of plasticized PVC: the driving force for the neutralization of the charges are ionic charge mobility and a volumetric charge balance in the film. This effect is only possible because of the level of ionic conductivity of the film which enables an internal electric field to be established to create a barrier for charge rebound in one direction. After separation (v and vi), both surfaces are now depleted of charges and the cycle restarts. As can be inferred, when plasticized PVC is used in the same conductor-dielectric TENG structure as pristine PVC, it produces DC. In this configuration, the output is generated exclusively during the separation stage of the process, while the contact stage is suppressed. This suppression leads to a unidirectional output, meaning that the current flows consistently in one direction rather than alternating. This behaviour contrasts with the conventional mechanism where both contact and separation stages contribute to the output, leading to AC generation. A similar principle underpins the sliding mode (Fig. 1f), in which charges are generated at contact points and dissipate upon separation due to electrostatic polarization (EP), creating a current flow during sliding motion. Extending this concept, the iDC-RTENG integrates multiple sliding layers in series, generating charges in one direction facilitated by the shielding effect of EP (Fig. 1g), which will be further discussed in the paper.

We propose that the behaviour at the contact stage occurs due to the phenomenon called electrode polarization (EP). This is the accumulation of charge on the surface of a film at the film electrode junction which inherently renders the true contact electrification charges altered, EP usually occurs in dielectric ionic conducting films which are endowed with the capability to have other forms of polarization²⁴. Using films which are partially conductive can lead to the free ions in the films moving toward the electrodes under the influence of an electric field²⁵. In other fields of study, EP is a challenge for the measurement of electrical properties because of the false electrical properties exposed by the high dielectric loss especially in the low-frequency range (EP characteristics are discussed and shown in Supplementary Fig. 5 and Supplementary Note 1). The DC output in this iDC-TENG might have been mistakenly attributed to the tribovoltaic effect; however, plasticized PVC lacks typical semiconductor characteristics (Supplementary Fig. 6). The EP does not directly produce or influence the output but rather screens the peak at the contact stage, allowing triboelectric charging based on triboelectric tendencies. We therefore assessed the contribution of the plasticity of our materials, specifically the influence of adhesion, and its effect on the output. Plasticized PVC exhibits the phenomenon of bond scission, which drives material transfer, thereby increasing the magnitude of the DC output. This phenomenon has been widely studied in TENGs as a driving force to enhance output in both AC and DC TENGs^{26–28}. Supplementary Fig. 7 and Supplementary Note 2 provide detailed insights

into the roles of ionic conductivity and plastic nature, helping to clarify their contributions to the generation of DC output in our system. Upon separation, the output shows a sharp peak, indicating the rapid transfer of charges to a neutral state. The dielectric layer generates triboelectric charges, while EP modulates charge transfer through the electric field across the film during the contact stage.

To further explore this mechanism using other materials, we fabricated ionic liquid (IL) 1-butyl-3-methylimidazolium bis (trifluoromethyl sulfonyl) imide (BMIM-TFSI) based PVC and PDMS films. We showed the gradual transition of the TENG output of these films with increasing WT% of ionic liquid. As can be seen in Supplementary Fig. 8a and c, the output drops quite sharply as the conductivity (Supplementary Fig. 8b and d) of the film increases until a transition point and then increases in unidirectional output to DC. Although the overall output reduces with increasing IL content due to more pronounced leakage paths, it is still clear that within a given ionic content range, the characteristics of a given ionic polymer film can display DC characteristics in the conductor-dielectric contact mode of TENGs. Considering the output behaviour of these materials, the mechanism of the iDC-TENG is influenced by the EP effect, usually indicated by higher conductivity levels. However, common triboelectric materials may not show good output with increasing conductivity, as increased conductivity usually means more charge leakage paths, which limits charge generation. EP can be prevented when an insulative material is placed between the ionic film, blocking the electrical connection between the electrodes²⁹. In Supplementary Fig. 9, we blocked one end of an electrode with a PI film, and studied the effect of the output, and it showed AC characteristics. We also explored single electrode mode Supplementary Fig. 10, and it yielded AC due to EP effect being absent. Supplementary Note 1 discusses further the EP and its implications in these structural alterations.

Output performance of the iDC-TENG based on material properties and electrode configuration in contact separation mode

Plasticized PVC has simultaneous ionic and electret properties, which make it very viable for this TENG. These electrical properties can be tuned by plasticizer type and plasticizer content to optimize the output and behaviour of the iDC-TENG. We utilized adipate plasticizers due to their environmental friendliness compared to other plasticizers such as phthalates. The chemical structures and properties of PVC and the plasticizers DDA, DOA, and DBA are presented in Supplementary Fig. 11. Their molecular interactions within the polymer matrix are further confirmed by FTIR analysis, where characteristic $C=O$ stretching peaks ($1729\text{--}1736\text{ cm}^{-1}$) indicate the presence of plasticizers and their varying degrees of interaction with PVC^{30,31}. These differences, influenced by the alkyl chain lengths of the plasticizers, contribute to variations in the material's structural and electrical properties²³. PVC gels were prepared at PVC: plasticizer weight ratios of 1:1, 1:3, and 1:5 for each plasticizer (DDA, DOA, DBA); the full fabrication procedure and validation of the gel-like film properties are provided in Supplementary Fig. 12. Notably, all films show DC behaviour but vary in the magnitude of output (at separation) and leakage (at contact). As shown in (Fig. 2a–c), all films of DDA and DOA showed an increase with increasing plasticizer content, from 1:1 to 1:5, where DDA increased from 29 V to 43 V, and DOA increased from 39 V to 92 V while DBA decreased from 28 V to 18 V. The optimal film was realized at 1:5 ratio of DOA with a total transferred charge of 45 nC per cycle. With these plasticizers, a higher content (above a 1:5 ratio) yields a film with insufficient tensile strength and hardness for TENG applications, and thus was excluded from this study (see mechanical properties of films in Supplementary Fig. 13). KPFM analysis (Supplementary Fig. 14) confirmed that plasticizers increase surface potential negativity with content and chain structure, consistent with our previous finding that they render PVC more electron-accepting in the order of DDA, DOA, and DBA²³. This trend was further supported by contact angle

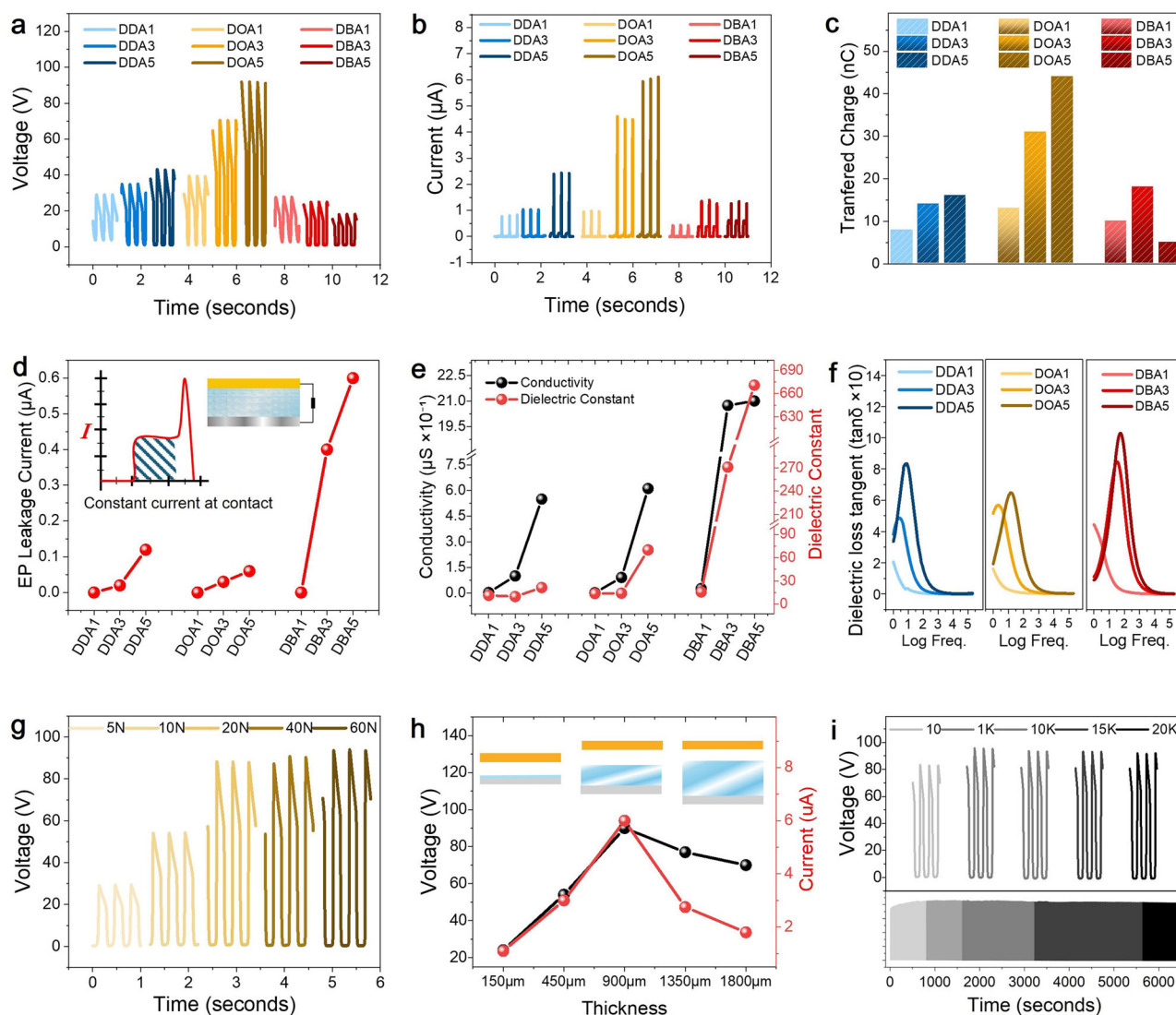


Fig. 2 | Optimizing DC triboelectric properties of the plasticized PVC.

a Open-circuit voltage (**b**) Short-circuit current and **c** Total transferred charge of iDC-TENGs using PVC films plasticized with DDA (diisodecyl adipate), DOA [bis(2-ethylhexyl) adipate], and DBA (dibutyl adipate) at ratios of 1:1, 1:3, and 1:5. **d** Electrode polarization leakage current values of the various films; inset: representative current–time profile highlighting the constant leakage current and

schematic of the electrode–film in contact (**e**) Conductivity and dielectric constant of various films at 5 Hz (**f**) Dielectric loss tangent of various films over frequency of 0 Hz–200,000 Hz (**g**) Force dependence of DOA5 films from 5 N–60 N (**h**) Thickness dependence of DOA5 film from 150 μm–1800 μm (**i**) Stability and durability test for over 20,000 cycles for contact separation DOA5 films.

measurements (Supplementary Fig. 15), which followed a similar pattern and are consistent with changes in surface dipole orientation³². Contrastingly, the observed reduction in output for DBA films can be attributed to their comparatively high conductivity potentially emanating from the high intrinsic ionic mobility. Despite their triboelectric tendency to generate higher charges, the smaller molecular size of DBA films results in increased EP leakage, which impedes the generation of effective output power. The EP leakage current, as perceived in the output, is shown in Fig. 2d; it is the static current measured at the contact stage of the contact–separate cycle of the TENG (Supplementary Fig. 16). Supplementary Fig. 17 further shows the transferred EP charge leakage details. When EP occurs there is a formation of a layer of charges akin to the formation of an electric double layer (EDL) which is seen as a static voltage and current in TENG output measurement (Supplementary Note 1 and Supplementary Fig. 5). This behaviour shows an intrinsic capacitance, or a built-in electric field formed at static contact. As can be seen, all films increase in leakage current with increasing plasticizer content. It should be clear that, in this iDC-TENG,

a substantial amount of this EP leakage is a condition for DC to occur; above or below a certain threshold would alter the output form. DOA films maintain comparatively low charge leakage values (Supplementary Fig. 18) which is sufficient to maintain DC and low enough to prevent charge loss by high EP leakage. Although DDA films have lower conductivities than DOA, there is a relatively large EP leakage observed in DDA5 as a result of molecular insolubility and plasticizer incompatibility with PVC due to DDA's high molecular weight³³. For DDA, we ascribe that the high leakage voltage values signify that leakage is not due to the mobility of blended plasticizer and PVC but rather the abundance of either PVC or plasticizer molecules showing insulative properties at the interface. The conductance and the dielectric constant (Fig. 2e) also increase in that same order and with an increasing ratio. As seen in the details of the resistance and Nyquist plots, and capacitance plots in Supplementary Fig. 19 and Supplementary Fig. 20 respectively, at the lower frequency, the plasticized PVC Films based on plasticizers with shorter molecular chains show increased capacitance and reduction in conductivity over two orders. This behaviour is

typical to dielectric films with the electrode polarization phenomenon and also indicates a transition of dielectric polarization from electronic polarization to ionic or interfacial polarization²⁴. The loss of the films was further evaluated by measuring the dielectric loss tangent ($\tan\delta$) in Fig. 2f. Comparing the highest ratios (1:5), DDA, DOA, and DBA have a $\tan\delta$ peak of 82, 79, and 103 respectively, along with increasing area and a right shifting of the loss peak to higher frequency. The $\tan\delta$ represents the losses in dielectric materials when subjected to an electric field. The balance of electrical and triboelectric properties of DOA, achieved uniform DC output can be attributed to low loss fuelled by the achievement of steric balance of PVC and DOA unlike in the other plasticizers.

For the optimized DOA5 films, device oscillation parameters were investigated to analyze the occurrence of DC. According to Fig. 2g, increasing the pushing force linearly increases the output voltage and current (Supplementary Fig. 21 and Movie 2) reaching a saturation point of about 100 V and 9 μ A at 60 N of pushing force corresponding to a pressure of 150 kPa. The softness of the plasticized PVC contributes to this nature, whereby there is an improvement of output based on enhanced compression and release at the contact surfaces. It is observed that as the thickness increases, the output of the iDC-TENG with DOA5 follows a rising trend, reaching its peak at 900 μ m. Above this, the dynamics of deformation³⁴, the charge depth³⁵, internal resistance³⁶ as well as capacitance can attenuate the output (Fig. 2h). At a greater thickness than 900 μ m, charges are still generated but the thickness hinders the easy flow through the film to enable generation of optimal output. Increasing the area of the friction layers also results in increased output and approaches a saturation point beyond 9 cm^2 (Supplementary Fig. 22). This limitation is prone to TENGs and tribovoltaic nanogenerators as well, and can be attributed to mechanical limitations and electrical losses³⁷. Remarkably, this iDC-TENG maintains 98 % of its initial output after saturation and remains DC after 20,000 cycles showing stability as a reliable DC TENG in contact separation mode without any charge loss, degradation, or attenuation (Fig. 2i). This shows the formidability of our approach to designing DC TENGs with simple structure and polymer materials. In the Supplementary data, we describe the impact of contact and EP on TENG behaviour, focusing on the transition between AC and direct current DC output by using a separation distance oscillation sweep (see Supplementary Fig. 23 and Supplementary Note 3).

For CS TENGs with two dielectrics, the work function (ϕ) of the electrodes and the dependence on the output are hardly considered or studied in many TENG studies. However, a study by Pace et al.³⁸ highlighted on the preferred orientation of the electrode where it is shown that the more tribo-negative material should have the lower work function electrode to result in higher output even with the same friction layers. They suggest that there is a built-in potential created which boosts the total triboelectric potential. This proved that the electrode work function and their orientation should be taken into consideration for TENGs. In our study, like in the conductor-dielectric structure, the work function is not only important because of this built-in electric field but also, the tendency of electron transfer between the metal and the plasticized PVC, because the top electrode functions as both the electrode and tribo-pair. As described previously, EP is influenced by the applied voltage over the film. Therefore, if the two opposite electrodes have varying work functions, and are placed across an ionic conducting film, a defined electric field orientation can be expected across the film. We selected conventional electrode materials with known work functions Au (5.5 eV), Cu (4.8 eV), and Al (4.0 eV) shown in Fig. 3a to analyze this behaviour in the iDC-TENG. We evaluated the output using the same dielectric layer (DOA5) for various matched electrode pairs. Figure 3b shows that the iDC-TENG output increases with the top electrode having a higher work function and the bottom electrode having a lower work function. The I_{SC} follows suit and are

shown in Supplementary Fig. 24. The highest output was therefore recorded for Au as top electrode and Al as bottom electrode showing a threefold increase compared the opposite orientation.

We propose that the work function of electrodes influences the output of the TENG by affecting the built-in electric field created upon contact. This electric field is modified by the EP effect, particularly in conductive dielectric materials. According to Poisson's equation, the electric field $E(x)$ within the dielectric is impacted by ion concentration $\rho(x)$ and permittivity ϵ . The built-in potential V_{bi} , proportional to the work function difference ($\Delta\phi$), modifies the charge density σ . Thus, the effective charge density is adjusted to:

$$\sigma' = \sigma + \beta \Delta\phi \quad (1)$$

This adjustment leads to a modified V-Q-x relationship for the iDC-TENG, accounting for electric field leakage:

$$V = \frac{-\sigma'd}{\epsilon_0\epsilon_r} + \frac{Q}{S\epsilon_0} \left(\frac{d}{\epsilon_r} - x \right) + \frac{\sigma'x(t)}{\epsilon_0} \quad (2)$$

At short-circuit conditions, the transferred charge Q_{SC} is given by:

$$Q_{\text{SC}} = \frac{S(\sigma + \beta \Delta\phi)x(t)}{d_0 + x(t)} \quad (3)$$

This modified equation reveals that the built-in electric field due to work function differences affects charge distribution and leakage, highlighting its significant role in TENG performance. This contribution, not captured in simpler models, underscores the impact of the built-in electric field on both initial charge transfer and subsequent separation. A detailed discussion of these findings is provided in Supplementary Note 4. Consistent with the previous explanation of EP, we studied the output signals for the various electrode orientations. In Fig. 3c, it is shown that the electrode orientation influences the polarity of the output signal upon contact. This first establishes that the signal at contact is solely not an effect of contact electrification or electrostatic induction, but it is an effect of the EP caused by interfacial contact and ion displacement. As seen, in the current output, when the top electrode $\phi_{\text{Elec. 1}}$ is greater than the bottom electrode $\phi_{\text{Elec. 2}}$, the contact peak has an EP bias in the same direction as the output, and when the bottom electrode has a $\phi_{\text{Elec. 2}}$ greater than the top $\phi_{\text{Elec. 1}}$, the EP bias is in the opposite direction. The output current peak remains in the same direction for the separation stage (output) in both cases, but changes at the contact stage depending on the EP bias. The details of the three selected pairs are shown as follows: high work function-low work function (Au-Al), mid work function-mid work function (Cu-Cu), and low work function-high work function (Al-Au) (Fig. 3d). The optimal orientation, as previously discussed has a uniform DC form and the peak details are also evident in the current and charge plots (Fig. 3e-g), while the contact peaks of the unpreferred orientation plots appear to produce an AC output. The contact peak of DOA5 is small and almost negligible because it has an optimized level of EP, with a good charge generation ability. These could be the primary reasons why DC-TENGs have not been identified in many materials; the electrode orientation has not been considered. Not only does this orientation influence output, but it can also determine whether the behaviour is DC or AC.

Device operation parameters and its influence on behaviour of iDC-TENG

Essential parameters for output generation in TENGs can reveal more about charge generation and optimization for improvement. We evaluated the output of the iDC-TENG using frequencies from 1 Hz –15 Hz. The EP value is influenced by time because the movement of ions in response to the built-in electric field requires time to occur.

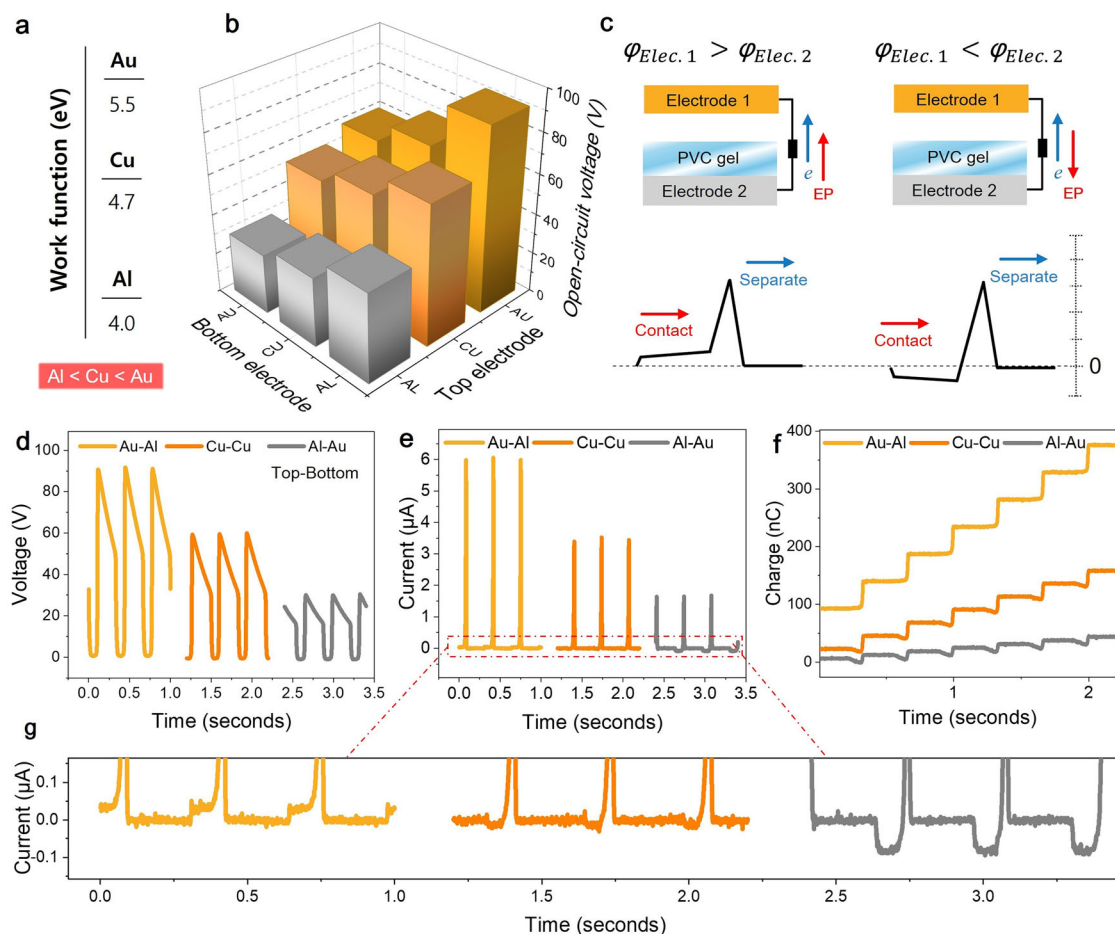


Fig. 3 | Electrode work function influence on the iDC-TENG. **a** Work function of selected electrode materials: Au (gold), Cu (copper), and Al (aluminum) **(b)** Open-circuit voltage of selected electrodes as top friction electrode and bottom electrode configuration **(c)** Schematic on short-circuit current waveform during contact and separate stages of one cycle depending on work function orientation **(d)**

Open-circuit voltage **(e)** short-circuit current and **f** total transferred charge of iDC-TENG with different orientations of electrodes **(g)** Magnification of short-circuit current waveform showing the transition from DC to AC Characteristics by work function orientation.

Therefore, the rapidness of contact and rapidness of separation influence the EP and output, respectively. The output increases at higher frequencies due to more CS cycles between the friction layers per unit of time until it reaches an inflection point, above 5 Hz. This behaviour can be attributed to changes that are more visible in the current output Fig. 4a. Herein, we describe the output dependence on frequency considering the I_{SC} . As seen in the inset of Fig. 4a the EP peaks for frequency are well formed at lower frequency values. This is because sufficient time was allowed for ion displacement. On the other hand, as the speed of the oscillation increases, the time for the formation of EP is reduced and as the two surfaces have less time to establish good contact and ions do not move as fast to form the EP, hence the EP reduces. At very high frequencies, AC behaviour is more prominent which buttresses the point that EP is influenced by time contact and that establishment of EP is essential for DC generation. Details on the charge output can be seen in Supplementary Fig. 25, where the transferred charge, leakage charge, and AC rebound charges are visualized. It can be observed that the AC rebound charge increased at the extremes of the highest frequency. More importantly, the output is influenced by the speed of charge transferred per second, therefore at optimal values at about 3 Hz – 5 Hz, the output attains a peak and then decreases as the frequency increases. Figure 4b expresses the contact/separation ratio as a percentage, where the transition from DC to AC is seen as the frequency gradually increases. A lower absolute value percentage signifies a better DC output. In Fig. 4c

the actual output power is shown and evaluated for different load resistance, with a constant frequency of 3 Hz. First, the current output was evaluated over various load resistances, from short-circuit ($0\ \Omega$ to $3\ \text{G}\Omega$). The gradual decrease in output was observed with an accompanying reduction in the EP leakage into AC at load resistance above $500\ \text{M}\Omega$ (Fig. 4c). The circuit setup for the measurement of the current at various loads is based on the conventional circuit setup for TENG, and the optimal load resistance is determined to be $20\ \text{M}\Omega$. The output of this DC TENG has two distinct regimes based on load resistance. At lower resistances up to about $500\ \text{M}\Omega$, the current is DC. However, above $500\ \text{M}\Omega$, the current becomes an AC waveform. This occurs because the high load resistance causes parasitic leakage and charge rebound, resulting in an AC output (Fig. 4d). Notably, this transition can be almost matched with the transition of AC to DC of the PVC film, as we increase the plasticizer; above $1.33\ \text{nS}$ or below the equivalent, $0.75\ \text{G}\Omega$, we attain full DC features (Supplementary Fig. 3a). In the AC regime of the load resistance dependence, high load resistance prevents electrons from flowing through the load as expected. This is like plasticized PVC used in a TENG with a blocking layer, indicating that output generation remains being related to the internal resistance of the film. The power density is calculated to be $260.2\ \text{mW}/\text{m}^2$ of DC output. To demonstrate the power usage in direct iDC-TENG, we compared capacitor charging and current waveforms from intrinsic DC-TENGs and rectified AC-TENGs under identical conditions (Supplementary Fig. S26).

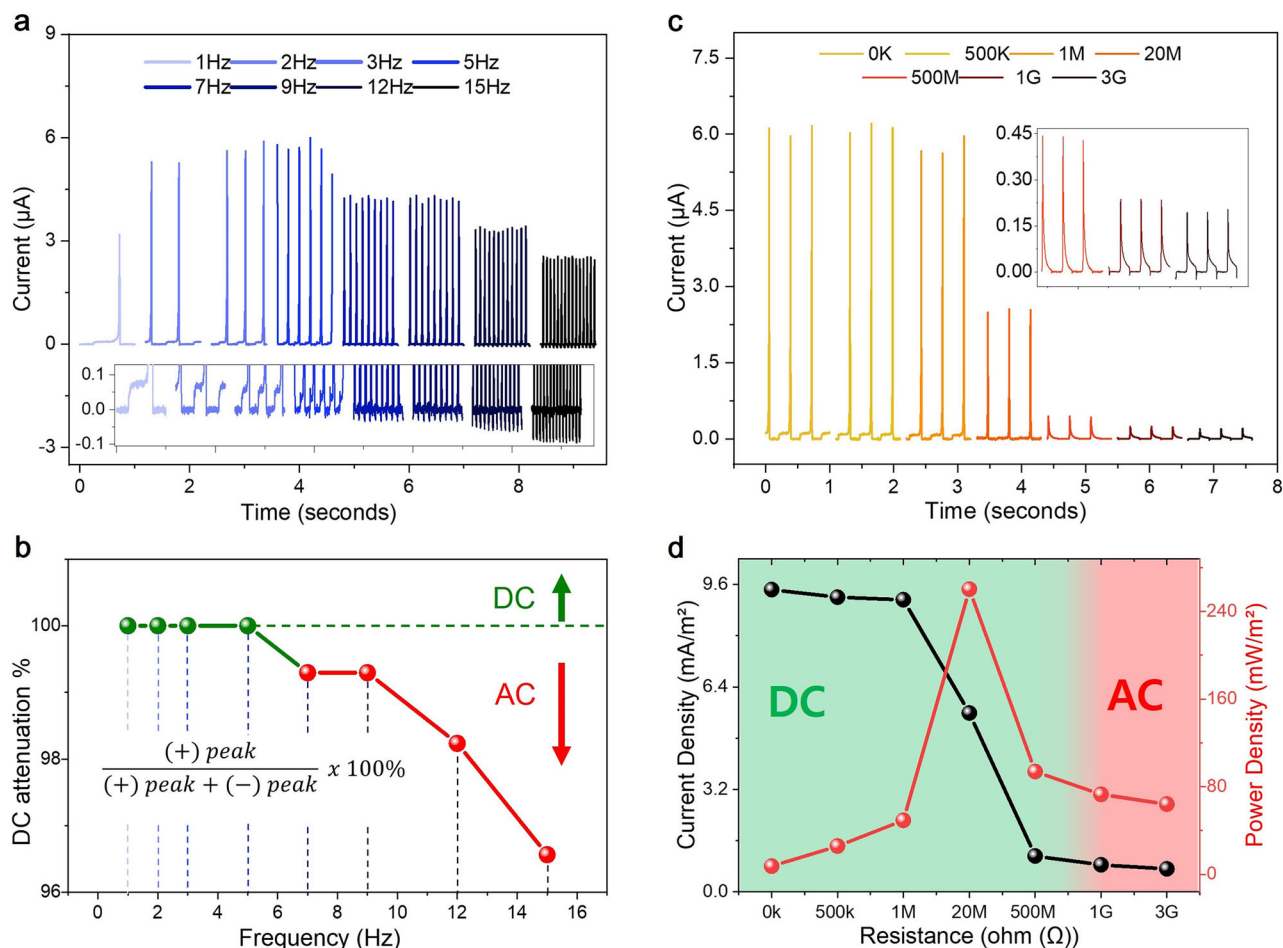


Fig. 4 | The behaviour of iDC-TENG depending on frequency and load resistance. **a** Short-circuit current of frequency dependence of iDC-TENG with magnification from 1 Hz–15 Hz; inset: Magnified view of the negative rebound peaks in the current signal at low frequencies **(b)** Contact/separation peak ratio percentage for frequencies from 1 Hz to 15 Hz showing the transition of DC to AC behaviour by

change in frequency in iDC-TENG; inset: Equation for calculating DC attenuation using (+) and (−) peak values **(c)** Short-circuit current dependence on load resistance; inset: Enlarged view of the high-resistance region ($\geq 1\text{ M}\Omega$) in the load resistance dependence plot **(d)** Output power density and current density dependence on load resistance transitioning from DC to AC at higher load resistance.

Sliding and rotary mode iDC-TENG

Most direct current (DC) triboelectric nanogenerators (TENGs) utilize the lateral sliding mode of operation³⁹. In this study, we investigated the behaviour of iDC-TENG in sliding mode to enhance our understanding and explore alternative mechanical harvesting techniques. For the sliding mode experiments, we used DOA1 film due to its favourable properties: it is neither jelly-like nor sticky, so its surface is ideal for sliding. As previously demonstrated in Fig. 2b, DOA1 film maintains stable DC output even with a low amount of plasticizer. We also employed the electrode orientation favourable for the contact-separation mode, positioning the higher work function metal as the top slider. Figure 5a presents a schematic of the slider, which is structured as a curved-deformable slider using PET-supported copper tape and secured by a rigid glass plate (Fig. 5b). We used foam within the slider so that the deformation can be well absorbed to reduce mechanical energy loss and wear during sliding. The bottom of the sliding iDC-TENG shares the same configuration as in the contact-separation (CS) mode, utilizing DOA1 film.

We propose the following sliding mechanism for the iDC-TENG (Fig. 1f and Fig. 5c): At the contact point (i), EP occurs and is maintained, ensuring that the charges transferred by contact electrification are overshadowed by the induced electric field. This polarization effect creates an internal electric field that significantly influences how

charges are generated and transferred. During movement (ii)—whether the sliding electrode remains in contact with another part of the surface or moves out of contact—the hindered charges are generated at points of contact and dissipate where the slider is separated. This perturbation in charge distribution generates a flow of current during the sliding motion. This mechanism is similar to the tribovoltaic effect, where the built-in electric field can orient the output in one direction, resulting in direct current (DC) generation⁴⁰. When there is complete separation and no contact (iii), the output remains null because the charges have been dissipated and are no longer present within the materials. As the sliding motion continues in the opposite direction (iv), the cycle restarts, producing output in the same direction as the forward motion. The output is generated instantaneously until the system returns to the initial state (i) at which point the cycle begins anew. If the dielectric could retain the charges instead of dissipating them, we would observe alternating current (AC) behaviour. The sliding mechanism involves simultaneous micro-contact and separation at the friction surfaces as the electrode moves over the PVC gel, leading to continuous generation of output. In the sliding mode, EP, contact electrification, and electrostatic induction occur simultaneously. The output exhibits clear DC characteristics in voltage, current, and transferred charge in both forward and backward directions, as shown in Fig. 5d–f, and Supplementary Movie 3. We further investigated the output at various sliding speeds, ranging from 20 mm/s to

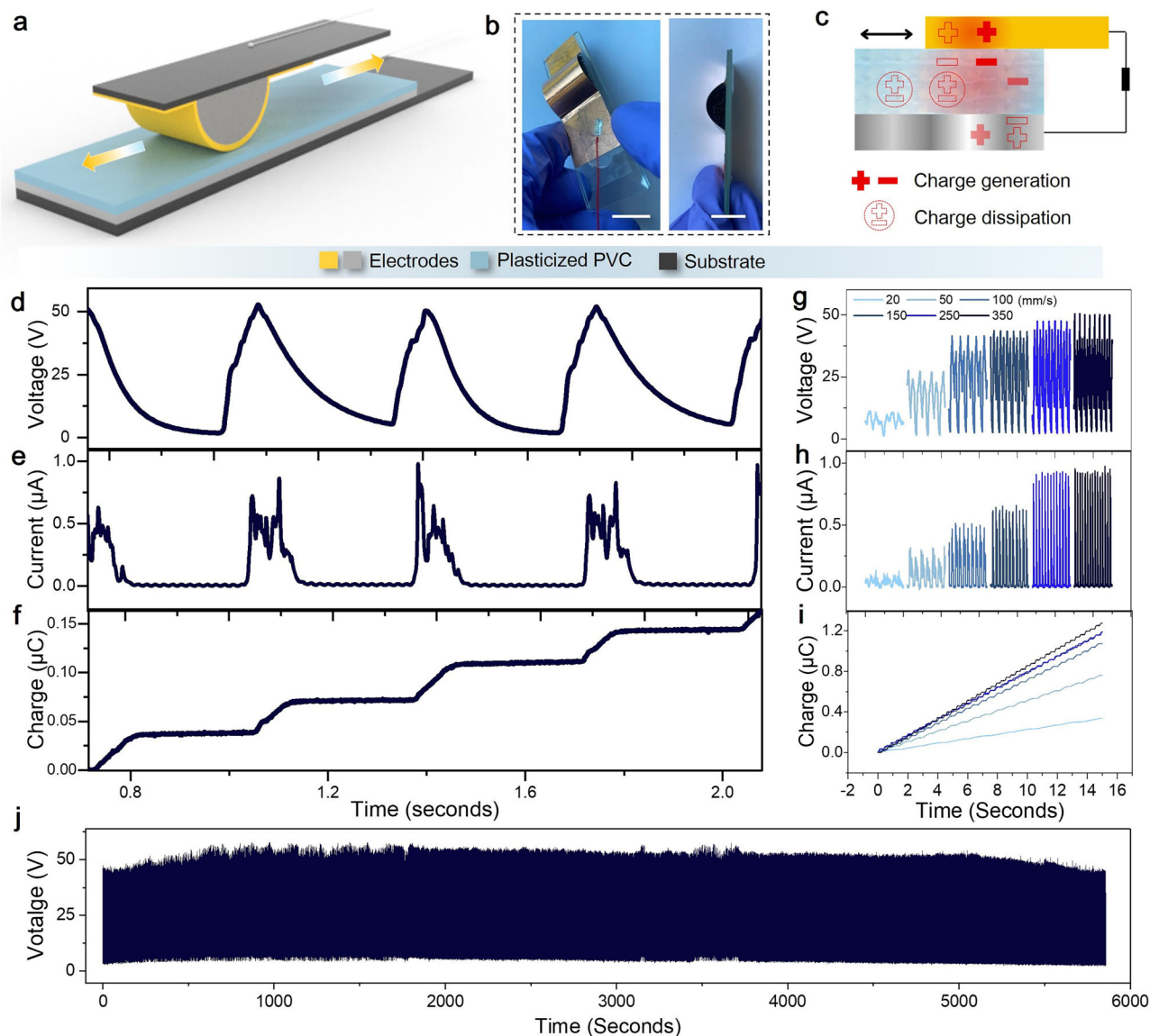


Fig. 5 | Output performance evaluation of iDC-TENG in sliding mode.

a Schematic of the structural design of the sliding-mode iDC-TENG incorporating a PVC (polyvinyl chloride) gel film **(b)** Photograph of the slider with foam-based copper film (Scale bar: 2 cm) **(c)** Mechanism for DC sliding mode operation **(d)** Open-circuit voltage **(e)** Short-circuit current, and **f** Transferred charge for sliding

mode iDC-TENG of the sliding mode **(g)** Open-circuit voltage and **h** Short-circuit current and **i** Transferred charge for sliding mode with different speeds from 20 mm/s to 350 mm/s **(j)** Voltage output of long-term output stability of the sliding-mode iDC-TENG over 6000 s.

180 mm/s, observing higher output as the speed increases. We suggest that higher speeds increase the frequency of micro contact-separation as the sliding electrode makes and breaks contact with the PVC gel. The output increased from 0.1 μA , 10 V at 20 mm/s to 0.9 μA , 50 V at 350 mm/s (Fig. 5g–i). Under varying temperature and humidity, the device retained over 50% of its output at 95 °C and remained stable below 60% RH, with full recovery after reconditioning (Supplementary Fig. 27). Environmental stability results for the contact-separation mode (DOA5) are also presented in Supplementary Fig. 27. The sliding-mode iDC-TENG (DOA1) maintained stable output over 6000 s of continuous operation with minimal degradation (Fig. 5j), demonstrating durability and consistent performance during prolonged operation.

To extend the functionality of sliding iDC-TENGs, we developed a palm-sized, ionic-based DC rotary TENG (iDC-RTENG) for DC energy harvesting from rotational motion. The rotational triboelectric structured devices leverage rotational energy to achieve high

power output without relying on complex structures, both to produce AC and DC output^{12,41,42}. This flap-based iDC-RTENG, based on the sliding mode, is specifically designed to increase the rate of energy harvested while maintaining good DC output. The structure, depicted in Fig. 6a, comprises three main components: the stator (plasticized PVC), the rotor (with flaps for the copper electrode supported by foam beneath), and a slip ring. A slip ring is an electromechanical device that facilitates the transfer of electric current from a rotating body to a stationary one (or vice versa). In iDC-TENG and iDC-RTENG configurations, the double electrode structure must always be grounded, to ensure EP. In our design, the slip ring enables this grounding without added complexity. The flap structure was meticulously chosen to ensure a mechanical favourable structure for smooth DC production. Figure 6b (i, ii, iii and iv) shows photographs of the rotor, stator, PVC gel film and the cross-sectional SEM image of the large area plasticized PVC film showing thickness for the iDC-RTENG. The mechanical properties of the PVC gel film were tested,

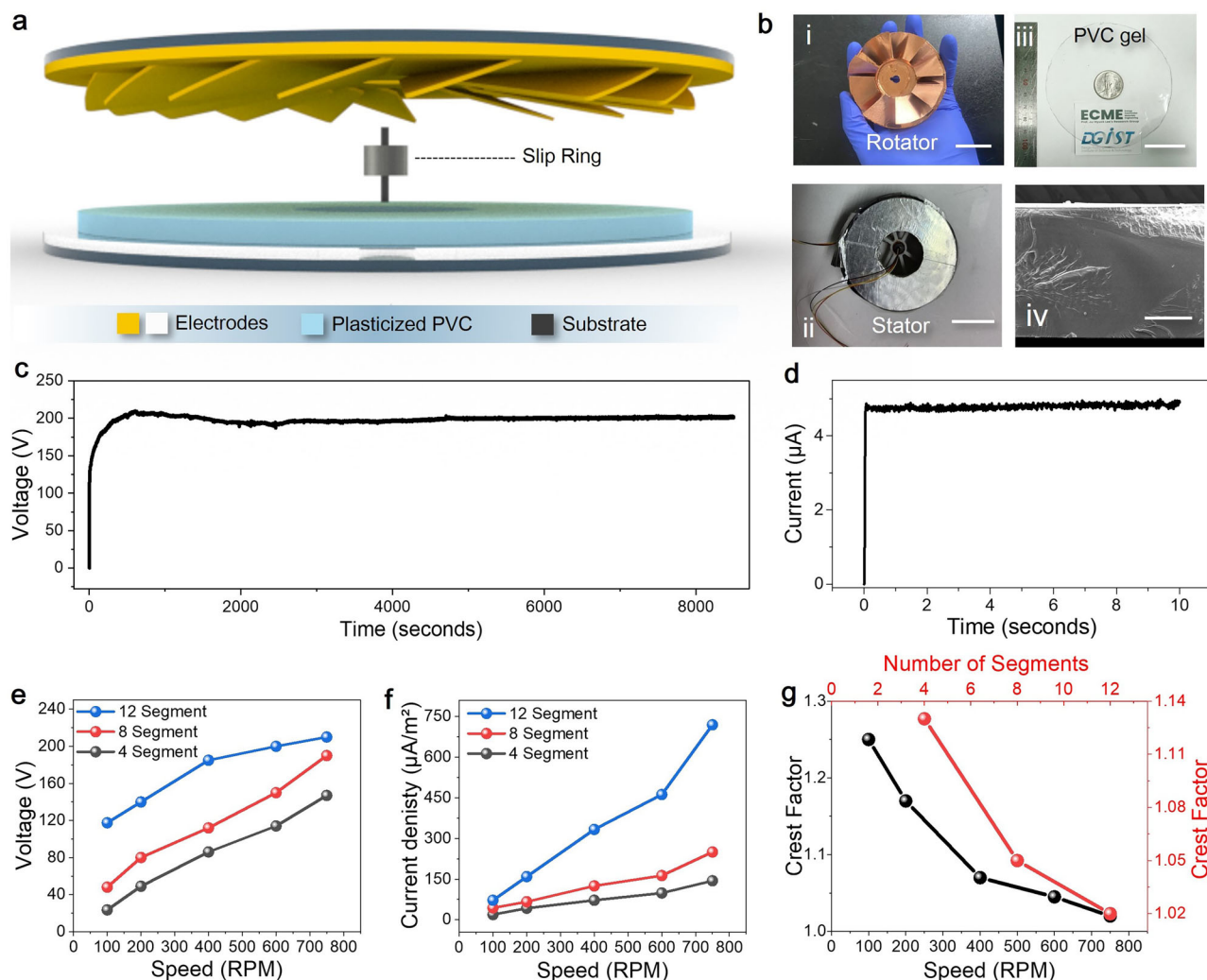


Fig. 6 | Output performance evaluation of iDC-RTENG. a Schematic of the structural design of the flap-based rotary-mode iDC-TENG incorporating a PVC (polyvinyl chloride) gel film. **b** Photograph of (i) rotor (scale bar: 3 cm) (ii) stator (scale bar: 3 cm) and, (iii) photograph (scale bar: 3 cm) and (iv) cross-section of large area PVC gel film (Scale bar: 100 μm) (c) Open-circuit voltage of the iDC-

RTENG with 12 segments at 750 rpm and its long-term stability over 8000 s (100,000 cycles) (d) Short-circuit current, and (e) Open-circuit voltage and (f) short-circuit current with different rpm's from 100 rpm up to 750 rpm (g) Crest factor of iDC-RTENG with different number of segments and various speeds of rotation.

revealing an ultimate tensile strength of 20.73 MPa and a remarkable stretching capability of up to four times its initial length (Supplementary Fig. 13). The flaps gently glide over the gel surface, facilitating intermittent micro-contact and separation, like the sliding mode. The iDC-RTENG conceptually functions as a series summation of multiple sliding layers, generating charges in one direction due to the shielding effect of EP (Fig. 2g). At equilibrium, the output is null; rotation induced by a motor initiates output generation. At 750 rpm, the V_{OC} and I_{SC} reach 200 V and 4.8 μA , respectively, demonstrating stable, continuous DC output (over 100k cycles) (Fig. 6c, d). DC operation is shown in Supplementary Movie 4, with output increasing instantaneously as the speed is increased. Supplementary Fig. 28 details the V_{OC} and I_{SC} of the iDC-RTENG with 12 segments at various speeds from 100 rpm up to 750 rpm. As rpm increases, sliding friction rises, enhancing charge transfer rates.

To further explore these mechanisms, we varied the number of segments in the flap-based TENG: 4 segments (90°), 8 segments (45°), and 12 segments (30°) Supplementary Fig. 29, all maintaining the same total area. The segmentation significantly influences the performance of rotary TENGs⁴². With the stator unchanged, the 12-segment TENG exhibits higher performance compared to fewer segments. This

increased output can be attributed to the better distribution of charges on the smaller area of the flaps⁴². However, this does not mean that output would always increase with increasing segmentation because, beyond a certain limit, the edge effect and other mechanical limitations become more prominent⁴³. As illustrated in Fig. 6e, f and Supplementary Fig. 30, at all speeds V_{OC} and I_{SC} the output increases for increasing segment number, demonstrating that segmentation is a critical factor. The crest factor indicates how uniform the DC output is. In our study, the continuous rotation of the iDC-RTENG results in a steady DC output, but the speed and crest factor influence the uniformity of the output waves. An increase in speed leads to higher output generation per unit time, resulting in a higher and steadier output. Additionally, segmentation affects the crest factor because EP contributes to the output form. It is well known that increasing the electrode area decreases the effect of EP^{29,44,45}. Therefore, with smaller segmented areas, the EP effect is more pronounced, leading to a more optimized and uniform DC output with a higher crest factor. Figure 6g shows the crest factor decreasing with increasing speed and the crest factor reduction with fewer segments, which corroborates the working mechanism of the DC TENG. Find further discussion on the crest factor equation and calculation in Supplementary Note 5. Therefore, for a

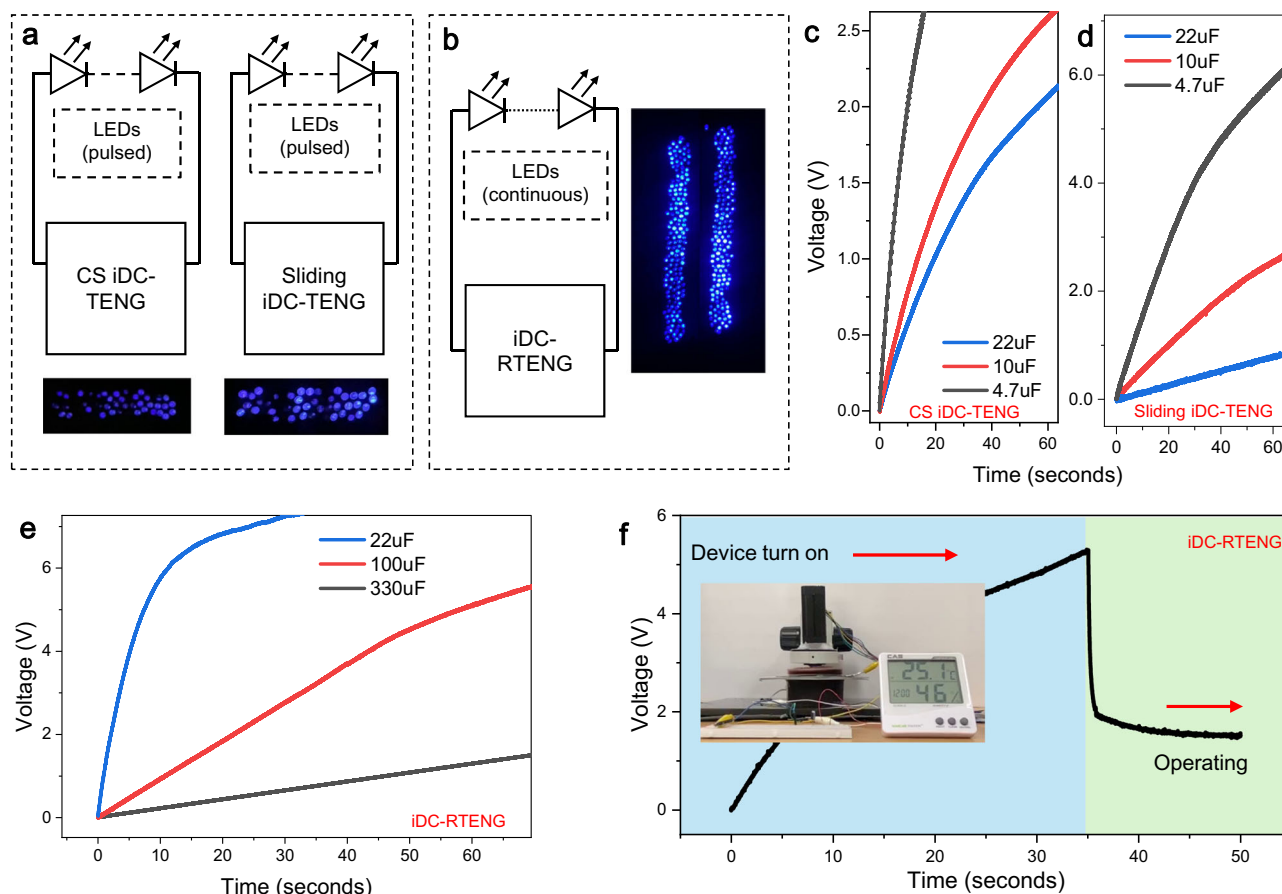


Fig. 7 | Demonstration of DC output applications of iDC-TENGs. **a** Pulsed LED (light-emitting diode) lighting powered by contact-separation and sliding iDC-TENGs (insets: photographs of illuminated LEDs; scale bars: 5 cm, left and right). **b** Continuous LED illumination driven by the iDC-RTENG (inset: photograph of continuously lit LEDs; scale bar: 4 cm). **c** Capacitor charging behaviour of the

contact-separation iDC-TENG. **d** Capacitor charging behaviour of the sliding iDC-TENG with capacitances of 4.7, 10, and 22 μF . **e** Capacitor charging behaviour of the iDC-RTENG for 22, 100, and 330 μF capacitors. **f** Real-time powering of an electronic device by the iDC-RTENG (inset: photograph of the experimental setup showing device connection and operation).

given area, a higher number of segments and increased speed can increase the output and efficiency of DC-based TENGs. Practical demonstrations of direct capacitor charging, LED illumination, and real-time device powering using the iDC-TENGs are presented in Fig. 7, with extended results in Supplementary Fig. 31 and Supplementary Movies 5 and 6. These results confirm stable DC generation across contact-separation, sliding, and rotary modes, further confirming its applicability across diverse mechanical inputs.

Through this work, we identify and validate a DC generation mechanism based on EP-induced ionic charge recombination in soft gels; distinct from conventional TENGs that rely on diode bridges and TVNGs that require rigid semiconductors. We show that plasticized PVC, traditionally a passive triboelectric layer, can be re-engineered into an active ionic-electronic material, supporting internal EDL formation and directional charge flow under mechanical input. This enables direct high-voltage DC output without rectification, bridging the mechanical-electrical conversion through polymer engineering. The mechanism is generalizable, but this study best optimizes it in PVC due to its processability and plasticizer compatibility. By tuning plasticizer type, ionic content, electrode configuration, and device structure, we reveal the interdependence between material properties and output behavior.

Achieving rectifier-free DC requires a targeted design strategy across material, structural, and operational levels for scaling and optimizing the mechanism. Dual-electrode setups are necessary to induce EP, limiting single-electrode use (Supplementary Fig. 10). Ionic

conductivity must be balanced: too low hinders mobility, too high causes leakage (Fig. 2e, Supplementary Fig. 8). Materials must also meet dielectric and triboelectric criteria, including sufficient surface charge density and dipole alignment. These principles enable stable DC output not only in contact-separation mode, but also in sliding and rotary configurations, all using a single material, which is a rare achievement in TENG or TVNG systems. The ability to deliver up to 200 V DC output, combined with durability and environmental robustness, establishes a scalable and flexible pathway for rectifier-free energy harvesting. Unlike prior PVC-based AC TENGs or rigid TVNGs, this work offers both a mechanistic foundation and a materials design framework for high-performance DC generation in soft, conformable electronics.

This study introduces and investigates ionic-based direct current triboelectric nanogenerators (iDC-TENGs), highlighting the potential of ionic-modified dielectric materials within a conductor-dielectric structure to achieve stable DC output. By tuning materials like plasticized PVC, we demonstrate that modifying dielectric materials with ionic components enables dynamic control over charge magnitude and direction, overcoming the traditional tradeoff between conductivity and triboelectric charge. We identify electrode polarization (EP) as a key mechanism for achieving DC output, shielding contact electrification during the operation. We further describe how EP influences DC generation in TENGs, revealing that work function orientation significantly impacts output form and magnitude, with the most effective orientation being a higher work function for the top

contacting electrode. Our findings also show that parameters like frequency, contact force, and load resistance play a critical role in optimizing DC behavior. Extending this approach, we successfully demonstrate stable DC generation in multiple modes, including contact-separation, sliding, and rotary motion. The palm-sized rotary TENG achieved continuous DC output of 200 V and 727 $\mu\text{A}/\text{m}^2$ with a low crest factor. This study emphasizes the significance of ionic modification in dielectric TENGs and provides a pathway to scalable, efficient DC energy harvesting, bridging the limitations of traditional TENGs and tribovoltaic nanogenerators (TVNGs) while advancing sustainable energy solutions.

Methods

Materials

The following materials were used for the preparation of plasticized PVC Gel, IL based PVDF, PDMS and PVC films. Commercial PVC powder (M_w 275,000) was sourced from Scientific Polymer Products, Inc., USA. Tetrahydrofuran (THF, 99.5 %) was utilized as the solvent, obtained from DAEJUNG, Korea. The plasticizers used in this study were dibutyl adipate (DBA), bis (2-ethylhexyl adipate) (DOA), and diisodecyl adipate (DDA), all procured from TCI, Japan. Additionally, 1-butyl-3-methylimidazolium bis (trifluoromethyl sulfonyl) imide (BMIM-TFSI) was purchased from TCI, Japan.

Synthesis of ionic Gel films

The PVC gel solution was prepared by dissolving the PVC powder in THF to create a homogeneous solution. Subsequently, plasticizers DBA, DOA, and DDA were added to the solution in specific PVC: plasticizer ratios of 1:0 (only PVC), 1:0.2, 1:0.4, 1:0.6, 1:0.8, 1:1, 1:3, and 1:5. The mixture was then stirred continuously for 12 h at a speed of 150 rpm and a temperature of 30 °C to ensure thorough mixing and dissolution. After this, the prepared PVC-plasticizer solutions were poured into glass dishes and left to dry for over 24 h at room temperature to ensure the complete evaporation of the solvent, THF. The resulting dried PVC gels were then cut into various sizes as required for subsequent testing and characterization. The thickness of the PVC gels was controlled by adjusting the amount of solution poured into the glass dishes during the casting process. This method ensured uniform distribution of the plasticizer within the PVC matrix and consistent gel properties across different samples. The PDMS-IL was prepared by stirring a silicone elastomer base with various weight percentages of ionic liquid, degassing the mixture, adding a curing agent, and then curing the samples. PVDF-HFP was used with the DMF solvent for the preparation of the films.

Fabrication of vertical contact separation iDC-TENG

For the vertical contact separation DC-TENG, glass substrates are utilized for device assembly. Copper tape is applied to the substrate as the bottom electrode, while an Au-coated wafer is affixed as the top substrate. The PVC gel layer is carefully deposited onto the bottom electrode.

Fabrication of foam-based lateral slider iDC-TENG

In the fabrication of the foam-based lateral slider DC-TENG, acrylic plate substrates are used as the base material. These substrates are covered with copper tape electrodes, and foam layers are attached beneath the PET electrode flaps to enhance flexibility and resilience. Electrical connectivity is established through a slip ring for integration into external circuits.

Foam-based rotary iDC-TENG

The foam-based rotary DC-TENG is fabricated using a carefully designed method. Acrylic plates are employed as substrates for both the rotor and stator components. The rotor is taped with copper. The segmentation of the rotor flaps is based on specific angles, with

options for 4 Segments–90°, 8 Segments–45°, and 12 Segments–30° PET film with copper tape is utilized for the rotor flap. Additionally, foam layers are affixed to the base of all flaps, providing mechanical support. For the stator, aluminium tape is used, and a large-sized PVC gel with an uncut radius of 10 cm is placed on it. The slip ring used for electrical connectivity is the “slip ring 8.5 mm 4 wire 1 A U type [SZH-SLI001],” sourced from Device Mart. This methodical fabrication approach ensures the efficient construction of the rotary iDC-TENG device.

Materials characterization and device performance

The electrical properties (capacitance, resistance, EIS and dielectric loss tangent) of the PVC gels were evaluated using an LCR meter (IM3533; HIOKI, Japan) at room temperature (25 °C) under standard conditions. Measurements were conducted over a frequency range of 1 Hz to 200 kHz with a signal amplitude of 1 V. To ensure accurate measurements, the films were sandwiched between two aluminium plates. Output voltage, current, and charge of all TENG devices were recorded utilizing a Keithley 6514 system electrometer (Keithley Instruments, Cleveland, OH, USA). Measurement setups included a shaker (ET-139; Labworks Inc., Costa Mesa, CA, USA), a function generator (33210 A; Keysight, Santa Rosa, CA, USA), and a linear power amplifier (PA-138; Labworks Inc.). The experiments were conducted at 20 N and 3 Hz, with a separation distance of 1 cm unless otherwise specified in the paper for measurements.

Data availability

All relevant data supporting the key findings of this study are available within the article and its Supplementary Information files. Source data are provided with this paper.

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Acknowledgements

This research was supported by the Nano & Material Technology Development Program through the National Research Foundation of Korea (NRF) funded by Ministry of Science and ICT(RS-2024-00451318). This work was also supported by the National Research Foundation of Korea (NRF) funded by the Ministry of Science and ICT(RS-2025-00560648).

Author contributions

J.-H.L. supervised the project. G.S.G. conceived the idea. G.S.G., H.P. and H.J. designed the experiments, fabricated the devices, and performed the data measurements. C.L. and S.G. assisted with experiments. G.S.G., H.-J.Y., H.R., S.M.L. and J.H.L. analyzed the data. G.S.G. drafted the manuscript with input from H.P., H.-J.Y., H.R., S.M.L., J.H.L. and J.-H.L. All authors discussed the results and contributed to the final manuscript.

Competing interests

The authors declare no competing interest.

Additional information

Supplementary information The online version contains supplementary material available at <https://doi.org/10.1038/s41467-025-64582-w>.

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Peer review information *Nature Communications* thanks Arunkumar Chandrasekhar, Wencong He and Linards Lapčinskis for their contribution to the peer review of this work. A peer review file is available.

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