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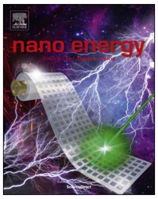

A size-unlimited surface microstructure modification method for achieving high performance triboelectric nanogenerator

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ABSTRACT

Triboelectric nanogenerator (TENG) is a promising mechanical energy harvesting device, which has many advantages such as low cost, high efficiency and easy fabrication. Surface modification on tribo-surface has been proved to be an efficient way to improve the output performance of TENG. However, the existing methods are time-consuming, high-cost and difficult for large-area processing, which, to some extent, limited the industrial production of TENG technology. In this work, we demonstrated a simple, fast and low-cost method, in which the tribo-surfaces of TENG were processed without size limitations to improve its output performance. Through this method, a large-scale treated TENG can be easily achieved and its output voltage and power can be increased by a factor of 3 and 5, respectively, providing us a size-unlimited, low-cost and time-saving surface micro-structure modification method for improving the output of TENG. This work showed great chance for industrial fabrication of TENG, which was crucial for sustainable power supply in the future.

1. Introduction

Triboelectrification and static electrification are common phenomenon in nature. However, the triboelectricity and static electricity were hard to be utilized in the past and thought to be harmful to industry and human health, so people have made many efforts to eliminate and prevent the triboelectricity and static electricity [1]. Recently, a novel device termed triboelectric nanogenerator (TENG) has attracted growing interests, which can harvest triboelectricity and static electricity caused by mechanical motion in the surrounding environment and power electronics [2]. The energy sources of the TENG were ubiquitous, ranging from large-scale energy to small-scale energy, such as the wind, tide, walking, touching, breathing and heart beating and so on [1,3–9]. Considering its high efficiency, low cost, light-weight and easy fabrication, the TENG can be as one of the most promising devices to provide sustainable power source under the background of energy exhaustion and degradation [1,10–14].

In the past few years, researchers have done much work to improve the output performance of TENG, including optimizing its structure [15–19], surface morphology [20–24] and materials [26–28]. The output performance of TENG was significantly improved by modifying the tribo-surface of TENG at microscale or nanoscale, which obviously increased the friction area. Hence, people produced different micro/nano patterns on tribo-surface [20–25,29]. In 2012, Fan et al. fabricated micro patterns on PDMS surfaces by soft lithography, such as line, cube and pyramid. The pyramid pattern had provided the maximum output, which was about 4 times of that with plane surface [20]. In 2014, Lee et al. obtained dots, lines, holes and rings at nanoscale through block copolymer (BCP) technology. In addition, inductively coupled plasma (ICP) treatment method was also widely used to form microstructures on polymer films [30,31]. However, these methods are time-consuming, high-cost and difficult for large-area processing, which, to some extent, limited the industrial production of TENG technology [23].

Here, we demonstrated a simple, fast and low-cost method, in which the tribo-surfaces of TENG were processed without size limitation to improve its output performance. A systematical study of the grit size of sandpaper and polishing times on the influence of the output performance of TENGs was made to obtain the optimum treatment method. This method was very convenient and cheap, and most importantly, it allowed large area surface modification without size limitations. Through this method, a large-size treated TENG (20 cm × 20 cm) with micro-scale surface structures can be easily achieved and its output voltage and power can be increased by a factor of 3 and 5, respectively, providing us a

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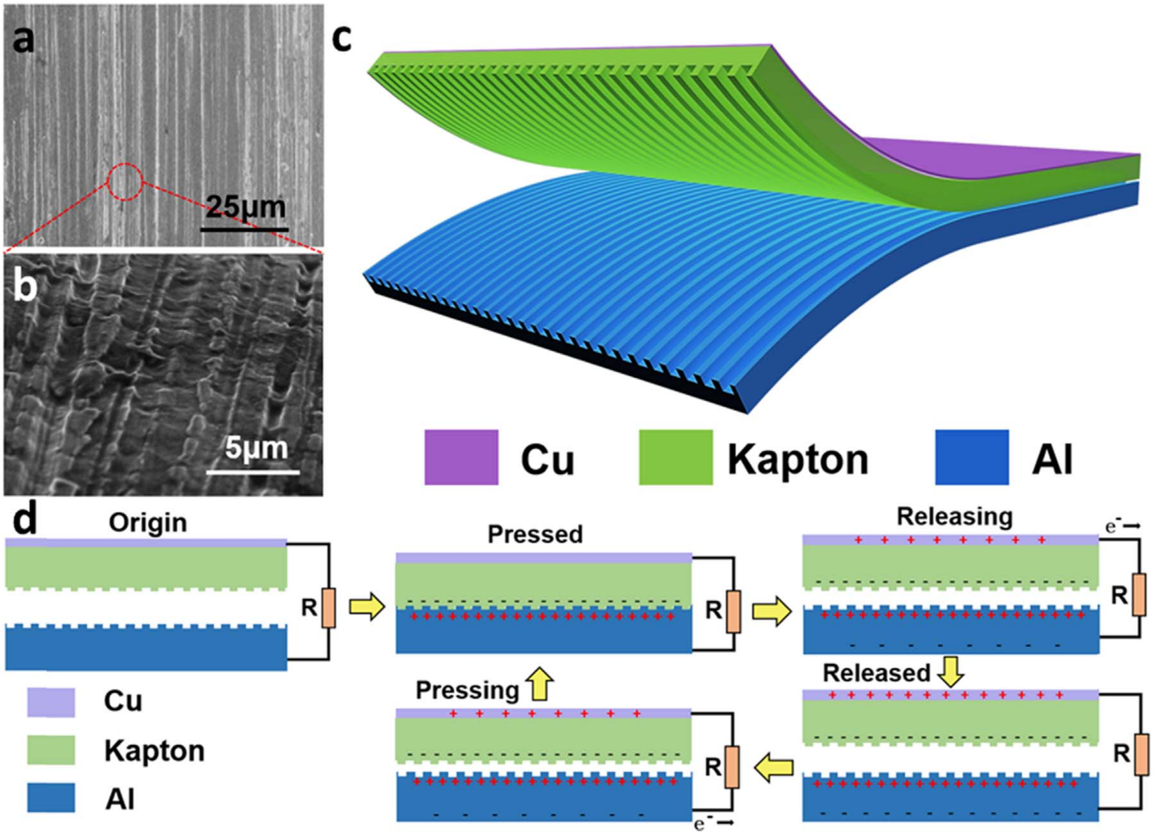

Fig. 1. (a–b) SEM images of the linear micro-structures on Al surface polished for 2 times with 3000# sandpaper. (c) Schematic of the TENG with tribo-surfaces treated by sandpaper. (d) The detailed working principle of the as-fabricated TENGs with vertical contact-separation mode.

size-unlimited, low-cost and time-saving surface micro-structure modification method for improving the output of TENG. This work showed great chance for industrial fabrication of TENG, which was crucial for sustainable power supply in the future.

2. Experimental section

2.1. Fabrication of the nanogenerator

Both of the Al foils and kapton films were prepared with the same size (2 cm × 2.5 cm × 100 μm) for standby application. The Al foils and kapton films were cleaned with alcohol. Cu film (50 nm) was deposited on the surface of kapton film by magnetron sputtering.

2.1.1. Section A

Here, three large groups of TENGs were fabricated to research the relationship between the grit size of sandpaper and the output performance of TENG. The three large groups were named untreated group, Group A and Group B, respectively. The tribo-surfaces of the four TENGs in untreated group were not processed by sandpapers. Both of the large Group A and B contain eight small groups, and each small group contains 4 TENGs. In large Group A, the Al tribo-surfaces were polished in the same direction for 2 times using sandpapers with different grit size (240#, 400#, 600#, 800#, 1000#, 1200#, 3000# and 5000#) separately. The kapton tribo-surfaces were not treated by sandpapers. In large group B, the kapton tribo-surfaces were polished in the same direction for 2 times using sandpaper with different grit size. The Al tribo-surfaces were not treated by sandpapers.

2.1.2. Section B

Three large groups of TENGs, named C, D and E, were fabricated to research the relationship between the polishing times and the output performance of TENG. All three large groups were made up of three small groups and each small group contains 4 TENGs. In large group C, the Al tribo-surfaces were polished for 1, 2 and 4 times in the same direction using sandpaper with the same grit size, respectively. The kapton tribo-surfaces were not treated by sandpapers. In large group D, the kapton tribo-surfaces were polished for different times in the same direction using sandpaper with the same grit size, while the Al tribo-surfaces were untreated by sandpapers. In large group E, the Al tribo-surfaces and the kapton tribo-surfaces were all polished for different times in the same direction using sandpaper with the same grit size.

2.2. Characterization

Morphologies of the Al foils and kapton films were observed using an optical microscope (Nikon ECLIPSE 3 × 2 STAGE JAPAN) and scanning electron microscope (SEM; SU8020), The output voltage was measured by digital phosphor oscilloscope (Tektronix DPO 3034) and the output current and the transferred charges were measured by electrometer (KEITHLEY 6517A).

3. Result and discussion

A TENG is mainly consist of two parts: the tribo-layers which are made up of materials have different electron-attracting abilities and the electrode-layers which are often conductive metal layers, such as gold, copper and aluminum. Metal layers sputtered on the back of the two tribo-surfaces as two electrodes directly

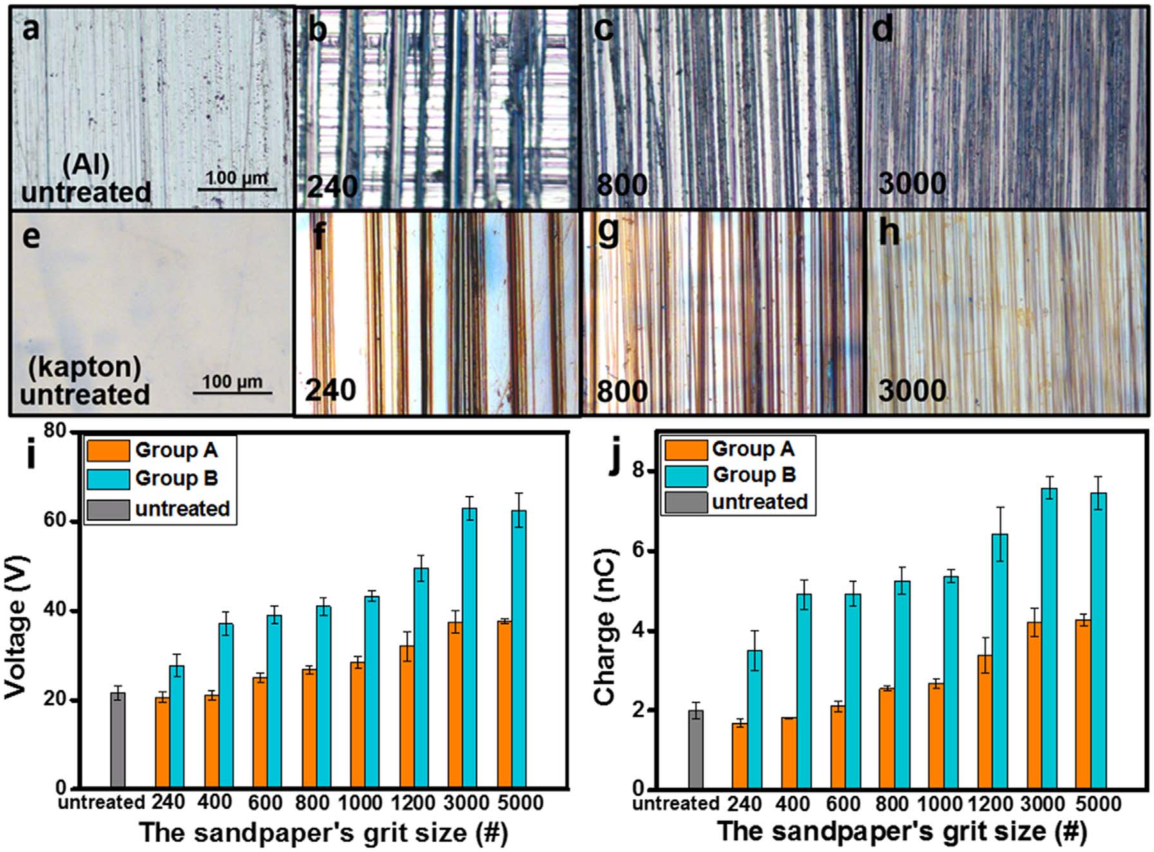

Fig. 2. Optical microscope images of (a) untreated Al foil, (b–d) Al foils treated with sandpapers of grit size 240#, 800# and 3000#, respectively. (e–h) Optical microscope images of (e) untreated kapton film surface, (f–h) kapton films surface treated with sandpapers of grit size 240#, 800# and 3000#, respectively. (i–j) The output voltage and transferred charges of the TENGs in untreated group, Group A and Group B, respectively. (Group A: only Al foils were treated. Group B: only kapton films were treated.).

connected the external circuit. In previous works, people produced nano/micro patterns on the TENG's tribo-surfaces to improve its output performance [1].

In this experiment, we fabricated the TENG mainly with Al foil and kapton film. A layer of Cu (50 nm) was deposited on the back of kapton film served as one electrode, and the Al foil was applied as both another tribo-layer and electrode. We produced linear structures on both tribo-surfaces via sandpaper treatment method (Fig. 1a). In Fig. 1a, the linear microgrooves on the surfaces of Al foil and kapton film represented the scratches produced by sandpaper. Through polishing the surface of Al foil in the same direction for 2 times with 3000# sandpaper, we can obtain obvious and regular linear scratches of 2 μm in width (Fig. 1b and c).

The detailed working principle of the as-fabricated TENGs with vertical contact-separation mode was shown in Fig. 1d. At the beginning, no charge was generated on the surfaces of both films in the original state. As an external pressure was applied on the as-fabricated TENG, the polished surfaces contacted and rubbed with each other, induced charges generated and transferred on the contact surfaces because of their different electron-attracting abilities. When the polished surfaces were separated, in order to balance the electric potential difference established by the tribo-charges on the Al foil and kapton film, the electrons in the attached induction electrodes will be driven to flow back and forth through the external circuit. So when the TENG was pressed and released periodically, it can generate alternative current pulse output [1,16].

To research the relationship of the scale of micro-structures and the output performance of TENG, we processed the tribo-surfaces of TENGs with sandpapers of different grit size. Here, to diminish the influence to the tested output, the applied sanding force was carefully controlled for it can infect the surface structure scale produced by sandpaper (Fig. S10). In our experiments, the larger the grit size of sandpaper, the smaller the diameter of the particles on it. After we processed the TENGs with sandpapers of 8 grit sizes respectively, micro-structures of different scales can be achieved on both metallic (Al) and organic (kapton) tribo-surfaces compared with their initial state (Fig. 2a–h, Supplymentary Figs. S1 and S2). The average width of the as produced linear shaped micro-structures was ranging from 2 μm to 20 μm as shown in Fig. S3, and the average density of the surface micro-structures was ranging from 45/mm to 390/mm as shown in Fig. S4. The larger the grit size of the sandpaper being used, the narrower the linear structures was produced and the higher density of surface micro-structures was obtained, which subsequently contributed to the output performance of TENG.

Two groups of TENGs were adopted for electrical tests. The group with treated Al surface was set as Group A, and the group with treated kapton surface was set as Group B. As shown in Fig. 2i and j, the output voltage as well as the transferred triboelectric charges increased as increase of the grit size. This phenomenon can be attributed to two reasons: (I) The larger the grit size of sandpaper being used, the narrower and higher density of the produced liner micro-structures, and the more complex the patterns on the tribo-surfaces of TENG. (II) The larger the grit size of sandpaper being used, the shallower the scratches, and the larger the friction area when the TENG was pressed. It has to be noted that both Group A and Group B achieved their maximum output at 3000# and maintained stable even if the grit size was still increasing. This was mainly because when the grit size of sandpaper exceed 3000#, the minimum size of the produced linear structures was about 1.5 μm, the decline was insignificant. When the grit size

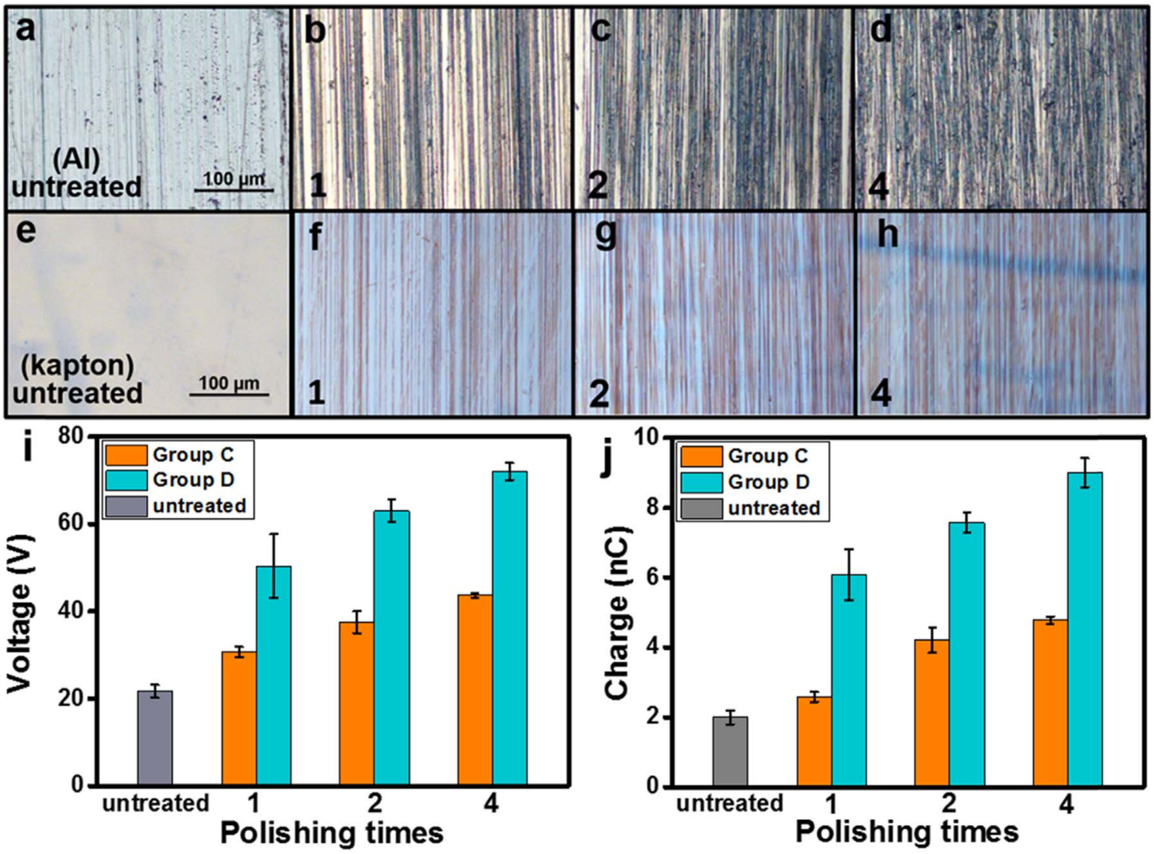

Fig. 3. The output performance of the TENG with tribo-surfaces treated with 3000# sandpaper for different times. Optical microscope images of (a) untreated Al foil surface, (b–d) Al foil surfaces polished for 1, 2 and 4 times with 3000# sandpaper, respectively. Optical microscope images of (e) untreated kapton film surface, (f–h) kapton film surfaces polished for 1, 2 and 4 times with 3000# sandpaper, respectively. (i) and (j) were the output voltage and the transferred charges of the TENGs in untreated group, Group C and Group D, respectively. (Group C: only Al foils were treated. Group D: only kapton films were treated).

of sandpaper was large enough, the grain size of the particles on its surface was too small to produce scratches with obvious linear structures. Therefore, the output performance might even decrease slightly. The highest output voltage of TENG with surface modification can reached up to 65 V, which was nearly triple of that in untreated group exhibiting a great potential of this method for improving the output performance of TENG.

As shown in Fig. 2i and j, we can also find that the output voltage and charge in Group A were generally lower than that in Group B. This phenomenon can be attributed to the reason that the Mons' hardness of Al foils' was smaller than that of the kapton films. So the Al foils can be scratched easier and deeper compared with scratching kapton films with the same sandpaper and processing strength. The produced surface linear micro-structures on kapton films in Group B were smaller, shallower and denser than that on Al foils in Group A, (Figs. S3 and S4). Therefore, when TENGs were pressed, the contact areas in Group B were generally larger than that in Group A, and the output performance in Group B were generally higher than that in Group A.

In Group A, the output performance in group 240# and 400# were slightly lower than that in untreated group (Fig. 2i and j), this phenomenon can be can be attributed to the smaller Mons' hardness scale of Al foil. The linear micro-structures produced by sandpapers with lower grit size were wide and deep, resulting in smaller contact areas compared with the untreated group.

We can conclude from i and j in Fig. 2 that when we treated the TENGs with sandpapers of 3000#, their output performance reached maximum. Hence, we used 3000# sandpapers to research the relationship between the polishing times and the output performance of TENG. After polishing different times in the same direction, micro-structures of different density were achieved on both metallic (Al) and organic (kapton) tribo-surfaces compared with their initial state (Figs. 3a–h and S5). The more the polishing times, the denser the produced linear micro-structures on tribo-surfaces. So the linear micro-structures on kapton surfaces in Group D were denser than that on Al surfaces in Group C (The group with treated Al surface was set as Group C, and the group with treated kapton surface was set as Group D).

We did grounding handling to the sandpaper treated films to effectively rule out the accumulation of triboelectric charges. The TENGs in untreated group, Group C and Group D were adopted for electrical tests. As was shown in Fig. 3i and j that the output performance of TENG can be improved by increasing the polishing times. This tendency can be attributed to the increased density of linear micro-structures on tribo-surfaces related to the polishing times. Moreover, the output performance of TENGs were not in linear with the polishing times, because the density of the linear micro-structures can reach to its saturation state when the polishing times increased to a certain level. Therefore, if the polishing times were excess to this level, the output performance of the as-fabricated TENG will not increase obviously. When the kapton surface was polished for 4 times by sandpaper of 3000#, the average width and density of the produced linear micro-structures were about 2 μm and 460/mm, respectively, which almost reached to its saturation state (see Figs. S3 and S5). So, the optimum polishing times was 4 times.

We also estimated the difference between the output performances of TENGs with double-sided pattern (Group E: The group with Al and kapton surfaces both treated) and the output performances of TENGs with single-side pattern (Group C and D). It can

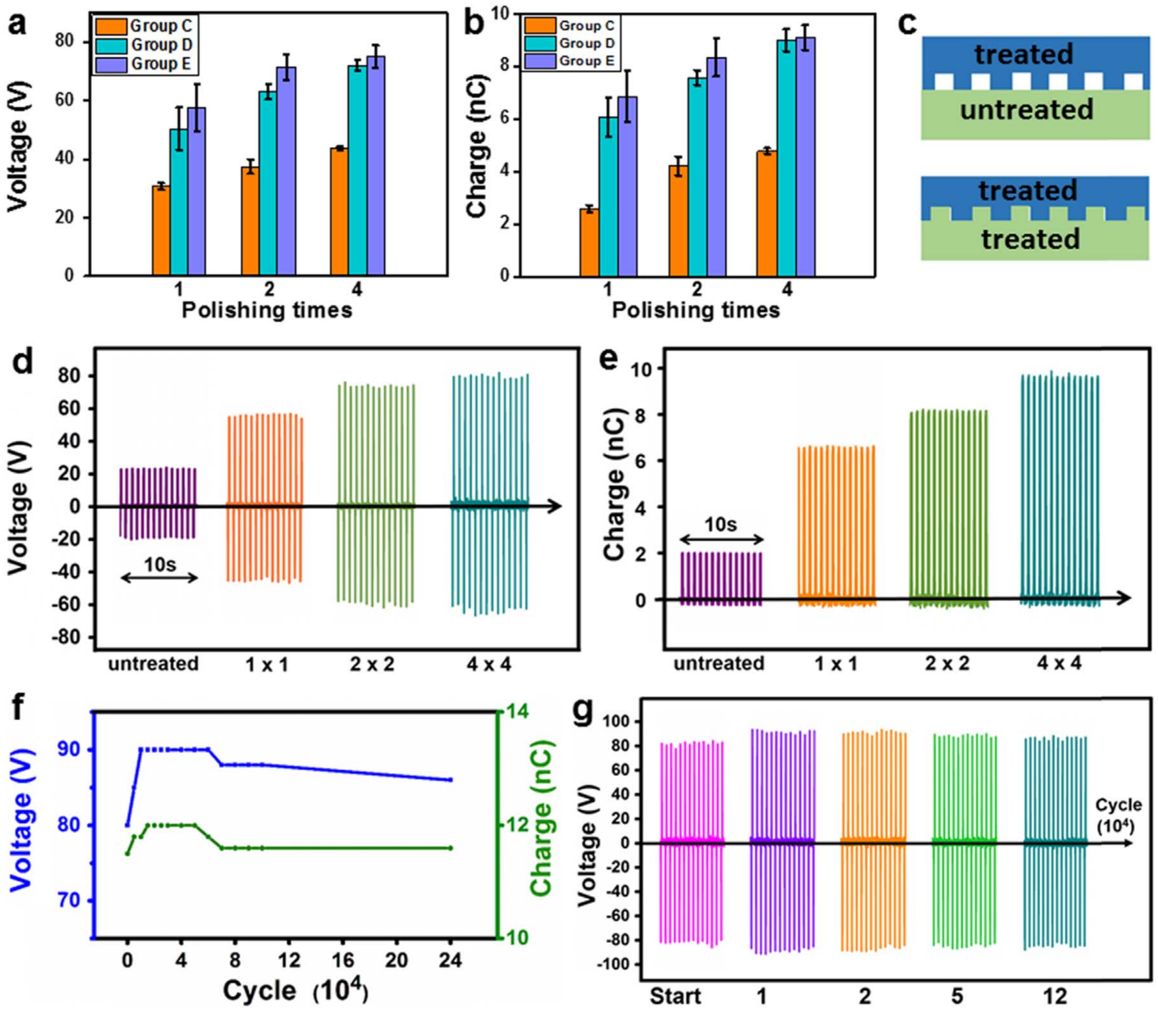

Fig. 4. (a–b) The output voltage and transferred charges of the TENGs in Group C, D and E. (Group C: only Al foils was treated. Group D: only kapton films treated. Group E: both Al and kapton films were treated). (c) The diagram above shows the interface of TENGs in Group C and D, the diagram below shows the partial interface of TENGs in Group E; (d–e) The open-circuit voltage and the transferred charge waveforms of the TENGs in Group E (1 × 1; 2 × 2; 4 × 4 denote Al foils and kapton films were all polished by 3000# sandpapers for 1 time; 2 times; 4 times, respectively); (f–g) Stability tests of TENG (4 × 4).

be seen in Fig. 4a and b that the output performances of TENGs in Group E were higher than those in Group C and D consisting with previous reports that the TENGs with nano/micro structures on both tribo-surfaces exhibited higher output [24]. In Group E, the Al foils and kapton films constituted TENGs were all treated by sandpapers, and there were micro linear structures on both tribo-surfaces. When these TENGs were pressed, the sufficient contact area in Group E was larger than that in Group C and D, because the linear structures on both surfaces were partly coincided with each other, which was not existed in the TENG with single treated tribo-surface (Fig. 4c).

The open-circuit voltage and transferred charges of the TENGs in untreated group and Group E were maintained regular and stable (Fig. 4d and e). It was worth noting that the maximum output voltage can reach to 80 V, which was almost 3.5 times as high as that in the untreated group. The output performance of treated TENG was also comparable with that treated by ICP method which was widely used in today's TENG treatment (Fig. S8). Moreover, through our test, the sandpaper treated TENG showed outstanding stability. It can maintain good working performance after being tested for 1.2×10^5 cycles. According to the above results, we can optimized the sandpaper processing parameters, which is to polish both tribo-surfaces of the TENG with sandpaper of 3000# for 4 times in one direction.

Compare to other micro/nano structure processing methods, sandpaper treatment method have advantages in treating large-area tribo-surface even without size limitation (Fig. S6). An examination was designed to demonstrate the feasibility for size-unlimited surface treatment. As shown in Fig. 5a, triboelectric films (Al and kapton) with large area (20 cm × 20 cm) can be easily processed with the sandpaper treatment method mentioned above and then used to fabricate a large-sized TENG. Another TENG with the same size and untreated tribo-surface were also fabricated used as a control experiment group. Besides, when applied to different materials that former used as friction layers, such as PET and PTFE, this method also achieved very good results, showing its great versatility (Fig. S11).

The treated large-sized TENG can light up 350 LEDs at the same time (Fig. 5b, Video S1). The output voltage and current of the optimum method treated large-sized TENG were about 1500 V and 75 μA, respectively, which were about 3 times of the untreated one (500 V and 26 μA) (Figs. 5c and S7). The output power of both TENGs at different load resistance were also measured (Fig. 5d). When the external load was set to about 13 MΩ, the output voltage and current of the treated TENG were about 560 V and 44.5 μA, respectively, and the output power density can reach to its maximum value (62.3 μW/cm^2). (Fig. S8a). While the output voltage and current of the untreated TENG were about 280 V and

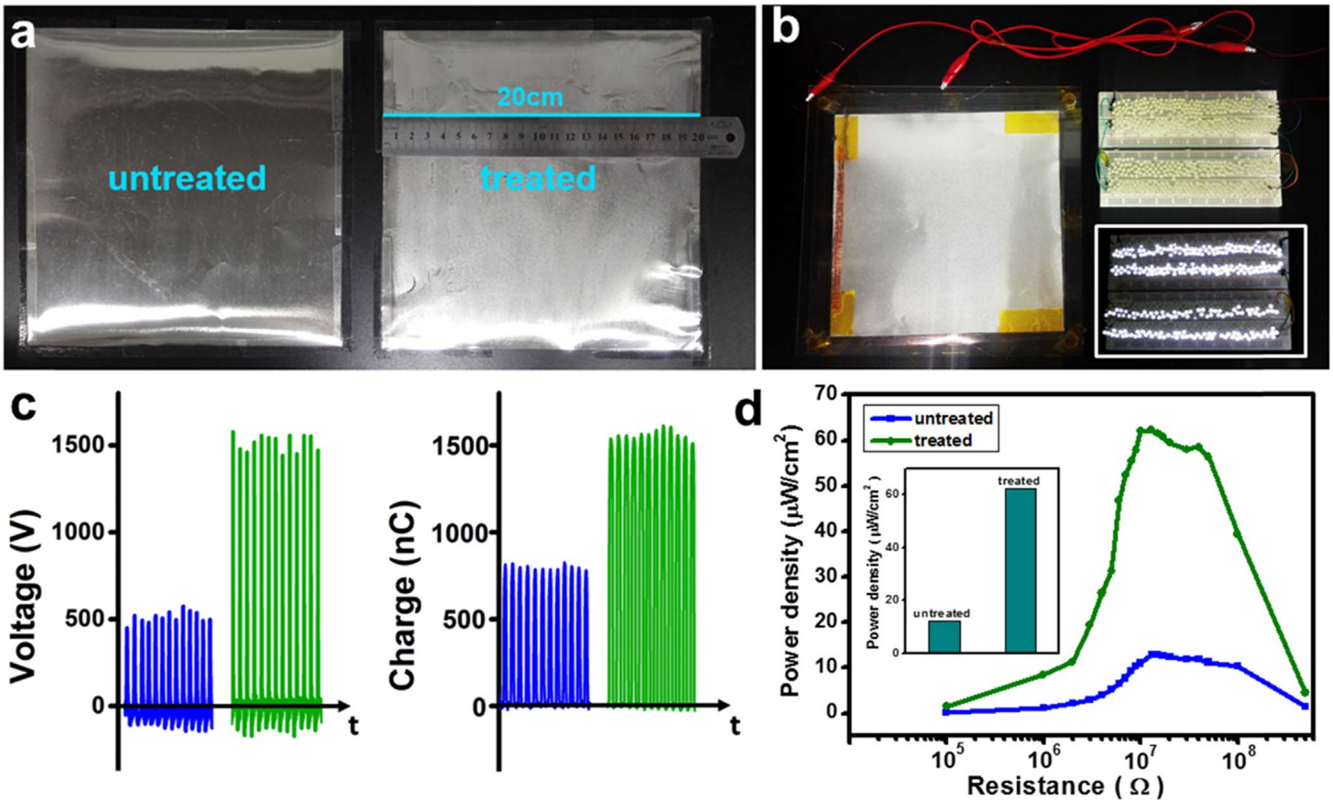

Fig. 5. (a) The image of the Al foils (20 cm × 20 cm) untreated and treated by sandpaper. (b) The image of the large-sized TENG (20 cm × 20 cm) processed by optimum method lighting up 350 LEDs at the same time. (c) The output performance of the untreated large-sized TENG (blue) and the large-sized TENG processed by the optimum method (green). (d) The output power density curve of both large-size TENGs at different load resistance, respectively; the inset in this picture is the contrast power of both large-sized TENGs.

18 μA, respectively, and the maximum output power were only 13 μW/cm^2 at the external load resistance of 13 MΩ. (Fig. S8b). If the untreated large-sized TENG was treated by the optimum sandpaper treatment method, its output power can be increased by about 5 times (see inset of Fig. 5d).

Supplementary material related to this article can be found online at http://dx.doi.org/10.1016/j.nanoen.2016.08.024.

4. Conclusion

In summary, we have demonstrated a sandpaper treatment method to modify the tribo-surface of TENG and improve the output performance effectively. The greater the grit size of sandpaper being used and the larger the polishing times, the higher the output. The optimum sandpaper treatment method is to polish both tribo-surfaces of the TENG with 3000# sandpaper for 4 times in the same direction. After treating the TENG with this optimum method, the output voltage and power of TENG can be increased by a factor of 3 and 5, respectively. This method provide us a simple, fast and low-cost way to create micro-structure on tribo-surface of TENG even without size limitations for improving the output of TENG. This work exhibited great potential for industrial fabrication of TENG in the future.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.nanoen.2016.08.024.

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