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# Flexible stretchable tribo-negative films with exceptional output performance for high-temperature energy harvesting and self-powered sensor

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### ABSTRACT

High temperature is a significant cause of failure of electromechanical products, achieving effective collection of triboelectric energy at high-temperatures is a major research emphasis. Improving the electrical outputs from triboelectric nanogenerator (TENG) in extreme environments is crucial for its large-scale application and remains challenging. Further considering the major potential application for TENG is wearable electronics, a series of SEBS/fatty acid (SEBS/FA) skin-like flexible films were fabricated, which exhibit adjustable surface triboelectricity and enhanced triboelectric outputs at high-temperatures. The SEBS/FA based TENGs possess remarkable durability of over 10,000 contact-separation cycles, and exhibit exceptional electrical outputs in the heat. Thereinto, the output efficiency of SEBS-OA (octanoic acid) is up to 302.32 % at 110 °C and still above 100 % at 150 °C. The SEBS/FA based TENGs can efficiently harvest the mechanical energy in high-temperature environments to power commercial capacitors and LEDs. Notably, the SEBS/FA films still show enhanced electrical outputs even under extreme conditions with working ambient temperatures up to 90 °C and significant deformation ( $\lambda \leq 4$ ). The excellent electromechanical performances enable SEBS/FA based TENGs to be served as flexible energy collector and self-powered touch sensor for motion capture. The real-time tactile trajectory can be accurately mapped by a 5 × 5 sensing array. The SEBS/FA films exhibit great potential in energy harvesting and active real-time tactile sensing system in extreme environments.

#### 1. Introduction

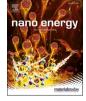
With the escalating requirements for the application of self-powered wearable microelectronics and self-powered sensing technology to scientific missions and energy generation in utmost environments, such as scorching and arid deserts, industrial plants, petroleum extraction, wildfires, military and space exploration, achieving effective collection of triboelectric energy at high-temperatures has gradually become an important research focus [1–3]. Therefore, the development of generators that can function efficiently in extreme temperatures has

outstanding application value but challenging to accomplish. This is mainly because, despite significant advances in triboelectric technology, it still faces several challenges to applying this technology to appalling conditions. For instance, military grade microelectronics need to work normally at drastic temperatures of up to 125 °C, where most traditional triboelectric materials fail to function [1,4]. It is well-known that the electrical output of triboelectric devices mainly rests on their surface charge density, which is largely affected by operating ambient temperature [5–7]. High temperature not only affects and destroys the chemical and physical structure properties of materials, thereby reducing

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their performance [3,8], but also is a major reason for the failure of a range of electromechanical products. Whereas, current triboelectric materials are principally customized for a single external environment (room temperature) energy collection, neglecting the impact of environmental variability (such as temperature) on the electrical outputs of triboelectric materials. Meanwhile, the fault of electromechanical equipments caused by high temperature produces enormous financial losses [7]. In this instance, the utilize of triboelectric nanogenerator (TENG) for high-temperature conditions is bound to be profoundly restricted. Consequently, further exploring advanced triboelectric materials from triboelectric series is the crux point to resolve this issue and one of the prime challenging assignments.

Polymeric materials, as the main conventional triboelectric matesuch as polytetrafluoroethylene (PTFE) [9,10], polyrials. dimethylsiloxane (PDMS) [11], polystyrene (PS) [12], poly(vinylidene fluoride-hexafluoropropylene) copolymer (PVDF-HFP) [13], polyurethane (PU) [14], polyamide 6,6 (PA66) [15], aramid [1] and fluorinated polyimide (F-PI) [6], have the virtues of diverse material choices, marvelous flexibility, processability and light quality [16], and are commonly employed to manufacture TENGs. However, electrons migrated to the surface of the above polymeric materials may be loosed into the vacuum after contact electrification (CE) in view of thermionic emission effect, bring about the inability of the corresponding TENGs to maintain efficacious electrical outputs at elevated temperature conditions [1,17]. Recently, there have indeed been many studies on TENG devices that can operate at high temperatures [7,12,15,18]. Despite their steady operation under high temperature conditions, these TENG devices experience a significant decrease in electrical output performance due to continuous thermionic emission effect [7,12,15,18]. For instance, the output efficiency of PI based TENG [7] and PS based TENG [12] can only maintain 52.12 % at 120 °C and 11.42 % at 100 °C, respectively, which is even worse. In addition, most of them are synthetic polymers with high costs, non-stretchable and non-degradable, which also hinder the practical application of TENG. Further considering the major potential application for TENG is wearable electronics, flexibility and stretchability are critical for triboelectric materials utilized in TENGs [19,20]. However, up to now, there are few reports on flexible and stretchable triboelectric materials with exceptional electrical outputs for mechanical energy harvesting in high-temperature environments.

For wearable electronics, a skin-like flexible substrate that can accommodate deformation during body movement should be used. Through detailed investigation, it is found that poly[styrene-b-(ethylene-co-butylene)-b-styrene] (SEBS), as one of the most frequently applied styrene-based thermoplastic elastomers, has extraordinary properties including exceptional thermal and chemical resistance, good flexible, resilience and stretchability, and can accommodate a variety of different deformation [21,22]. Additionally, it is also biodegradable and environmental friendly. Hence, SEBS can be considered as one of the most appealing flexible and stretchable materials for making use of the TENG fabrication for extensive mechanical energy harvesting [22–24]. However, so far, there are few studies about SEBS as the triboelectric material for fabricating TENG. Meanwhile, SEBS belongs to the triboelectric material located in the triboelectric series with slightly negative polarity [25]. There is still some way off its commercial use due to the insufficient electrical outputs of TENG. More importantly, one of the critical challenges currently recognized in TENG-based wearable power supplies is how to boost the electrical outputs of triboelectric materials, especially in harsh environments [26,27]. Therefore, improving the electrical outputs of SEBS based TENGs in a wide temperature range is crucial for expanding its large-scale applications, particularly for flexible and wearable bioelectronics [26,27].

In this study, SEBS, a skin-like flexible substrate, is first used as tribonegative material to prepare flexible stretchable TENG. Fatty acids (FA) are used as dopant to further improve the electronegativity of SEBS, to enhance the electrical outputs of SEBS based TENGs and promote their large-scale applications. The relevant results show that, at an optimized OA (octanoic acid) concentration of 1.0 wt%, the dielectric constant and surface potential at most 54 % and 587 % respectively increase compared to pure SEBS. Energy-harvesting depends on the SEBS-OA based TENG, which represents the maximum  $V_{oc}$  of 47 V, 3.27 times that of pure SEBS film, significantly higher than the previous technical levels of available rubber elastic triboelectric materials with similar structures [28]. Meanwhile, when the resistance is about 30 M $\Omega$ , the SEBS-OA based TENG has a maximum output power of 131.92 µW, which is 10.72 times of that of pure SEBS based TENG. Specially, all SEBS and SEBS/FA films possess excellent enhanced electrical outputs at high-temperatures and outstanding stretchability. The SEBS/FA based TENGs can efficiently harvest the mechanical energy in high-temperature environments to power commercial capacitors and LEDs, and the charging voltage of capacitors and the brightness of LEDs increase significantly with increase of operating ambient temperature. Additionally, the SEBS/FA based TENGs still show enhanced electrical outputs even under extreme conditions with working ambient temperatures up to 90 °C and significant deformation ( $\lambda < 4$ ). Therefore, the SEBS/FA films possess excellent enhanced triboelectric performance at high-temperature environments and different deformation. The excellent electromechanical performances enable the SEBS/FA based TENGs to be used as a flexible energy harvester and self-powered tactile sensor for motion capture. This work is the first to use SEBS as a triboelectric material and realize efficient collection of triboelectric energy at high temperatures. Sophisticated selection/preparation of SEBS/FA composite films not only offers a promisingly new approach for developing advanced TENGs with aggrandized triboelectric outputs over a wide temperature range and tensile strain, but also shows great potential in energy harvesting and active real-time tactile sensing system in harsh environments.

### 2. Results and discussion

Fig. 1a illustrates the detailed structure diagram of the singleelectrode mode SEBS/FA based TENGs, which comprise SEBS/FA film, commercial PI film and Cu tape as the tribo-negative layer, dielectric layer, and electrode, respectively. Fig. S1 displays the typical fabrication process of SEBS/FA composite films through a simple blade coating technology. The as-fabricated SEBS/FA films are shown in Fig. 1b, which exhibit excellent transparency. Fig. 1c-f and Fig. S2a depict the FT-IR spectra of SEBS/ODA (octadecenoic acid) films with different ODA doping content, as well as the electrical outputs of SEBS-ODA based TENGs. As depicted in Fig. 1c and Fig. S2a, the peak observed at 1711  $cm^{-1}$  corresponds to the C=O group of ODA [12], implying that the SEBS/ODA films contain ODA molecules. And with increasing ODA doping content, its intensity increases gradually. It is well-known that the majorization of mixing conditions is the hinge to realizing dependable output performance of composite based TENGs [29,30]. All electrical outputs of SEBS and SEBS/ODA based TENGs with different ODA doping content are investigated using a Cu tape triboelectric pair under the same conditions at 1.5 Hz operating frequency, and the corresponding results are dispalyed in Fig. 1d-f. It can be found that the electrical outputs of SEBS/ODA based TENGs increase with higher doping content of ODA, and the maximum peak-to-peak voltage  $(V_{oc})$ , transferred charge  $(Q_{sc})$  and short-circuit current  $(I_{sc})$  were recorded for the SEBS/ODA films with a 1.0 wt% ODA, which are 2.84, 2.55 and 5.27 times of that of the pure SEBS film, respectively. To more evidence the impact of ODA on the output characteristics of SEBS based TENGs, the electric power of SEBS and SEBS-ODA (1.0 wt%) based TENGs were measured by loading different external resistances. As presented in Fig. 1g, the output power of SEBS-ODA (1.0 wt%) based TENG achieved the maximum value of 93.17  $\mu W$  when the resistance is about 30 MΩ, which is 7.57 times of that the pure SEBS with the value of 12.31  $\mu$ W. The above results distinctly indicate that doping ODA into SEBS is an useful way to enhance the triboelectric outputs of SEBS based TENGs,

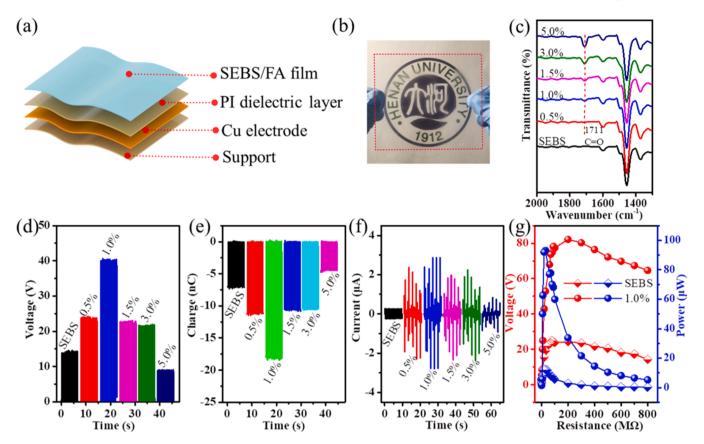


Fig. 1. (a) Structure diagram of SEBS/FA based TENGs. (b) Photo of SEBS/FA films. (c) FT-IR spectra of SEBS-ODA films with different ODA content. (d-f) Electrical outputs of SEBS-ODA based TENGs with different ODA content. (g) The effective electric power of SEBS and SEBS-ODA (1.0 wt%) based TENG.

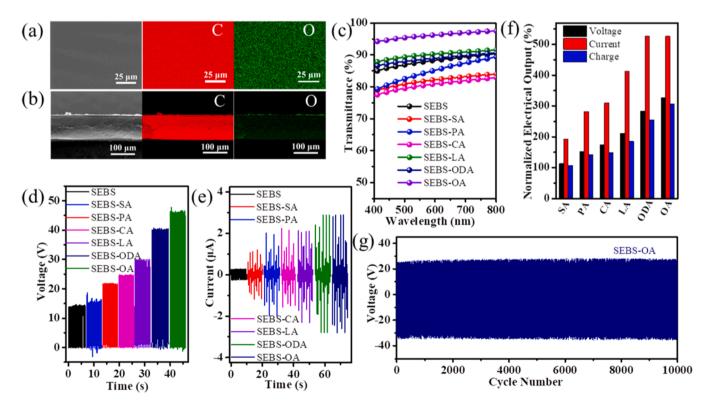


Fig. 2. (a) Surface SEM and relevant EDS mapping images of SEBS-OA film. (b) Sectional SEM and relevant EDS mapping images of SEBS-OA film. (c) Transmittance of SEBS and SEBS/FA films. (d, e) Electrical outputs and (f) normalized electrical outputs of the SEBS and SEBS/FA based TENGs. (g) Mechanical stability of SEBS-OA based TENG.

and the SEBS-ODA (1.0 wt%) film has a stronger triboelectric ability.

As is known from the above, ODA, as one of the fatty acids (FA), can effectively improve the triboelectric outputs of SEBS. To verify the universality of SEBS triboelectric outputs improved by fatty acids, a series of SEBS/FA composite films with a 1.0 wt% doping content were fabricated, such as SEBS-SA (stearic acid), SEBS-PA (palmitic acid), SEBS-CA (capric acid), SEBS-LA (lauric acid) and SEBS-OA (octanoic acid). Previous studies have shown that the homodisperse of fillers in polymer matrices is one key point to generate excellent triboelectric outputs [31]. Therefore, taking the SEBS-OA film as an example, the distribution of fatty acids in polymer matrix SEBS was firstly studied by EDS mapping and elemental analysis, and the corresponding results are exhibited in Fig. 2a-b and Fig. S3. Fig. 2a-b exhibit the surface and cross-sectional morphologies of SEBS-OA film, as well as the corresponding elemental mapping images of SEBS-OA film, respectively. As seen in Fig. 2a-b and Fig. S3, the even distribution of C and O elements in the film evidences the homodisperse of fatty acids in SEBS matrix and the favorable compatibility of fatty acids and SEBS matrix.

Next, the effects of fatty acids (i.e., OA, ODA, LA, CA, PA, and SA) doped with 1.0 wt% on the output performance of SEBS matrix were also investigated, the relevant results are manifested in Figs. 2d-2f and Fig. S4. As exhibited in Fig. 2d-f and Fig. S4a, all SEBS/FA films have much higher electrical outputs than that of pure SEBS film, indicating that it is a widespread method to enhance the triboelectric characteristics of SEBS matrix by doping fatty acids. Thereinto, as shown in Fig. 2d, the SEBS-OA film shows the highest triboelectric output with a  $V_{oc}$  of about 47 V, which is 3.27 times that of pure SEBS film, prominently higher than the previous technical levels of available rubber elastic triboelectric materials with similar structures (such as, styreneisoprene block copolymer [19,32], bromobutyl rubber [33], and natural rubber [34]). Meanwhile, as exhibited in Fig. 1g and Fig. S4b, the SEBS-OA based TENG has a maximum output power of 131.92  $\mu W$  when the resistance is about 30 M $\Omega$ , which is 10.72 times of that of pure SEBS. From Fig. 2g, it can be observed that SEBS/FA films could be function consecutively for over 10,000 cycles without degrading output performance, which explicates that the SEBS/FA films exhibit a transcendental mechanical stability and the SEBS/FA based TENGs have excellent durability. Moreover, as shown in Fig. 1b and Fig. 2c, the SEBS/FA

composite films have excellent transparency. Combined with transparent electrodes, the SEBS/FA films show enormous promise in manufacturing transparent TENG devices.

Due to the operating mechanism of TENG being a conjunction of CE and electrostatic induction [35], the output performance can be improved through increasing the triboelectric polarity difference between two contact surfaces in order to add the amount of charge generated through triboelectric effect [36]. Therefore, to unveil the mechanism of fatty acids improving the triboelectric outputs of SEBS based TENG, the electronegativity of SEBS and SEBS/FA films were first studied. It is widely known that the group electronegativity value of -COOH is greater than that of -C<sub>6</sub>H<sub>5</sub> [12], which means fatty acids are more easily to attract a pair of shared electrons than SEBS. In view of this, it is reasonable to believe that doping fatty acids can change the triboelectric polarity and charge retention ability of the SEBS matrix film, which could enhance the total charge capacitance of SEBS/FA films, thereby greatly improving the output characteristics of SEBS based TENGs. To attest this guess, we devised a TENG consisting of pure SEBS and SEBS-OA films, as presented in Fig. 3a. In addition, Fig. 3b comes out the collected instantaneous electrical output signal from this configuration. From Fig. 3b, it can be clearly seen that the signal has a transient positive and negative peak form, which signifies that SEBS-OA film is more triboelectronegative than the pure SEBS film. The above result not only is coincident with electronegativity, but also suggests that the triboelectronegativity of SEBS can be improved doping fatty acids to improve the triboelectric output performance of SEBS based TENGS

Further, abundant studies have indicated that the surface potential of triboelectric materials is a prominent element determining the output characteristics of a TENG [29,37]. KPFM has been extensively used to study the surface charge distribution within the triboelectric interface. As a tribo-negative material, SEBS typically exhibits a negative surface charge potential, and the change to a more negative surface potential can result in a greater electrical outputs [37]. Fig. 3c-d provide the KPFM results of the pure SEBS and SEBE/FA films. From these figures, all SBES/FA films exhibit a more negative surface potential compared with pure SEBS film, which indicates that the addition of fatty acids could increase the charge capture capacity to enhance the electrical

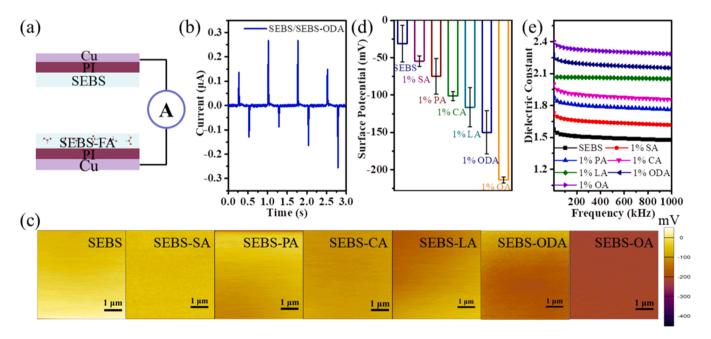


Fig. 3. (a) Diagram of measurement connection for discriminating the triboelectric series of SEBS films with and without FA doping. (b) Output signal generated by friction between SEBS-OA film and pure SEBS film. (c, d) Kelvin probe force microscope (KPFM) results of SEBS and SEBS-FA films. (e) Correlation of dielectric constants for SEBS and SEBS-FA films.

outputs of SEBS based TENGs. Moreover, the changing tendency of the surface potential of SEBS/FA films is accordance with that of the triboelectric outputs of SEBS/FA based TENGs in Fig. 2d-e and Fig. S4a above. And the SEBS-OA film shows a maximum surface potential value of -213 mV, which is about 6.9 times that of pure SEBS film.

Meanwhile, as a capacitive energy equipment, the triboelectric characteristic of the TENG widely depends on its capacitance [37,38]. The higher permittivity typically results in higher capacitance and greater charge transfer density, thus helping to enhance the electrical outputs of TENG [13]. In view of the positive relationship between surface charge transfer density and permittivity, upgrading the dielectric properties of triboelectric materials is very important for enhancing the triboelectric outputs of TENG. Fig. 3e displays the dielectric constant (permittivity) of pure SEBS and SEBS/FA films doped with different fatty acids. As shown in Fig. 3e, all as-fabricated SEBS/FA films possess a significantly higher dielectric constant than pure SEBS film, and similar trends were observed in the electrical outputs and surface potential results for SEBS/FA films. This is mainly because: (i) when the addition amount of FAs is fixed 1 wt%, the smaller the relative molecular weight of FAs, the stronger the ability to change the polarity and charge retention ability of the SEBS film. As a result, the higher the surface potential, dielectric constant, and electrical output of the prepared SEBS/FA films. (ii) Recent studies have found that the introduction of proper liquid lubrication can not only provide a super wear-resistive TENG inhibiting wear and transfer of polymer tribomaterials, but also increase the charge-capturing capability of the triboelectroactive layer, thereby increasing the electrical output of the TENG device [12,39]. As well known, FAs are considered to be one of the classical boundary lubrication additives [40]. Compared with solid FAs (SA, PA, etc), the presence of liquid FAs (ODA, OA) lubricants can increase the effective solid-solid contact area, capture more transferred electrons, and thus improve the total charge capacitance and dielectric constant of SEBS/FA film, resulting in higher electrical output [12,39–41]. Considering the influence of the above two factors, the surface potential, dielectric constant and electrical output of SEBS/FA films showed the trend as shown in Fig. 2d-e and Fig. 3c-e. In conclusion, based on the above characterization results, it can be validated again that doping fatty acids to ameliorate the triboelectric characteristics of SEBS matrix is a general method.

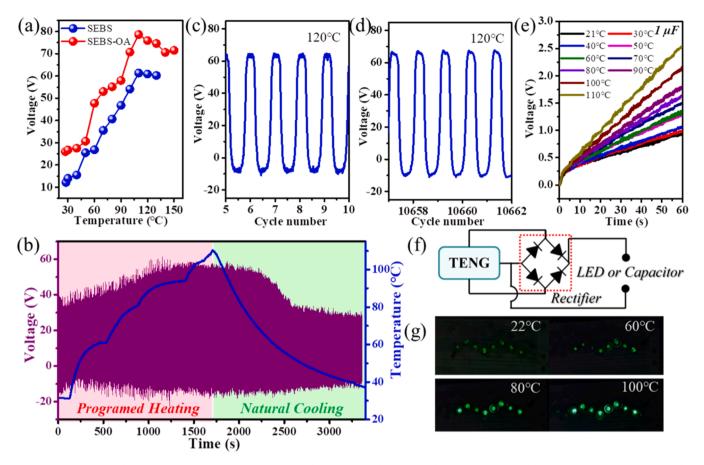
The TENG is known for its ability to effectively translate small mechanical energy into high-performance power supplies for portable and wearable electronics, which has aroused widespread interests and exhibited immense application potential [42,43]. However, the triboelectric layers of TENG are seriously affected by operating ambient temperature [4,9]. The high temperatures not only influence the triboelectric outputs of TENG, but also is a major reason for the failure of a range of electromechanical products, which undoubtedly limits the further large-scale applications of TENG in harsh environments. Additionally, with the growing requirement for self-powered wearable microelectronics for scientific missions and energy generation in harsh environments, there is a significant demand for high-temperature energy collectors to power low-power sensors [44]. Consequently, excavating advanced triboelectric materials with exceptional triboelectric output performance, excellent flexibility and stretchability for mechanical energy harvesting in high-temperature environments is crucial to expand their large-scale applications, especially for flexible and wearable bioelectronics.

In this study, through detailed investigation, SEBS is first used as tribo-negative material to prepare flexible stretchable TENGs. Fatty acids as dopants further improve the electronegativity of SEBS and enhance the output characteristics of SEBS based TENGs. As seen in Fig. S2b and Table S1, the initial thermal decomposition temperature of the fabricated SEBS/FA films is above 400 °C, exhibiting excellent thermal stability and environmental temperature stability, which are favorable when considering their potential applications in energy harvesting and active real-time tactile sensing system in extreme

environments. On top of that, and more importantly, as presented in Fig. 4 and Figs. S5-S8, all as-fabricated SEBS and SEBS/FA films possess exceptional amplified electrical outputs at high-temperatures and outstanding stretchability. From these figures, it can be visually seen that the electrical outputs of SEBS and SEBS/FA based TENGs could remain steady at each temperature point. The  $V_{oc}$ ,  $I_{sc}$  and  $Q_{sc}$  show an obvious increasing tendency with increasing temperature (from room tempetature to 110 °C or 120 °C), but the output voltage has a decreasing tend when the temperature exceeded the critical point. Still, even at 150 °C, their electrical outputs are still noticeably higher than that at room temperature. This phenomenon can be explicated by electron-driven relevant factors and material relevant factors, both of which are influenced when operating at high-temperatures [17,45,46]. First, the former factor: rising temperature accelerate the electron transfer and introduce disorder, which is related to the driving force of electron shift from one layer to another, producing a better output at higher temperatures [47]. However, enormous thermal changes may conquer barrier heights, promote reverse electron transfer processes, and reduce the output characteristics [17]. Hence, there exists an optimum temperature point for obtaining the best electrical outputs. Second, the latter factor: for the TENG, mechanical strength is the dominant element determining the friction efficiency, as the polymers are impressionable to heat, their properties intensively depend on the operating temperature, and both ductility and stiffness change as the temperature increases [17]. Thus, a suitable temperature is demanded to promote the friction process, thereby increasing the output characteristics. As displayed in Fig. 4a and Fig. S7c, it can be concluded that the temperature range of 110-120 °C is the ideal temperature range for obtaining the optimum electrical outputs of SEBS/FA based TENGs. Thereinto, the output efficiency ( $V_T/V_{Room}$ ) of SEBS-OA at 110 °C is up to 302.32 % and is still higher than 100 % at 150 °C. Specially, the  $V_T/V_{Room}$  of SEBS-ODA at 110 °C is as high as 797.93 %. Moreover, Fig. 4b displays the Voc of SEBS-OA based TENG during consecutive procedural heating and natural cooling, from which it can be clearly seen that the  $V_{oc}$  exhibits a distinct increasing trend with increasing operating temperature, which is completely consistent with the temperature-by-temperature test results in Fig. 4a and verified again that the SEBS/FA films prepared in this work have exceptional enhanced output characteristics at high-temperatures.

In practical applications, in addition to exceptional output performance, high-temperature stability, repeatability and durability are also extremely vital property indexes. Therefore, we measured the output voltage under more than 10,000 external force tests at 120 °C, and the correspond results showed that the SEBS/FA based TENGs have outstanding high-temperature stability and durability, as shown in Fig. S7d. From Fig. 4c-d, the output voltage of SEBS/FA based TENGs not only don't decay, but also exhibits a slight increase after a long working cycle at 120 °C high temperature condition. Additionally, as listed in Table S2, the fabricated SEBS-OA film in this study not only possesses higher output voltage at high temperature, but also shows significantly higher normalized voltage values at various temperatures than those of the triboelectric materials at corresponding temperatures reported in similar studies [7,15,48-52]. Meanwhile, the SEBS-OA film also exhibits excellent stretchability and transparency. In summary, the SEBS/FA films fabricated in this study have various excellent characteristics at a wide temperature range and high temperatures, making them widely suitable for energy harvesting and wearable self-powered sensor devices in extreme temperature environments.

To explicate the practical applications under high temperature operating conditions, the SEBS/FA based TENGs were placed on a hot stage to simulate energy harvesting in high-temperature environments. Noteworthy, as shown in Fig. 4e-g, the SEBS/FA based TENGs can efficiently harvest the mechanical energy in high-temperature environments to power commercial capacitors and LEDs, and the charging voltage of the capacitors and the brightness of LEDs increase significantly with increasing operating temperature. The detailed process can



**Fig. 4.** (a) Temperature-related output voltage of SEBS and SEBS-OA based TENGs. (b) Output voltage of SEBS-OA based TENG during consecutive procedural heating and natural cooling.  $V_{oc}$  before (c) and after (d) stability and robustness test of SEBS-OA based TENG at 120 °C. (e) Charging characteristic of SEBS-OA based TENG to a commercial capacitor. (f) Circuit diagram of SEBS-OA based TENG for charging capacitors or lighting LEDs. (g) Photos of SEBS-OA based TENG to light commercial LEDs at different operating temperatures.

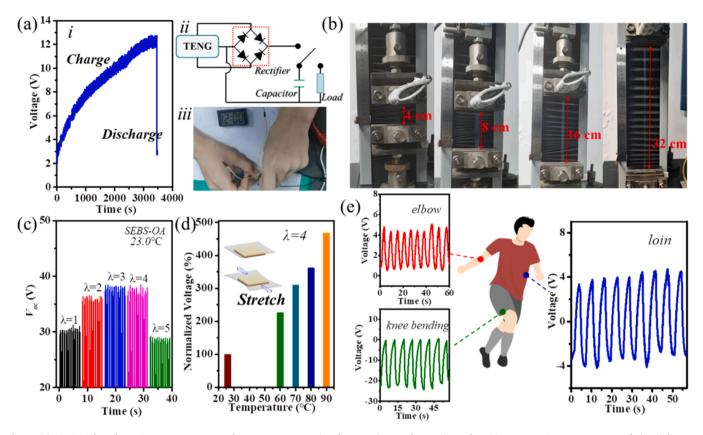
be observed in the Video S1. Fig. 4f shows the circuit diagram of SEBS-OA based TENG for charging capacitors or lighting LEDs. Even at 130 °C operating temperature, the SEBS-OA based TENG has been proven to be as self-powered system that can charge a 4.7  $\mu$ F commercial capacitor, which still can power the electronic watch, as displayed in Fig. 5a. The corresponding real-time charge-discharge curve of the 4.7  $\mu$ F commercial capacitor powering the electric watch is displayed in Fig. 5ai. These results demonstrate the great potential of SEBS/FA based TENGs for energy harvesting and self-powered microelectronic systems in harsh environments. To sum up, this study provides an efficacious solution for self-powered microelectronics applied in high-temperature harsh environments, which not only effectively improves the electrical outputs of triboelectric material SEBS, but also perfectly solves the limitation of practical application of TENG in high-temperature conditions.

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Considering the major potential application for TENGs is wearable electronics, flexibility and stretchability are critical for triboelectric materials utilized in TENGs [19,20]. Therefore, for wearable electronics, a skin-like flexible substrate that can accommodate deformation during body movement should be used. Thanks to good flexible, resilience and stretchability of SEBS, the as-fabricated SEBS/FA films exhibit outstanding stretchability as well, which is demonstrated by the photographs in Fig. 5b. Further, their mechanical properties were estimated through uniaxial and cyclic tensile tests. As seen in Fig. S8a, the as-fabricated SEBS/FA films (thickness of about 105  $\mu$ m) rupture at a stress of 6.83–27.63 MPa respectively and reach the tensile strain of 500 %– 821 % (strain rate 50 mm/min). Benefiting from lower tensile

stress, the SEBS/FA films exhibit favorable recovery even by applying 500 % tensile strain 5 cycles, as displayed Figs. S8b-S8h, which indicates that SEBS/FA films can recover well upon cyclic stretching. Additionally, since human epidermal skin is rarely revealing to more than 100% tension in daily life, the as-fabricated SEBS/FA films are suitable for stretchable human skin to capture biomechanical energy. Subsequently, the triboelectric output voltage of SEBS-OA film under different uniaxial strain conditions was studied, as displayed Fig. 5c. The output voltage of SEBS-OA film exhibits a trend of enhancement when the strain does not exceed 400 % ( $\lambda = 4$ ), which is consistent with recent literature report [53]. The increase of output voltage is attributed to the variations of thickness of tribo-negative layer [53]. In detail, the output voltage achieves approximately 38 V at stress state ( $\lambda = 4$ ), which is higher than that of 10 V at initial state without strain ( $\lambda = 1$ ). As well, taking SEBS-OA film as an example, it can be observed intuitively from Fig. 5d and Fig. S9 that the electrical outputs of SEBS/FA films under different uniaxial strain conditions could remain stable at each temperature point and show an obvious increasing tendency with increasing temperature. As shown in Fig. 5e, a series of experiments has verified that the flexible stretchable SEBS/FA based TENGs can capture biomechanical energy from various types of body movements. These results prove once again that the as-fabricated SEBS/FA films have enormous promise in the field of energy harvesting and self-powered wearable electronic devices in harsh environments.

The excellent electromechanical performances enable SEBS/FA based TENGs to be served as a flexible energy harvester, but also as self-powered tactile sensor for motion caption. As schematically shown in Fig. 6a, the triboelectric sensor array (TSA) consists of  $5 \times 5$  pixels, and

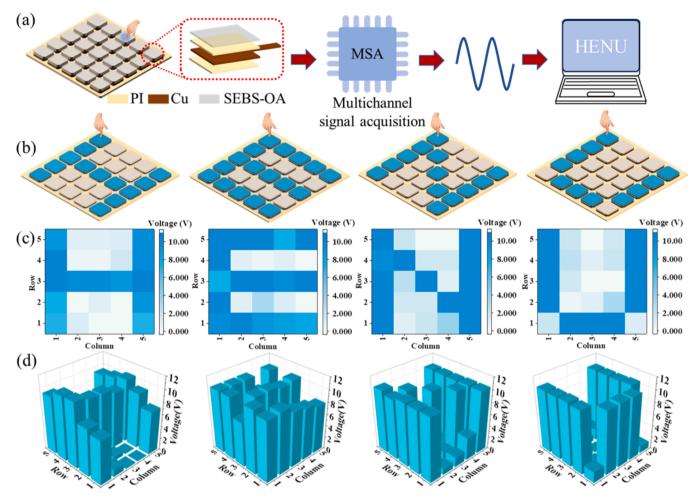


**Fig. 5.** (a) SEBS-OA based TENG as power source to charge a 4.7  $\mu$ F capacitor for powering an electronic watch at 130 °C operating temperature and circuit diagram for SEBS-OA based TENG works as power source for electronic device. (b) Photos of the SEBS-OA triboelectric layer at initial state ( $\lambda = 1$ ) and various tensile states. (c) Output voltage of the SEBS-OA based TENG under various tensile states. (d) The corresponding output voltage of SEBS-OA based TENG when the  $\lambda = 4$  and at different operating temperatures. (e) Low-frequency biomechanical energy-harvesting performances of SEBS-OA based TENG as a wearable device.

each pixel contains four layers of materials from top to bottom, including a SEBS-OA layer, a PI dielectric layer, a Cu electrode layer, and a supportive PI layer. The sensor performance of TSA was systematically characterized by the testing system as displayed in Fig. 6a, And the operating principle is displayed in Fig. S10. Briefly, when the TSA is touched, a corresponding electrical output will be generated at the corresponding pixel of the TSA by the coupling of CE and electrostatic induction. Sequentially, the electrical outputs were registered through a multi-channel data acquisition card system, and eventually recorded by a computer. As illustrated in Fig. 6b, to study the application of character identification, we showed a testing through writing "H", "E", "N", and "U" on a  $5 \times 5$  pixels TSA in sequence. By processing the real-time output mapping accordingly, the write trajectory can be acquired based on the recorded output peak voltage [54,55]. The corresponding 2-D and 3-D maps are respectively displayed in Fig. 6c-d. By setting 12.00 V as the cutting off value in the 2-D and 3-D maps, the "H", "E", "N", and "U" patters can be clearly recognized. The experiment results exhibit that the TSA has excellent property in tactile sensing, particularly in contact point detection and trajectory recognition, which reveals the feasibility and practicability of the TSA for real-time tactile sensing. Therefore, the SEBS/FA films fabricated in this study exhibit great potential in energy harvesting and active real-time tactile sensing system in extreme environments.

# 3. Conclusion

A series of SEBS/FA films were prepared through facile blade coating technique and appropriate ratio optimization. The introduction of FA enables the SEBS/FA composite films to exhibit tunable surface triboelectricity, and the fabricated SEBS/FA films show significant improvement in dielectric property and electronegativity. All SEBS/FA films have much higher electrical outputs than that of pure SEBS film, indicating that it is a widespread method to enhance the triboelectric characteristics of SEBS matrix by doping fatty acids. Thereinto, the SEBS-OA film shows the highest triboelectric output with a  $V_{oc}$  of about 47 V, which is 3.27 times that of pure SEBS film. Meanwhile, the SEBS-OA based TENG has a maximum output power of 131.92 µW when the resistance is about 30 M $\Omega$ , which is 10.72 times of that of pure SEBS. More significantly, all as-fabricated SEBS and SEBS/FA films have remarkable improved electrical output performance at hightemperatures and outstanding stretchability. The output efficiency of SEBS-OA at 110 °C is up to 302.32 % and is still higher than 100 % at 150 °C. Specially, the  $V_T/V_{Room}$  of SEBS-ODA at 110 °C is as high as 797.93 %. Additionally, the SEBS/FA based TENGs can efficiently harvest the mechanical energy in high-temperature environments to power commercial capacitors and LEDs, and the charging voltage of the capacitors and the brightness of LEDs increase significantly with increase of operating ambient temperature. Notably, the SEBS/FA films still show enhanced electrical outputs even under extreme conditions with working ambient temperatures up to 90 °C and significant deformation ( $\lambda < 4$ ). The excellent electromechanical performances enable the SEBS/ FA based TENGs to be served as a flexible energy harvester and selfpowered tactile sensor for human motion capture. The sensing capability of SEBS/FA based TENGs is demonstrated by a 5  $\times$  5 sensing array to trace the tapping positions by generating electrical signals. This study provides an effective solution for efficient collection of triboelectric energy in extreme environments, which not only proposes an advanced tribo-negative material with improved output performance over a wide temperature range and tensile strain, but also further facilitates the large-scale applications of TENG devices, especially for flexible and wearable bioelectronics in extreme environments.



**Fig. 6.** (a) Tactile sensing signal recording system based on SEBS-OA based TENGs. (b) Schematic of a finger writing the alphabets "H" "E" "N" "U" (short for Henan University) on the surface of 5 × 5 pixelated SEBS-OA based TENGs. (c) Top views of 2D mapping and (d) 3D diagram of output voltage intensity when a finger writes the alphabets "H" "E" "N" "U".

# 4. Experimental section

*Preparation of SEBS and SEBS/FA films*: Octadecenoic acid (ODA) with different content (i.e., 0.0, 0.5, 1.0, 1.5, 3.0 and 5.0 wt%) was combined with 4 g poly[styrene-*b*-(ethylene-*co*-butylene)-*b*-styrene] (SEBS, G1650, Kraton) triblock copolymer and 16 g toluene (AR) solvent in a 50-mL sample flask. The above mixture was stirred for 6 h to prepare the homogeneous and transparent SEBS-ODA solutions. Then, the prepared SEBS/ODA solution was scraped onto a flat glass surface and dried naturally to obtain the corresponding SEBS-ODA films with different ODA content, as shown in Fig. S1. In addition, the SEBS-SA (stearic acid), SEBS-PA (palmitic acid), SEBS-CA (capric acid), SEBS-LA (lauric acid) and SEBS-OA (octanoic acid) composite films with a 1.0 wt% doping content were prepared individually by the same operation as above.

Assemble of SEBS and SEBS/FA based TENGs: The layer-by-layer selfassembled sandwich structure composed of PI film and copper (Cu) tape was cut into  $4.5 \times 4.5$  cm<sup>2</sup>, wherein PI film was respectively used as support substrate and dielectric layer, and the Cu tape was used as electrode layer. After that, the SEBS and SEBS/FA films were separately assembled into single-electrode mode TENG as displayed in Fig. 1a.

SEBS-OA based triboelectric sensor array (TSA) for self-powered tactile sensing: The PI film  $(6.5 \times 6.5 \text{ cm}^2, \sim 100 \,\mu\text{m}$  thick) was as the support substrate. Twenty-five SEBS-OA based sensor units  $(0.5 \times 0.5 \text{ cm}^2,$ 1.0 cm interval) were anchored on the support substrate. Each terminal of triboelectric sensor unit was connected with Cu wire for electrical measurement. Finally, a fully integrated flexible TSA, which composed of a 5  $\times$  5 SEBS-OA based sensor units, was assembled for self-powered tactile sensing.

*Characterization*: The chemical structural property, surface and crosssection morphology, transmittance, triboelectric outputs, dielectric spectroscopy, and surface potential of the fabricated SEBS and SEBS/FA films were investigated using spectroscopic and microscopic analyses, a high-impedance electrometer, KPFM-mode AFM, and so on. For more deails, see Supporting information.

## CRediT authorship contribution statement

Yangjiu Zhao and Shaowei Shen performed the experiments, data collection and analysis. Ruirui Cao contributed to conceptualization, formal analysis, the original manuscript writing, funding acquisition, and resources. Haoyi Wu, Haoran Yu, and Xin Li assisted in data analysis. Weifeng Zhang contributed to supervision. Caofeng Pan commented on and revised the manuscript, and contributed to funding acquisition, and resources.

## **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

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# Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.nanoen.2023.108654.

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