Exploring theoretical and experimental optimization towards high-performance triboelectric nanogenerators using microarchitecture silk cocoon films

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Abstract

Triboelectric nanogenerators (TENGs) developed using eco-friendly natural materials instead of traditional electronic materials are more favorable for biocompatible applications as well as from a sustainable life-cycle analysis perspective. Microarchitected silkworm fibroin films with high surface roughness and an outstanding ability to lose electrons are used to design TENGs. An alcohol-annealing treatment is utilized to strengthen the resistance of the silk film against humidity and aqueous solubility. Herein, for the first time, the distance-dependent electric field (DDEF) theoretical model is employed to optimize the TENG parameters to achieve high-output, which shows excellent agreement with the experimental outputs of silk film-based TENG (SF-TENG). The alcohol-treated microarchitected SF (AT-MASF) with a polytetrafluoroethylene (PTFE) positive contact exhibit a stable and high electrical output even in harsh environments. These studies can lead us closer to the attractive future vision of realizing the biodegradable TENG systems for harness/sensing various biomechanical activities even under real/humid environments. The potential and real-time application of the proposed AT-MASF-

TENG is demonstrated by directly employing its electric power to drive a number of low-power portable electronics and for sensing in human-body centric activities.

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1. Introduction

Clean energy via renewables for human society has become one of the most important and demanding concerns faced by humanity. Despite many forms of energy in our surrounding environment such as solar, wind, thermal, chemical, and mechanical energies, etc.,^[1] each has its own advantages and disadvantages to society.^[2] However, among the above energy resources, mechanical energy can be considered as an abundant energy source available through movements in everyday human life. In this regard, researchers have been investigating methods to harvest mechanical energy to support the needs for human life.^[3-7] Triboelectric nanogenerators have been a great invention for mechanical energy harvesting, reported first by Wang et al. in 2012.^[8,9] Since

then, many studies have been carried out to improve their electrical performance, sustainability, quality, flexibility, etc.^[10-19] Theoretical models such as the capacitor-based model was developed to approximate the output trends of TENGs,^[20-23] however, more recently, improved theoretical platforms such as the distance-dependent electric field (DDEF) model have been presented to fully describe the performance and to optimize the device structure to achieve a high output in TENG.^[24-27] In the DDEF model, a detailed analysis of the electric fields generated by each of the TENG surfaces, and the effect of material, structure, and motion properties of the device is used for optimization resulting in relatively high accuracy, when compared to the simpler parallel plate capacitor model.^[24,25] Furthermore, the DDEF model has been demonstrated to show a high degree of adaptability covering a wide variety of device structures and working modes, while producing analytical and numerical simulations closely matching with the experimental outputs.^[24-27] In comparison to highly specialized finite element models, the DDEF model has shown more versatility especially when simulating dynamic device conditions.^[24,25]

Despite the theoretical and application-level device improvements in TENG research, there is a necessity to develop nanogenerators in eco-friendly materials and these compatible with human life. Being aware of this, fabrication with natural materials can be more favorable for biocompatibility, biodegradability, implantability, and non-toxicity as well as eco-friendliness to the environment.^[5] Silk, produced by cultivation of silkworms, is a well-known naturally abundant and eco-friendly biomaterial used in the textile industry because of its luster and mechanically high strength large-scale production at a viable price.^[28-30] In particular, silk fibroin as a protein polymer can be processed into a variety of regenerated forms such as solutions, porous scaffolds, nano/microspheres, and films for various research fields including optical, electrical, and biomedical applications.^[31-37] Usually, silk can also be produced from different anthropods such

as silkworms, spiders, scorpions, mites, and bees. Silk is spun into fibers during their metamorphosis to form cocoons. The primary advantages of silk compared to other natural polymers are its excellent mechanical properties and the presence of easily accessible chemical groups for functional modifications.^[34,35,37,38] Moreover, there are various source organisms for the silk polymeric fibers, such as *Bombyx mori*, *Antheraea mylitta*, *Philosamia cynthia ricini*, *Coscinocera hercules*, *Hyalophora euryalus*, *Rothschildia hesperis*, *Eupackardia calleta*, *Rothschildia lebeau*, *Antheraea oclea*, *Hyalophora gloveri*, and *Copaxa multifenestrata*. Among all these source organisms, the *Bombyx mori* exhibits the highest tensile strength and tensile modulus of 4.3-5.2 g/den and 84-121 g/den, respectively.^[38] It also shows excellent mechanical properties, which leads to good durability and use under external stress.^[32,36] In addition, silk fibroin film can be applied to flexible systems as a substrate or an active layer.

In addition, silk fibroin can also exhibit a strong electron-donating tendency, which can be utilized as a promising positive triboelectric material, allowing it to be used in contact with the human skin and body (or even implantable).^[20,38] Due to these advantageous features, silk fibroin films were utilized to fabricate TENGs, in which the silk films or electrospun silk fibers are used as triboelectric materials and supporting substrates of the transparent and biodegradable TENGs.^[34,39-45] However, the effect of several parameters like the selection of triboelectric materials, surface roughness, and compression frequency, etc., on the output characteristics of such silk film-based TENGs are not extensively studied in previous reports. For example, the quantity of charge separation in TENGs is more likely to be proportional to the surface area in contact. Thus, the surface area of triboelectric layers plays a crucial role in the charge separation. Here we adopt the DDEF theoretical model to fully understand the working mechanism and to examine the effects of such parameters on the output performance of silk-based TENG. Furthermore, these

theoretical results are compared with an experimental analysis and optimized device structures to obtain a high output performance. Experimentally, the surface roughness of the silk film was improved by microarchitecture pattern-transfer onto its surface. In addition, the patterned silk films were post-annealed with alcohol to strengthen the resistance of silk to humidity and liquid phase.^[32,40] Another objective is to employ a bio-degradable, long-lasting, and humid resistant silk film to harness mechanical energies even in a harsh environment in contact with the human body. So, we systematically investigate the water solubility of silk before and after the alcohol-annealing treatment, examining the performance of corresponding silk-film based TENGs. Thus far, the electrical response of the silk film-based TENGs in humid environments has been rarely studied. Except AT-MASF, the remained parts of the proposed TENG device are non-biocompatible materials. It can lead to future realizing of complete biodegradable TENG systems for real-time applications like sensing biomechanical activities even under harsh environments. Furthermore, the effective power density and robustness of TENG devices with the alcohol-treated MASF are also examined. Ultimately, the theoretically and experimentally analyzed optimum silk-based TENGs are employed for real-world and real-time applications like powering low-power electronics and sensing, etc.

2. Results and Discussion

Figure 1 depicts the schematic diagrams for the fabrication process of (a) silk fibroin solution and (b) microarchitecture silk film (MASF) and the design of (c) alcohol-treated microarchitecture silk film-based TENG device (MASF-TENG). Silk fibroin solution was prepared using previously reported protocols,^[33] which is summarized as follows (Figure 1a). As shown in Figure 1a,

commercially available silk cocoons were cut into pieces and treated with sodium bicarbonate (NaHCO_3) solution by a boiling process for the removal of sericin in silk fibers, followed by a rinse with de-ionized (DI) water. Dried silk fibroins were completely dissolved in a lithium bromide (LiBr) solution and the silk fibroin solution was filtered through a Mira-cloth, subsequently dialyzed to remove the salt (i.e., LiBr). To fabricate the MASF, the purified silk solution was coated on the micro-grooved polydimethylsiloxane (MG-PDMS) mold, followed by casting it at room temperature (Figure 1b). Herein, the MG-PDMS mold was fabricated using micro-pyramidal architected silicon (Si) substrates, which are prepared using a hydrogen chloride chemical etching method *via* soft imprinting lithography (SIL) technique and the detailed information can be found in our previous work.^[46] The as-developed MASF is further treated with alcohol (i.e., methanol) by spray coating followed by the annealing treatment, as shown in Figure S1 (In the Supporting Information). Such post-alcohol treatment can be expected to enhance the crystallization of silk,^[39,47,48] which reduces its water solubility. The alcohol treatment of silk film is discussed in detail, in the next section. Finally, the alcohol-treated MASF (AT-MASF) and polytetrafluoroethylene (PTFE) are employed as positive and negative triboelectric layers in a TENG device, respectively, as shown in Figure 1c. Detailed fabrication procedures of the silk solution, MASF, and MASF-TENG devices are summarized in the experimental section.

2.1. Structural characterization of silk film:

The structural characteristics and degradability (i.e., water solubility) of silk fibroin films are examined. Fourier-transform infrared (FTIR) spectroscopy and X-ray diffraction (XRD) analyses are performed to investigate the functional groups and crystallinity of silk films. Figure 2a shows the XRD pattern of silk film before and after the alcohol treatment. The diffraction patterns of the silk film and alcohol-treated silk film showed at $2\theta = 11.66, 19.59, 22.24, 25.64$ and 40.82° ,

suggesting that the obtained silk film formed in silk I structure.^[49,50] Although, even after the alcohol treatment the diffractions peak positions of silk films did not significantly vary except for the little variations in their intensities. This might be ascribed either to the fabrication processes or the thickness of silk film, which is in the micrometer range. Actually, we have prepared the SF by the simple casting technique, which is formed by liquid-based silk to the silk film at slow crystallization in the ambient condition. Due to these reason, eventhough the diffraction peak intensities varied, we do not except significantly change in the XRD patterns of the alcohol treated silk film. The changes in the structure of silk film before and after the alcohol-treatment are further characterized by FTIR spectroscopy, as shown in Figure 2b. Typically, treating the silk film with alcohol can increase the β -sheet crystalline domains.^[51,52] Herein, the silk film treated with ethanol assembles the hydrophobic blocks to form micelles, eventually resulting in regular and stable β -sheet crystallites. As shown in Figure 2b, the basic absorption peak at around 3400 cm^{-1} corresponds to the O-H stretching vibration. Further, the untreated film showed characteristic absorption bands at 1637 , 1512 and 1229 cm^{-1} for the amide I, II and III groups, representing the typical random coil protein conformation of the silk I structure.^[53] Furthermore, after the ethanol treatment, these bands shifted to 1618 , 1510 , and 1226 cm^{-1} , respectively. This shift in band determines that the alcohol-treated silk film is converted to the β -sheet formation of the silk II structure,^[54] due to the hydrogen bonds rearrangement activated by low dielectric organic solvents.^[55,56]

Silk fibroin film is well known as an easily soluble material in water or with high humid environments.^[32,33,35,57] Thus, it is meaningful to study the effect of alcohol treatment on water solubility of silk fibroin films. Figure 2c exhibits the water solubility of the as-prepared silk fibroin film without any alcohol post-annealing treatment. After immersing the silk film into DI water, it

is completely dissolved within 2 min. Here, a small amount of dye was loaded into silk film for easy discrimination. Although the silk film with several advantageous features such as biocompatibility, high transparency, and flexibility, can be widely used for a variety of applications including wearable and implantable devices, the silk-based TENGs cannot be long-lasting on the human skin and the body in humid environments.^[34,35,37,40] To prevent the device performance degradation of silk-based TENGs in humid conditions, the water-solubility of the silk film needs to be programmed by the post-alcohol annealing process.^[32] Methanol (99%) was sprayed on the surface of silk films 2 times with an interval of 5 min, followed by dry at 30 °C in the oven. Figure 2d shows the sequential photographic images of the post-alcohol treated silk film immersed in the DI water for 30 min, 5 h, 10 h, and 24 h. These images are clearly demonstrating that the post-treated silk film is not completely dissolved even after 24 h, unlike the untreated silk film. To further affirm the water-insoluble nature of alcohol-treated silk films, it is soaked in water for a week (Figure S2). As shown in Figure S2, the alcohol-treated silk film is insoluble in water even after a week and these results can evidently support its water-insoluble nature. Moreover, they also confirm that even though the silk film is bent or partially shrunk after soaking it for a week, it is still flexible and durable. These studies clearly suggest that water-insoluble silk films can be realized by treating with methanol. This might be ascribed to the transformation of the random coil molecular structure of silk film into β -sheet formation during the methanol treatment.^[40,58] Therefore, the TENG developed with the alcohol-treated silk film can be utilized as various potential wearable and implantable energy sources.

2.2. Working mechanism of silk film-based TENG (SF-TENG) device

According to the triboelectric series, silk and PTFE are classified as positive and negative triboelectric materials respectively,^[59] which are attached individually onto an Al tape and utilized

as bottom tribo-positive and top tribo-negative layers, as stated in the preceding section. The working mechanism of the SF-TENG device is illustrated in Figure 2e. When an external compressive force is applied on the TENG (Figure 2e-i), the top triboelectric material (i.e., PTFE) can experience a force and displace vertically towards the bottom material (i.e., silk). Consequently, both the materials contact with each other and generate the opposite triboelectric charges on their surfaces owing to the triboelectrification (or contact electrification). Thus, the equal number of positive and negative charges accumulated across the surfaces of MASF and PTFE can result in zero electrostatic potential differences between two layers (Figure 2e-i). When the vertically influenced force is slowly released from the MASF-TENG, the PTFE contemplates beginning returns to its actual position due to the flexibility of the attached springs. As a result, the separation in the triboelectric materials can generate an electrostatic potential difference between them and result in the electron flow from the top to bottom Al electrodes (Figure 2e-ii). Once the influencing force is entirely removed from the MASF-TENG, both the triboelectric materials are completely separated and reach their initial positions, which is in the equilibrium state of triboelectric charge distribution (Figure 2e-iii). And then, when the external compressive force is again applied to MASF-TENG, both the triboelectric materials begin to move towards contacting direction each other, which causes a reverse electrostatic potential difference between them due to the influence of applied force. Consequently, the electron flow from the bottom to top Al electrodes can be observed (Figure 2e-iv). Continuously applied and relaxed compressive forces on the MASF-TENG can repeatedly generate an electrostatic potential difference between two Al electrodes, thus resulting in the charge flow as well as an electrical output current through an external circuit.

2.3. Theoretical and experimental studies to examine the effect of surface roughness of triboelectric materials on the output performance of TENG

2.3.1 Surface morphology characterization of silk films: Typically, the surface roughness of triboelectric materials plays a key role in enhancing the output performance of TENGs. Thus, to examine the effect of surface roughness on the performance of the TENG device, microarchitectures (MAs) were developed on the surface of silk film by the SIL technique and the electrical output of the corresponding device investigated. Figure S3 shows the field-emission scanning electron microscopic (FE-SEM) images of (a-i) flat and (b-i) MA silk films. The surface morphological difference between the flat and MA silk films clearly indicates that the MGs on the surface of PDMS is well and inversely replicated on the surface of the silk film (Figure S3). The size of MAs on MASF was estimated in the range of 0.5-4 μm , whereas none of any architectures was observed on the surface of flat SF. As discussed above, to reduce the water solubility of silk film, its surface was treated with methanol. Figure S3 shows the FE-SEM images of the (a-ii) flat and (b-ii) MA silk films after the methanol treatment. Owing to the methanol spray-coated on the surface of flat and MA silk films, many small globular fractures appeared on their surfaces.^[57] As a result, the surface morphology of both the flat and MA silk films appears to be modified by the alcohol annealing treatment. However, the surface roughness (R_q) of these flat and MA silk films before and after the alcohol treatment was examined by the atomic force microscopy (AFM), as shown in **Figure 3a-b**. The AFM images of flat SFs (Figure 3a) reveal that after the alcohol treatment, small holes and globular nano fractures are formed on its surface. Resultantly, the R_q of alcohol-treated flat SF (AT-flat SF) is noticed to be higher (i.e., ~32.53 nm) than that of the untreated flat SF (i.e., ~8.36 nm). From the AFM images, the surface area (SA) is also estimated and it's also slightly increased from 900.2 to 900.9 μm^2 by treating the flat SF with alcohol. Whereas, both the R_q and SA values of MASF are predominantly increased to 0.53 μm and 974.8 μm^2 , as compared to the untreated and alcohol-treated flat SF. On the contrary, both the R_q and

SA values of MASF after the alcohol treatment are decreased to $0.49\ \mu\text{m}$ and $963.8\ \mu\text{m}^2$, respectively. This may be attributed to the slight damage of MA shapes, in the presence of the spray-coated alcohol. So, even though the globular nano fractures are formed on the surface of AT-MASF, slight damage of its MA can influence the surface roughness of the corresponding film (see Figure S4). Therefore, the surface roughness of AT-MASF is slightly reduced (i.e., only about $40\ \text{nm}$), as compared to the untreated MASF. However, the effect of such surface roughness variations of the silk films on the output performance of corresponding TENG devices is examined through theoretical and experimental perspectives.

2.3.2 COMSOL Multiphysics simulations: Firstly, the influence of electrical potential distribution across the active materials of TENG by altering their surface roughness was theoretically studied using COMSOL Multiphysics; finite elemental simulation, as shown in Figure 3c. To design the simulation model, the SF, and PTFE with the width of $500\ \mu\text{m}$ are placed on the two Al electrodes with the same width, while the thickness of all the materials is assumed to be $100\ \mu\text{m}$. The Al/PTFE and SF/Al stacks are used as a top and bottom component of TENG, and the distance between them is fixed to $400\ \mu\text{m}$. While performing the simulations, the surface charge density across the active materials (i.e., silk film and PTFE) of TENG is assumed as $\pm 90\ \mu\text{C}/\text{m}^2$. The theoretical simulation results are shown in Figure 3c clarify that the electrical potential distribution across the MASF-TENG is significantly higher as compared to the TENG with flat SF. Such variation in the potential distribution is mainly attributed due to the high surface roughness of MASF, which leads to the enhanced contact area between the active materials as well as the output performance of TENG.

2.3.3 Distance-dependent electric field (DDEF) model: To further affirm the above discussion, the variations in the electrical outputs such as voltage, charge and current densities are also calculated

by the unified DDEF simulation model. During these simulations, length and width of triboelectric layers (L , W), triboelectric layer thickness (x_1 , x_2), separation between the TENG layers ($z(t)$), permittivity (ϵ), and triboelectric charge density (σ_T) were considered as the main parameters, and the electric field of the TENG layers were represented using:

$$E_z = \frac{\sigma_T}{\pi\epsilon} \arctan\left(\frac{L/W}{2(z/W)\sqrt{4(z/W)^2 + (L/W)^2 + 1}}\right) = \frac{\sigma}{\pi\epsilon_0} f(z) \quad (1)$$

Where the effect of x_1 , x_2 , and $z(t)$ are represented by the separation between TENG layers (z) in equation (1).^[24,25] Using equation (1), the voltage, current and charge outputs of the TENG were calculated using the DDEF model throughout this work (considering the structure depicted in Figure 3d), as indicated Supplementary Note-I. Considering experimental conditions, device parameters corresponding to each of the experiments were used for the simulations, as summarized in Table S1. The triboelectric charge density (Figure S5) was estimated using the short circuit output charge characteristics for each experiment, similar to our previous work.^[24,25,27] For the simulations related to the silk layers, the increase in the surface area was considered to be proportional to the increasing surface charge density, assuming a relatively even distribution of triboelectric charges on the triboelectric contact surfaces. The estimated peak charge density and current density (J_{SC}) values of TENG with various silk films from the distance-dependent model are shown in Figure 3e, respectively. The corresponding calculated peak open-circuit voltage (V_{OC}) values are also shown in Figure S6. The charge density, J_{SC} and V_{OC} values of TENG with an untreated and alcohol-treated MASF are noticed to be very high than that of the untreated and alcohol-treated flat SF (Figure 3e and S6). Furthermore, the output performance of TENG with the corresponding silk films are experimentally investigated.

2.3.4 Measured electrical outputs of TENG with different silk films: The measured charge density, J_{SC} , and V_{OC} curves of the TENG devices with and without alcohol-treated flat SF and MASF are shown in **Figure 4a-c**. These measured charge density, J_{SC} and V_{OC} provide a close approximation of the trend to the distant-dependent model calculations (Figure 3e and S6). Moreover, the magnitude of both the measured charge density and J_{SC} values is almost matched with the DDEF model calculations. However, it should be noted that there is a mismatch between the magnitude of the V_{OC} predictions and the experimental scenario, which is widely observed for all of the finite element models as well as analytical theoretical models. This is known to be due to the high impedance nature of the output generation of TENG impeding the direct experimental measurement of V_{OC} .^[20-25,27,60,61] However, both the theoretical simulation (Figure 3c and 3e) and experimental results (Figure 4a-c) are clearly elucidating that, as compared to other SFs, the untreated and alcohol-treated MASFs with very high surface roughness are preferred to achieve high output performance of TENG. Since, the triboelectric materials with high surface roughness are more favorable to improve the contact area between the active materials, which can lead to enhanced surface charge density as well as the output performance of TENG. Although, the output performance of TENG with an alcohol-treated MASF is slightly decreased than that of the TENG with an untreated TENG. Such a slight decrement might be attributed to a small variation in the surface roughness of alcohol-treated and untreated MASFs (Figure 3b), which can be expected to slightly influence the surface contact area as well as the output performance of TENG. However, the silk film based TENG with high-performance as well as strong robustness even in the harsh environment is the main target of this work. Therefore, the AT-MASF based TENG (AT-MASF-TENG) with a peak output charge density, J_{SC} , and V_{OC} values of approximately $97 \mu C/m^2$, 65

mA/m^2 and 350 V, respectively, are considered as the device with optimum performance, and, employed for further investigations.

2.4 Theoretical and experimental analysis to identify an optimum triboelectric material for high-performance AT-MASF-TENG

Along with the surface roughness of triboelectric materials, the choice of the best pair of triboelectric materials is also an important factor for high-performance TENG devices. So, the effect of the electrical output performance of the AT-MASF-TENG as a function of different negative triboelectric materials is firstly examined by the DDEF model. A series of triboelectric materials with different charge separation properties and dielectric constants were used for the simulations. All the negative material types used in this step were identical in dimensions, and the outputs of the different TENG material types were simulated using the method discussed in section 2.3.3. Figure 4d indicates the DDEF model simulations for the electric field (positive) originating due to triboelectric surface charges of the AT-MASF sheet, plotted against distance from its surface. Considering the triboelectric charging behavior between the AT-MASF sheet and respective negative triboelectric materials, the triboelectric charge density changes. According to Figure 4d, the maximum electric field is observed when AT-MASF and PTFE are used as the triboelectric pair, indicating the highest charge separation for this case. This is followed by PDMS, kapton, copper in the descending order of charging, whereas paper provides minimum charging. This is consistent with the simulation outcomes for the corresponding negative triboelectric materials (Figure S7), where PTFE displays the largest negative electric field and paper the smallest negative electric field. This shows that the PTFE acts as the most suitable triboelectric surface against the AT-MASF, among the materials examined in this work. The DDEF simulation output charge density and J_{SC} of TENG, while the AT-MASF making continuous

contact/separations with other distinct triboelectric materials including paper, copper, kapton, PDMS, and PTFE, etc., are shown in Figure 4e and 4f. These simulation results are clearly indicating that the performance of AT-MAST-TENG is altering as a function of the triboelectric tendency of the counter surface material. These output trends are consistent with the electric field simulations, shown in Figure 4d, where the AT-MASF contacted with PTFE provides the highest performance, whereas the contact between AT-MASF and paper provides the lowest performance. Furthermore, the electrical output performance of TENG is also experimentally investigated with the different types of triboelectric counter materials opposite to AT-MASF. The corresponding measured charge density, J_{SC} and V_{OC} curves of AT-MASF-TENGs are shown in Figure 4g-h. These measured electrical output curves of TENG are also in good agreement with the DDEF simulations for the electric field (Figure 4d) as well as the output simulations (Figure 4e, f). Both the theoretical and experimental analysis results are clearly elucidating that the pair of AT-MASF and PTFE are optimal or selective to attain the superior electrical output performance, compared to other pairs (i.e., AT-MASF/paper, AT-MASF/copper, AT-MASF/kapton, and AT-MASF/PDMS). Such variations in the electrical output of TENGs are mainly attributed to the ability of triboelectric materials to lose/gain the triboelectric charges.

2.5. Theoretical and experimental analysis to examine the influence of compression frequencies on the output performance of AT-MASF-TENG

In addition to the selection of triboelectric materials and its surface roughness, the output performance of TENGs is also strongly dependent on the input (or applied) compression frequency and force. Thus, the effect of external compression frequencies on the output performance of AT-MASF-TENGs is theoretically (**Figure 5a** and **b**) and experimentally (Figure 5c-e) analyzed. During these measurements, the compression frequencies applied on AT-MASF-TENG are varied

in the range of 1-5 Hz, while the compression force is kept constant at 3 N. As reported previously,^[62,63] the peak J_{SC} values of AT-MASF-TENG are gradually increased from 28 to 65 mA/m² by increasing the frequency from 1 to 5 Hz, while the charge density and V_{OC} values remain almost constant. The DDEF theoretical output results have also followed the same trend. In this scenario the maximum amplitude of the movement does not change during different frequencies, therefore, the TENG surfaces follow a relatively similar path from contact to maximum separation. As a result, the maximum voltage and maximum charge outputs, which depends on the position of the TENG surfaces, do not change considerably. However, as described in our previous work,^[24,25,27] the increase of frequency of the movement increases the rate of charge exchange, resulting in higher current outputs. As a result, the electrical output current of AT-MASF-TENG increases by increasing the compression frequency.

Furthermore, the influence of an external compression force on the electrical output performance of AT-MASF-TENG was also studied, as shown in Figure 5f-h. These studies are clearly indicating that the output charge density, J_{SC} , and V_{OC} values of AT-MASF-TENG are strongly dependent on the applied compression force. The peak output charge density, J_{SC} , and V_{OC} values of AT-MASF-TENG device are gradually increased from approximately 76-102 $\mu\text{C}/\text{m}^2$, 43-106 mA/m², and 254-530 V, respectively, by increasing the compression force from 1-10 N. This trend on the output performance of the AT-MASF-TENG is mainly ascribed to the increase of the surface contact area between the AT-MASF and PTFE layers in the presence of a strong applied compression force.^[64,65]

2.6. Tests for an effective output power density and robustness of MASF-TENG

For practical applications of TENG devices, it's an optimal/effective output power density value is very crucial to be known. So, an optimal/effective output power density of flat SF- and AT-

MASF-TENG devices were explored by measuring its electrical output performance at different external resistors loaded in the circuit. The peak output voltage/current values of flat SF- and AT-MASF-TENG devices with distinct load resistances are displayed in **Figures 6a** and **b**. For both the devices, as the load resistance increases, the voltage is raised, whereas the current density is reduced. Using both the voltage and current density values, the load resistance-dependent power density values of flat SF- and AT-MASF-TENG device was obtained (Figure 6c).^[7,65-67] As well known, the effective (or maximum) output power of TENG strongly depends on the short-circuit and open-circuit conditions. As expected, at very low or high load resistances, both the flat SF- and AT-MASF-TENG devices exhibited lower power density values. On the other hand, the effective output power density value of 22.05 W/m² was observed by the AT-MASF-TENG at the load resistance of 20 M Ω , which is higher than that of the flat SF-TENG (i.e., 15.13 W/m² at 40 M Ω). Therefore, our proposed AT-MASF-TENG device exhibited a peak charge density, J_{SC} , V_{OC} and power density values of 101 $\mu\text{C}/\text{m}^2$, 89 mA/m^2 , 395 V and 22.05 W/m², under the compression force and frequency of 5 N and 5 Hz, respectively, which are similar or even better as compared to the previously reported silk-film based nanogenerators, as shown in Table S2.^[39,41,68,69] Furthermore, the durability test of the AT-MASF-TENG device was also carried out to examine its reliability and electrical stability. In this respect, the electrical output voltage of the AT-MASF-TENG device was measured by continuously applying an external compression force of 5 N, for approximately 10,000 cycles (for 33 min) at the frequency of 5 Hz, as shown in Figure 6d. There is no significant change in an average output voltage value of AT-MASF-TENG between initial 3 min and final 33 min, supporting that the output performance of AT-MASF-TENG is relatively stable for several cyclic compressions. For the long-term cyclic operations, the output performance of AT-MASF-TENG was maintained, showing the reduction of only 5-10 V (i.e., 1.22%

decrement) after 30 days (Figure 6e), while the device is placed in the lab environment where the relative humidity is always maintained between the 30-45%. These results indicate its outstanding durability/reliability. Although, the performance of the AT-MASF-TENG device is further examined under relatively high humidity conditions. In order to examine the influence of moisture in the air on the output performance of TENG, its electrical output is measured under various relative humidity conditions (Figure 6f-g). While the alcohol-treated and untreated MASFs are employed as triboelectric materials and compared their performance. As shown in Figure 6f-g, an average/peak output voltage value of TENG with an untreated MASF is dramatically dropped from ~380 to ~30 V (i.e., ~92 % decrement) when the RH is increased from 40 to 80 %. Whereas the output voltage of the TENG device with the AT-MASF is almost maintained up to the RH of 60 % and it's reduced further by increasing the RH to 80 %. However, the decrement percentage in the electrical output of AT-MASF-TENG at the RH of 80 % is noticed as ~50 %, which is not a high loss than compared to the TENG with untreated MASF. These results demonstrate that the AT-MASF is more reliable when compared to the untreated MASF to attain higher electrical output even in a humid environment. By detecting an almost stable electrical output in the AT-MASF-TENG even at the RH of 60 % the alcohol treatment of silk film is shown to performs better in humid environments. The proposed AT-MASF-TENG device with a stable electrical output for several weeks and an RH of 60 % is desirable for many practical and real-time applications.

2.7. Practical and real-time applications of MASF-TENG device

To demonstrate the feasibility of AT-MASF-TENGs in practical applications, the electrical output generated by the AT-MASF-TENG device is linked to several low-power electronics including LEDs and liquid crystal displays, as shown in **Figure 7**. The electrical output generated by the TENG is an alternating signal, which is not suitable to directly supply and operate portable

electronic devices. So, the alternating signal generated by the AT-MASF-TENG is initially converted into the direct signal using a full-bridge rectifier, which was further utilized to charge a commercial capacitor. At last, the electric energy stored in the capacitor was utilized to operate various types of portable electronic equipment such as a timer and wristwatch. Figure 7a depicts the equivalent circuit model of the AT-MASF-TENG to charge the capacitor *via* the rectifier, followed by powering the portable electronic equipment.^[70,71] Figure 7b shows the rectified output voltage of AT-MASF-TENG under the compressive force and frequency of 3 N and 5 Hz, respectively, showing ~380 V. The voltage accumulated across various capacitors with the capacitances of 0.47, 4.7, 10, 22, and 47 μF was also measured while they were charged by the rectified voltage of AT-MASF-TENG (Figure 7c). As shown in Figure 7c, the capacitor with low capacitance (i.e., 0.47 μF) was charged instantly within few seconds and reached a high voltage of 27 V, whereas the capacitor with high capacitance took a long time to reach the high voltages. The charging-discharging curve of a 0.47 μF capacitor noted by the rectified voltage of AT-MASF-TENG is shown in Figure S8. Finally, the practical feasibility of AT-MASF-TENG is demonstrated by employing the electric energy stored in the capacitor to operate various portable electronic equipment. Figure 7e-f and Video S1 show the photographic images and movie of portable electronic equipment (i.e., timers and wristwatches), which are powered by the electric energy stored in two series-connected 47 μF capacitors (i.e., the resultant capacitance is noted as 23.5 μF). The charging and discharging curves of the capacitors by the rectified voltage of AT-MASF-TENG for powering different types of portable electronic equipment are shown in Figures 7g and h, respectively. Moreover, these curves reveal that within 60-70 s, the capacitors were charged up to 2 V by the rectified electric signal generated by the AT-MASF-TENG device. Whereas, both the timer and wristwatch connected to the capacitor were turned on for ~40-60 s,

which is also noted as the discharge lifetime of the capacitor. Besides, several green LEDs connected in series, spelled as “MASF TENG” directly turned on by the operating AT-MASF-TENG without any additional electrical components (e.g., rectifiers and capacitors), as shown in Figure 7d and Video S2. Therefore, these demonstrations verify that the high-performance AT-MASF-TENG prepared by the simple and inexpensive technique can be successfully employed for powering various types of portable/low-power electronic equipment for commercial applications.

Since the electrical response of TENGs is very sensitive to the strength and rate of mechanical agitations, it can be also employed for self-power sensing applications.^[72-76] Besides, the power or the strength of energy produced by each body part of a human is different during their corresponding activity.^[61] So, the proposed AT-MASF-TENG is also employed to sense various human body activities as well as the intensities of the corresponding activity, as shown in **Figure 8** and Figure S9. Herein, the electrical response (i.e., either output voltage/current) of the TENG is considered as the principal parameters to identify or sense the various types of such human activities. In this regard, a flexible device is developed by utilizing the flexible plastics sheets instead of the rigid supporting acrylic plates. The photographic images of the flexible AT-MASF-TENG device are shown in Figure 8a. Such a flexible AT-MASF-TENG device is located either on an object (like a table) or at different places of the body such as elbow, opisthenar of the hand, and the knee (Figure 8b). As shown in Figure 8c-f and Video S3, the AT-MASF-TENG exhibited a distinct electrical output response during the dissimilar types of human body activities. However, the average output voltage/current values of TENGs located on various parts of body are noted as ~150 V/13 μ A, 55 V/6 μ A, 11 V/1.8 μ A, and 15 V/3.8 μ A, respectively, while touching it with the fingers, folding/unfolding of hand, closing/opening the palm, and moving leg, respectively.

Consequently, by correlating the distinct electrical response of TENG, the movement of the corresponding human body part can be significantly identified or sensed.

To further demonstrate the potential applications of the proposed TENG device, it's also employed to sense the stress or pressure levels of the person, bending angle of arm/leg, and the other daily human body exercises like walking, running and punching a boxing bag, etc. (Figure 8g and S9). As shown in Figure 8g and Video S4, the strength of the force applied to the stress ball by the person can be recognized in terms of the output voltage generated by the flexible TENG device located on opisthenar. The person under very high stress may press the stress ball strongly, whereas it could be lighter under low stress. Accordingly, the stress level felt by the person or the strength of the corresponding human activity can be sensed by the flexible AT-MASF-TENG. Besides, the bending angle of the arm or leg is also sensed by the proposed TENG located either on the elbow or knee, respectively (Figure S9a, b). In the above cases, the variation in the electrical response of the TENG device can be mainly attributed to the variation in the contact area between the active materials as well as the strength applied on the device during the various human activities. Similarly, the daily human body exercises like standing, walking, running, jumping, and punching a boxing bag are also sensed or recognized by the electrical output response of the AT-MASF-TENG (Figure S9c, d, and Video S5). Therefore, all the above studies are revealing that the action of each human body part can be sensed by correlating its strength and rate with an electrical output response of the proposed AT-MASF-TENG. Also, it is elucidating that it can be located at various human body parts and utilized to sense daily human activities as well as the intensities of the corresponding firmness of activities. Such a system can simply be used with artificial intelligence tools to identify the different functionality and monitor everyday activities of humans.

3. Conclusions

In summary, a natural eco-friendly silk film with high surface roughness and resistance to aqueous solubility was developed by the facile and inexpensive SIL technique and alcohol annealing treatment. The post-alcohol treatment considerably enhances the water insolubility of silk film from a few minutes to several hours, and thus it can be expected to be employed in wearable and implantable devices for human skin and body. For the first time, the influence of various parameters like the selection of triboelectric materials, its surface roughness, external compression frequency, and force, etc., on the electrical output performance of silk film based TENG is analyzed by the theoretical DDEF model as well as the experimental measurements. Resultantly, the AT-MASF-TENG exhibited very high surface charge density, J_{SC} , V_{OC} , and power density values of $101 \mu\text{C}/\text{m}^2$, $89 \text{ mA}/\text{m}^2$, 395 V and $22.05 \text{ W}/\text{m}^2$, respectively, as compared to the flat SF-TENG (i.e., $78 \mu\text{C}/\text{m}^2$, $40 \text{ mA}/\text{m}^2$, 230 V and $15.13 \text{ W}/\text{m}^2$). Moreover, it also exhibited good durability after 10000 cycles and long-term stability after several weeks. Finally, the feasibility of AT-MASF-TENG is successfully demonstrated by applying its electric output to power various types of commercial portable electronics and employed for real-time human body sensing applications. From all the above results it could be concluded that the proposed TENGs integrated with biocompatible silk films can be successfully employed for sensing daily human activities and wearable/implantable biomedical applications.

4. Experimental Details

Materials: *Bombyx mori* silkworm silk cocoons were obtained from the local silkworm farmers in Suwon, South Korea. ACS reagent grade chemicals, such as sodium bicarbonate (NaHCO_3 , $\geq 99\%$)

and lithium bromide (LiBr, $\geq 99\%$), dialysis sacks (MW > 12,000 Da.), and Mira cloth were purchased from Sigma-Aldrich Co., South Korea, used directly without any further purification to synthesize silk fibroin film. Sylgard 184 (Dow Corning Co.), Si <100> wafers (p-type, boron-doped; iTASCO) were used to fabricate an MG-PDMS mold.

Synthesis of silk fibroin solution: Silk cocoons were cut into pieces with a size of < 1 cm. Firstly, to remove the sericin of silk fibers, cocoon pieces were degummed in boiling water with 50 mM NaHCO₃ for 30 min, followed by rinsed with deionized (DI) water for 4-5 times and dried under the ambient conditions (23 ± 2 °C and $40 \pm 10\%$ relative humidity). Secondly, the silk fibroins were completely dissolved in 9.3 M LiBr solution at 60 °C for 4 h. Thirdly, the silk fibroin solution was filtered through Mira cloth and then dialyzed with a generated cellulose dialysis membrane against the Millipore water for 1-2 days to eliminate salt (i.e., LiBr) in the solution, producing 10% (wt/vol) silk fibroin solution. The resultant silk fibroin solution was filtered again through Mira cloth and was stored at 4 °C before use.

Fabrication of MASF by SIL technique: The prepared silk fibroin solution was simply poured onto the flat substrate (i.e., plastic petri-dish) and MG-PDMS mold to fabricate a flat- or MA-silk films (Figure 1b) and is dried (or cured) at room temperature for 24 h. Afterward, the flat- or MA-silk films were carefully peeled off from the corresponding molds.

Fabrication of MG-PDMS: The MG-PDMS mold was produced by simply pouring the mixture of the base resin and curing agent (Sylgard 184, Dow Corning Co.) with a 10:1 weight ratio onto the microarchitected Si molds, followed by the curing treatment at 75 °C for 2 h. Herein, the microarchitected Si molds are developed by the well-known potassium hydroxide (KOH) alkaline etchant process.

Design of HASF-TENG: The regenerated MASF before or after the alcohol treatment was used as one of the triboelectric friction layers to design a TENG device. Whereas, the polytetrafluoroethylene (PTFE) film was used as another triboelectric friction layer. Both the films with the dimensions of $2 \times 2 \text{ cm}^2$ were stuck on the top of the aluminum foils with the same dimensions, which are employed as the top and bottom electrodes of the TENG, respectively. The top and bottom stacked layers (i.e., PTFE/Al and MASF/Al) were further glued on to the acrylic plates as supporting substrates, respectively. Finally, four springs were located at the corners of the supporting substrates to design a MASF-TENG, while both the PTFE and silk films were opposite to each other (Figure 1c).

Characterization: The surface morphologies of the as-prepared flat- and MA-silk films are analyzed using an FE-SEM (LEO SUPRA 55 Carl Zeiss) system. The structural characteristics of silk films are further examined by FTIR (JASCO FT-IR 470 Plus spectrometer) and XRD (M18XHF-SRA) measurements. The electrical output characteristics (i.e., V_{OC} , I_{SC} , and charge density) of SF- or MASF-TENG were analyzed using a digital phosphor oscilloscope (DPO4104; Tektronix) through an input impedance of $40 \text{ M}\Omega$, a low-noise current preamplifier (SR570; Stanford Research Systems) connected to the oscilloscope, and an electrometer (Keithley 6514), respectively. During this analysis, a compressive force was applied on the TENG device through a custom-made pushing machine, which is functioned with the nitrogen (N_2) gas flow.

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Figure 1. Three-dimensional schematic diagram to illustrate the fabrication/designing processes of (a) silk fibroin solution using typical white silk cocoons, (b) MASF by soft imprinting lithography technique *via* MGA-PDMS mold, and (c) AT-MASF-TENG device.

Figure 2. (a) XRD patterns and (b) FTIR spectrum of SFs before and after the alcohol treatment. Photographic images to demonstrate the (c) solubility or (d) insolubility function of silk fibroin films in aqueous solution, before or after the post-treatment process with alcohol, respectively. (e) Schematic representation to demonstrate the triboelectric electric energy generation mechanism.

Figure 3. Tilted-surface view atomic force microscopy (AFM) images of (a) flat and (b) microarchitected silk films i) before and ii) after the alcohol treatment. (c) COMSOL Multiphysics simulation results to demonstrate the surface potential distribution between the active materials of TENG by employing either the flat or microarchitected SF. (d) The structure model and charge distribution of a PTFE and silk films based TENG used for DDEF theoretical simulations. (e) The simulated peak output charge and current density values of TENG as a function of the surface area of silk films.

Figure 4. Measured (a) charge density, (b) J_{SC} and (c) V_{OC} curves of TENG with the flat and microarchitected SF before and after the alcohol treatment. (d) The distance-dependent variation of the electric field against the increasing distance from AT-MASF to other counter triboelectric materials like paper, copper, kapton, PDMS, and PTFE. DDEF theoretical simulation output (e) charge density and (f) J_{SC} curves of TENG, while the AT-MASF sheet, corresponding to other counter triboelectric materials; paper, copper, kapton, PDMS, and PTFE, etc. The measured (g) charge density (h) J_{SC} and (i) V_{OC} curves of corresponding TENG device with the respective triboelectric materials. During all these measurements, the compressive force and frequency applied on TENG are maintained at 3 N and 5 Hz, respectively.

Figure 5: Calculated electrical output (a) charge density and (b) J_{SC} curves of AT-MASF-TENG, by DDEF simulation model. Measured electrical output characteristics such as charge density, J_{SC} , and V_{OC} curves of the AT-MASF-TENG under various external compression (c-e) frequencies and (f-h) forces in the range of 1-5 Hz and 1-10 N, respectively.

Figure 6. Influence of external load resistance on the measured electrical output voltage and current values of (a) flat SF-TENG and (b) AT-MASF-TENG devices under the external compressive force and frequency of 3 N and 5 Hz. (c) Dependence of the estimated output power densities of flat SF- and AT-MASF-TENG on the external load resistance in the range of 10² to 10⁹ Ω. Enduringness test of the AT-MASF-TENG device was examined (d) over 33 min (or 10,000 compressive cycles) and (e) even after 30 days, under a compressive force of 3 N. Measured output voltage (f) curves and (g) average values of MASF- and AT-MASF-TENG devices at various relative humidities ranging from 40 to 80%.

Figure 7. (a) Equivalent circuit model of the bridge rectifier to convert the alternating current signals of AT-MASF-TENG into direct current to power the portable electronic equipment *via* commercial capacitors. (b) The rectified output voltage of AT-MASF-TENG. (c) Voltage accumulated across the capacitors with the capacitances of 0.47, 4.7, 10, 22, and 47 μF , which are charged by the AT-MASF-TENG through a bridge rectifier. (d) Photographic images of 85 green commercial LEDs connected in series, directly turned on by the AT-MASF-TENG. Portable electronic equipment like (e) timer and (f) wristwatch was successfully switched on by the voltage accumulated across two series-connected 47 μF capacitors (with the resultant capacitance of 23.5 μF), which are charged by the rectified electrical output generated from the AT-MASF-TENG device.

Figure 8. (a) Photographic images of flexible AT-MASF-TENG employed for real-time sensing applications. (b) Flexible AT-MASF-TENG device to sense various human activities locating it on different places of body such as under finger, on the elbow, opisthenar of hand, and knee, respectively. (c-f) The output voltage and current curves noticed by flexible AT-MASF-TENG while sensing various human body activities like knocking with finger, folding hand, closing palm, and moving leg, etc. (g) Output voltage generated by the AT-MASF-TENG while pressing the stress ball at distinct stress or pressure levels.

Supporting Information

Exploring theoretical and experimental optimization towards high-performance triboelectric nanogenerators using microarchitecture silk cocoon films

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Figure S1. The schematic diagram depicts the alcohol treatment processes of the MASF.

Figure S2. Photographic images to demonstrate the water insolubility function of alcohol-treated silk film up to almost a week. These images are also elucidating that even though the silk film is bent or shrink partly after soaking it for a week, the film is still flexible and not brittle.

Figure S3. SEM images of (a) flat and (b) microarchitected silk films i) before and ii) after the methanol annealing treatment.

Figure S4. AFM images of microarchitected silk films (i) before and (ii) after the alcohol treatment, while observing through different orientation (or angle). The AFM image of the MASF after the alcohol treatment is clearly indicating that the surfaces of MAs are slightly varied and result in the small nano scratches or particles owing to the alcohol treatment. Such variation on the surface of MASF has also slightly influenced its surface roughness.

Supplementary Note-I:

Considering the electric field equation for the charged triboelectric sheet given in eqn. 1, the output trends of the TENG can be derived as follows.

As discussed in our previous work,[S1,S2] considering the open circuit scenario, the potential of the negative (PTFE) TENG layer (Φ_N) and positive TENG layer (Φ_P) are given by:

$$\Phi_n = \frac{-\sigma_T}{\pi\epsilon_1} \left[\int_{x_1}^{x_1+z} f(x)dx \right] \quad (S1)$$

$$\Phi_P = \frac{\sigma_T}{\pi\epsilon_2} \left[\int_{x_2}^{x_2+z} f(x)dx \right] \quad (S2)$$

Whereas the open circuit voltage (V_{OC}) of the system is defined by:

$$V_{OC} = \Phi_P - \Phi_N \quad (S3)$$

Considering the short circuit scenario, the short circuit output charge density (σ_U) is given by:

$$\sigma_U = \frac{\sigma_T [\frac{1}{\varepsilon_1} \int_{x_1}^{x_1+z} f(x) dx + \frac{1}{\varepsilon_2} \int_{x_2}^{x_2+z} f(x) dx]}{(\frac{1}{\varepsilon_1} + \frac{1}{\varepsilon_2}) \int_0^{x_1+x_2+z} f(x) dx} \quad (S4)$$

Therefore, the short circuit current (I_{SC}) is evaluated using:

$$I_{SC} = A J_{SC} = A \frac{d\sigma_U}{dt} \quad (S5)$$

where J_{SC} is the short circuit current density, and A is the contact area of TENG layers.

Considering a load of resistance R connected between the electrodes of the TENG, and, using Ohm's law, the output generation of the TENG through the load can be evaluated using:

$$AR \frac{d}{dt} \sigma_U + \frac{\sigma_U}{\pi} \left(\frac{1}{\varepsilon_1} + \frac{1}{\varepsilon_2} \right) \int_0^{x_1+x_2+z} f(x) dx - \frac{\sigma_T}{\pi} \left[\frac{1}{\varepsilon_1} \int_{x_1}^{x_1+z} f(x) dx + \frac{1}{\varepsilon_2} \int_{x_2}^{x_2+z} f(x) dx \right] = 0 \quad (S6)$$

Using the charge output data from S6, the current and power output through the load can be evaluated. Therefore, eqn S1-S6 can be used to evaluate charge, current, voltage and power outputs of the TENG.

Note: The effect of the surface area of the TENG layers, and the motion parameters on the TENG outputs have been analysed in the theoretical and experimental studies of this work. Apart from that, the dielectric constant of triboelectric layers affects the output trends of the TENG, where, according to DDEF model simulations,[S2] lower dielectric constants are predicted to be favorable for higher output generation.

Supplementary References:

S1. R.D.I.G. Dharmasena, K.D.G.I. Jayawardena, C.A. Mills, J.H.B. Deane, R.A. Dorey, S.R.P. Silva, Triboelectric Nanogenerators: Providing a Fundamental Framework, Energy & Environ. Sci. 10 (2017) 1801-1811.

S2. R.D.I.G. Dharmasena, J.H.B. Deane, S.R.P. Silva, Nature of Power Generation and Output Optimisation of Triboelectric Nanogenerators, Adv. Energy Mater. 8 (2018) 1802190.

Figure S5. Triboelectric surface charge density approximation to perform the DDEF simulations.

Figure S6. The simulated peak output voltage values of TENG as a function of the surface area of silk films.

Figure S7. An opposite distance-dependent variation of the electric field observed by the counter triboelectric materials like paper, copper, kapton, PDMS, and PTFE, etc., while their distance increasing from AT-MASF.

Figure S8. Charging and self-discharging curves of a 0.47 μF capacitor observed by the rectified voltage of HASF-TENG.

Figure S9. The output voltage variations noticed by the AT-MASF-TENG located either on elbow or knee, while moving the (a) hand and (b) leg at different bending angles. Figure (a) and (b) also depicts the photographic images and schematic diagrams to illustrate the movement of hand and leg either in up or downward directions. (c, d) Output voltage observed by the AT-MASF-TENG either on elbow or knee, while sensing the various human activities like walking, running, jumping, and punching the boxing bag, etc.

Table.S1 Electrical output comparison of the proposed AT-MASF-TENG device with the recently reported silk film-based nanogenerators.

Type of material	length $\times$ width of materials	Thickness	Relative permittivity (ϵ_r)
Silk film	2 cm $\times$ 2 cm	100 μ m	3
PTFE film	2 cm $\times$ 2 cm	100 μ m	2
Al electrode	2 cm $\times$ 2 cm	100 μ m	10
<i>Instead of PTFE, we have also utilized some other triboelectric materials to optimize the output performance of the TENG</i>			
Paper	2 cm $\times$ 2 cm	100 μ m	3.7
Copper	2 cm $\times$ 2 cm	100 μ m	3
Kapton	2 cm $\times$ 2 cm	100 μ m	3.34
PDMS	2 cm $\times$ 2 cm	100 μ m	2.69
<i>For all the measurements and theoretical studies, the separation distance between the active materials of TENG is maintained to 5 mm (i.e., 5000 μm)</i>			

Table.S2 Electrical output comparison of the proposed AT-MASF-TENG device with the recently reported silk film-based nanogenerators.

Ref. No.	Type of nanogenerator; Mode of Operation	Triboelectric materials employed	Maximum voltage	Maximum current	Maximum power density
1	TENG; Contact-Separation Mode (CSM)	Electrospun silk film; Polyimide film	~ 18 V	~ 2.5 μA	~ 4.3 mW m^{-2}
2	Hybrid triboelectric piezoelectric NG (TPNG); CSM	Silk fibroin fibers; PVDF fibers	~ 500 V	~ 12 μA	~ 310 $\mu\text{W cm}^{-2}$
3	Transient TENG (T ² ENG); CSM	Silk fibroin film; Magnesium (Mg)	~ 60 V	~ 1 μA	~ 38.5 mW m^{-2}
4	Transparent TENG; CSM	Spray-coated silk fibroin; Polyethylene terephthalate (PET)	~ 213.9 V	-NA- (Not available)	~ 68 mW m^{-2}
5	Piezoelectric nanogenerator	Composite by silk fibroin solution, 1D wires, and lead-free ferroelectric nanoparticles	~ 2.2 V	~ 0.12 $\mu\text{A}\cdot\text{cm}^{-2}$ (Current density)	-NA-
Our work	TENG; CSM	Microarchitected silk film (MASF); polytetrafluoroethylene	~ 382 V	~ 26.3 μA	~ 22.05 $\text{W}\cdot\text{m}^{-2}$

Supporting Videos List:

Video S1. To demonstrate the practical applicability of HASF-TENG, by employing its electrical output to power various portable electronic tools *via* commercial capacitors.

Video S2. Employed an electric output generated by the HASF-TENG to directly turned on LEDs connected in series, without any additional electrical components.

Video S3. To demonstrate the ability of the flexible AT-MASF-TENG to sense various human body activities observed in everyday life, while it located on various body parts.

Video S4. Flexible AT-MASF-TENG device located on the opisthenar of the hand to identify the stress or pressure levels of the person.

Video S5. AT-MASF-TENG device device located on arm to sense its activity while punching a boxing bag.