멤브레인(Membrane Journal)
Vol. 33 No. 2 April 2023, 53-60

총 설

Print ISSN: 1226-0088
Online ISSN: 2288-7253
DOI: https://doi.org/10.14579/MEMBRANE_JOURNAL.2023.33.2.53

막 기반 마찰전기 나노 발전기: 총설

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(2023년 2월 21일 접수, 2023년 4월 15일 수정, 2023년 4월 17일 채택)

Membrane Based Triboelectric Nanogenerator: A Review

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(Received February 21, 2023, Revised April 15, 2023, Accepted April 17, 2023)

요 약: 기계적 에너지는 생물학 및 환경 시스템에서 트라이보 전기 나노제너레이터(TENG)로 얻을 수 있다. 웨어러블 전자제품에서 TENG는 진동 센서에 적용된 인간의 움직임에서 생체역학적 에너지를 수확할 수 있다는 점에서 많은 의미를 지닌다. 웨어러블 TENG은 습기에 취약하며, 폴리테트라플루오로에틸렌(PTFE)은 이러한 용도에 사용되는 우수한 소수성 물질이다. 높은 전기 음성 불소 원자의 존재는 매우 낮은 표면 에너지로 이어진다. 동시에 미세다공막 표면에 전자를 효율적으로 포획함으로써 소자의 성능이 증가한다. PTFE에 비해 상대적으로 적은 플루오라이드 원자의 존재로 인해 폴리비닐리덴 플루오라이드(PVDF)에서도 유사한 거동을 보인다.

Abstract: Mechanical energy can be harvested by triboelectric nanogenerators (TENG) from biological and environmental systems. In wearable electronics, TENG has a lot of significance as biomechanical energy can be harvested from the motion of humans, which is applied in vibrational sensors. Wearable TENG is prone to moisture and polytetrafluoroethylene (PTFE) is an excellent hydrophobic material used in these applications. The presence of highly electronegative fluorine atoms leads to very low surface energy. At the same time, the performance of the device increases due to the efficient capture of the electrons on the microporous membrane surface. This similar behavior occurs with polyvinylidene fluoride (PVDF) due to the presence of fluoride atoms, which is relatively less as compared to PTFE.

Keywords: *TENG, membrane, PVDF, PTFE*

1. Introduction

The need for sustainable and green energy is ever increasing in order to get rid of environmental pollution and global warming. Generating electricity by triboelectric nanogenerator (TENG) from mechanical energy is one of the exciting areas of research[1,2]. Advantages of TENG is that it is cheap and lightweight it can be easily fabricated and is highly efficient. TENG is mainly composed of a substrate, and two electrodes, one of which is negative and the other is positive. The most popular negative materials are polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF), polyimide (PI), and polydimethylsiloxane (PDMS) etc.

The PTFE membrane over the trapezoidal slit acts as a sensor in the system, which is made up of two pieces of PTFE membrane placed on an acrylic

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plate[3]. Harvesting of biomechanical energy with wearable devices is another application of TENG. It is useful to fabricate amphiphobic tribolelectric nanogenerators that can resist moisture and antistatic agents. FAS/PTFE was dip coated on garment–thermoplastic nanofiber membranes to achieve the above- mentioned goal. The growth of SiO_2 nanoparticles on the membranes improved the electrical output of the TENG[4].

To harvest biomechanical energy effectively, such as tensile joint movement from humans, wearable devices have to be stretchable. It was found that electrospinning of nanofibrous membranes improves TENG's performance since the method creates micro nanostructures that provide good flexibility. Hydrogel electrolytes in ionic conductors are embedded in polymer micro-networks rather than being mechanically doped, in contrast to the conductive composites such as silver nanowires, carbon nanotubes and conductive particles. This characteristic ensures that the conductive phase remains intact throughout deformation, producing materials with outstanding electrical conductivity, stretchability, and stickiness[5]. The introduction of functional groups enhances the performance of the triboelectric generators since they easily lose or gain electrons. For example, NH_2, which loses electrons easily, is present in polyaniline (PANI), a polymer that improves charge density and conductivity if introduced in the nanofiber. PANI can be synthesized on the nanofibers through chemical oxidative polymerization[6]. Extending the life of the TENG is important for their implementation. The addition of MoS_2 with outstanding lubricating characteristics to composite films efficiently boosts their surface charge density and increases their wear resistance[7].

The disk type TENG is an important TENG structure owing to its high output and energy-conversion efficiency, and static electrodes. It can be used in detection of rotating machinery faults. Another approach to the problem is to introduce prebent membranes to the rotating disk instead of the dielectric layer. What helps reduce friction is the deformation of the flexible membrane[8]. Implantable TENG can be used for spinal cord injury treatment by nerve stimulation. The wearable TENG harvests mechanical energy from human motion that provides the electrical energy required to stimulate the spinal cord to alleviate pain[9]. The comfort of wearable TENG is essential for their application. Through the use of an ultrasonic welding process, two layers of nanofiber membranes made of thermoplastic polyurethanes (TPU) supported by Ag elastic fabric and conducting polyvinylidene fluoride are joined to form arch structures. The TPU/Ag layer, which forms the base of the arch, ensures that the TENG may be stretched without breaking, which allows the harvesting of different types of biomechanical movements[10].

Increasing the surface area of the materials in contact enhances the performance of the TENG. It can be increased using patterned PVDF and nylon membranes and polymer membranes fabricated via the phase inversion method. The method can replace the electrospinning process for nanofibers and the patterning methods such as photolithography that cannot be used on PVDF because of the etching process[11]. PVDF and Nylon are usually used as triboelectric positive and triboelectric negative materials. However, in a recent study, peritoneum, a more sustainable material was used as a triboelectric positive material. It is found in the abdominal wall and diaphragm of mammals and is composed of a network of elastic fibers. It makes the TENG a greener and low cost energy harvester[12]. Graphene is an excellent conductor. Thus, it is used as an electrode material for electronics. When the graphene membrane is given a crumpled morphology, stretchability is improved due to improved areal density. Furthermore, the membrane maintains electrical properties after repeated cycles of deformation [13]. Ferroelectric materials, which convert mechanical and thermal stimuli into electricity, are flexible, treatable, lightweight, and strong. They are used to enhance the triboelectric effect when included in β-phase PVDF. The addition creates PVDF with a high phase composition, which has a higher dielectric constant and a higher negative charge. Thus increasing the β-phase

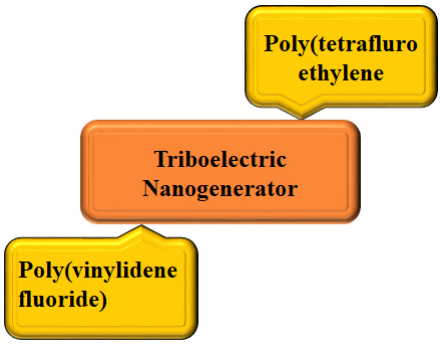

Fig. 1. Classification of TENG based membranes.

of PVDF increases the output energy of TENG[14]. This review is classified into PTFE and PVDF membrane based TENG and presented in Fig. 1.

2. Triboelectric Nanogenerator

Researchers have put a lot of emphasis on flexible and stretchy wearable electrical devices. This kind of electronics offers a lot of potential to create flexible, shape-adaptive gadgets that can be applied to a variety of uneven surfaces. Paper-based triboelectric nanogenerators (P-TENG) were fabricated with commercially available and environmentally friendly materials derived from cellulose via an easy and cheap method using print paper as the substrate and crepe cellulose paper (CCP) with nitrocellulose membrane (NCM) as friction layers[15]. The fabricated PTENG yielded great triboelectric performance with an output voltage of 198.6 V and 31.5 current and power density of 16.1W/m^2 along with high recyclability potential. This output is due to the considerably different tribopolarities of the NCM and the CCP. The device's capability for harvesting mechanical energy and performing self-powered sensing was demonstrated to be excellent through the use of a P-TENG- based paper piano for human-machine interfacing.

TENGs require highly stretchable components[16]. Hence, a lightweight, stretchy, porous, and thin adjustable Polyurethane (PU) membrane-based triboelectric nanogenerator was fabricated through an environmentally friendly and cost effective method. The TENG's maximum output voltage was 58.5 V, its current was 1.37, and power density of reached 9.7 Mw/m^2. The TENG demonstrated stable cyclic charging and discharging property. Furthermore, the device efficiently harvests mechanical energy and converts it to electrical energy.

2.1. Poly(tetrafluroethylene)

Due to the high electron-accepting properties of its carbon-fluorine bonds, PTFE is the most electronegative substance with a charge affinity. To improve the electrical output performance of TENGs, tribo-materials with unique differences in electron capture often create a greater surface charge. As a result, PTFE has been a material employed often in the creation of TENGs. This study focuses on the wearability of the triboelectric nanogenerator by making it waterproof and breathable[17]. It solves the tradeoff problem between wearability and triboelectric output. The TENG negative material is based on the expanded polytetrafluoroethylene (ePTFE) sheets that were stretched into microporous membrane. Increased pore size hinders the the output whereas increased porosity increases the permeability of the TENG. Therefore, the membrane was stretched at a lower temperature of 40°C into a membrane with small pores while maintaining a high percentage of porosity of 60%. Finally, the increased fibrils expanded the surface area, which favored the triboelectric output. The resulting short-circuit current, open-circuit voltage, and transferred charge was about 9.5 A, 120 V, and 44 nC, respectively. By continuing to stretch the membrane transversally, the moisture permeability was improved from 5200 g/m^2 · 24 h to 7000 g/m^2 · 24 h. The device exhibited the capacity of detecting knee motion and converting it into watchable electric signals.

In this study, a biocompatible TENG was fabricated using biomaterial eggshell membrane (ESM) as a positive material for its excellent piezoelectricity and environmental friendliness, and using porous expanded polytetrafluoroethylene (ePTFE) as a negative material

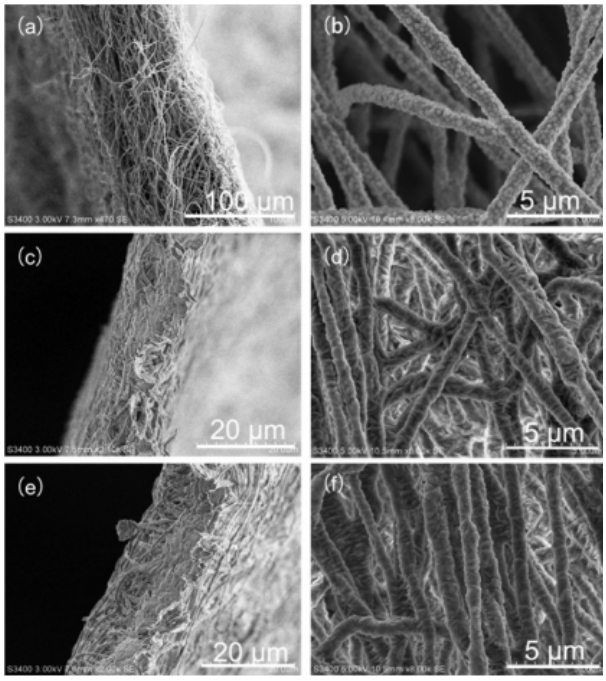

Fig. 2. Fiber cross-section SEM images of a PEO/PTFE composite nanofiber membrane (a) before and after the sintering process at (c) 350°C and (e) 370°C and surface morphologies of a pristine ES PTFE/PEO electrospun samples (b) before and after the sintering process at (d) 350°C and (f) 370°C, respectively (Reproduced with permission from Zhao *et al*.,[19], Copyright 2018, American Chemical society).

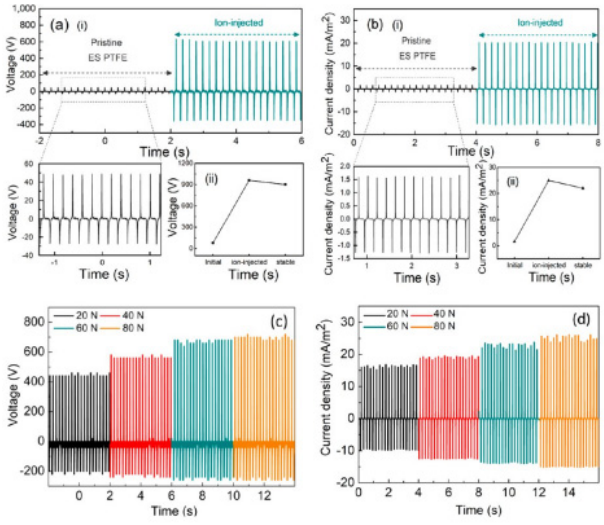

Fig. 3. Electrical measurement results of the sintered ES PTFE/PA6 TENG, before and after ion injection. The (a-i) open circuit voltage (V_{oc}) and (b-i) short-circuit current density (J_{sc}) measurements were carried out at an impact force of 50 N, a frequency of 5 Hz, and a spacer distance of 5 mm for the TENGs. Further measurements of Voc and current density as a function of impact force on the ion-injected ES PTFE/PA6 TENG are shown in (c) and (d), respectively. The TENG had received one ion-injection process. The insets (ii) in (a) and (b) show the stability of open voltage and current density after the ion-injection step (Reproduced with permission from Zhao *et al*.,[19], Copyright 2018, American Chemical society).

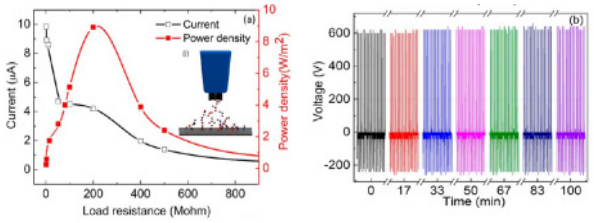

Fig. 4. a) Power density vs load resistance curve, showing a maximum output power of ~9 W m^{-2} at a load resistance of 200 MΩ; the inset (i) is the schematic of the ion-injection process. (b) Stability of the ion-injected surface charge on the ES PTFE surface as measured via the operation of ion-injected ES PTFE/PA6 TENG for 30 000 energy generation cycles at a working frequency of 5 Hz (Reproduced with permission from Zhao *et al*.,[19], Copyright 2018, American Chemical society).

[18]. ESM was carbonized to acquire enhanced piezoelectricity due to its carbon structure. Its piezoelectricity was compared to heated ESM and raw ESM. ESM was paired with ePTFE that showed excellent ability in harvesting energy because of ePTFE's great acquisition capability and ESM's excellent electron donating capacity. The device showed great sensitivity, performance, and biocompatibility which makes it suitable for biomedical applications.

In this study electrospun polytetrafluoroethylene (PTFE) is used in applications of TENG. First, emulsion electrospinning was used to synthesize polytetrafluoroethylene/polyethene Oxide (PTFE/PEO) membranes[19] (Figs. 2-4).

Following that, through the sintering process, they obtained pure PTFE fibrous membranes. Polyamide 6 (PA6) membranes were synthesized by the phase inversion process. PTFE was in the bottom contact layer while PA 6 was in the upper contact layer of the vertical contact mode TENG. EFM electrostatic force mi-

croscopy and FTIR showed negative and positive charges on the PTFE sintered membrane due to the transfer of polyethene oxide positive charge. Therefore, to improve the triboelectric surface charge, a negative charge injection process was done on the electrospun PTFE, which replaced the positive charge on the surface. The voltage, current, charge density, and highly stable power output density reached 900 V, 20mA m^{-2}, 149 C m^{-1}, and 9 W m^{-2} respectively. The method of synthesizing the membranes was cost effective and ecofriendly and the membranes showed high chemical and thermal resistance in the TENG field.

2.2. Poly(vinylidene fluoride)

Up until recently, research in TENG has typically employed PVDF and its copolymers. For instance, TENG's contact material of choice is electrospun P(VDF-TrFE) poly(vinylidene fluoride/trifluoroethylene). In this study, 2D siloxene-polyvinylidene fluoride (S-PVDF) composite nanofibrous membrane is used as the negative material and nylon 6/6 as the positive material of the TENG[20]. Because of adequate ratio optimization and electrospinning, the membranes demonstrated the enhancement in dielectricity, electronegativity, and compressibility. The TENG delivered a peak power density of 13.5 Wm^{-2} and runs low power electronics and internet of things (IoT). TENG is highly sensitive to dynamic pressure while the capacitive pressure sensor (CPS) based on poly (vinyl alcohol) (PVA)-lithium bis (trifluoromethanesulfonyl)imide (PVA-LiTFSI) is highly sensitive to static pressure. Their combination realizes a hybrid pressure sensor unit (HPS) that can measure dynamic and static pressure at the same time without external power. The system is self-powered since the rectified voltage of the TENG acts as a source of charging for CPS. The HPS demonstrated great dynamic pressure sensitivity of 12.062 $VkPa^{-1}$ and 2.58 $VkPa^{-1}$ and static pressure sensitivity of 25.07 mV kPa^{-1} and 5.96 mV kPa^{-1}. With the integration of AI, 2 × 2 HPS was developed for a user identification access system. The user accuracy increased from 92% to 98%, which indicates the enhancement in security.

The addition of nanofillers in electrospun PVDF can enhance the β-phase component in PVDF and improve TENG performance[21]. Thus, new TENGs based on polyvinylidene fluoride PVDF, nylon 11 nanofibers, and zinc oxide nanowires (PVDF-ZnO NWs/nylon-ZnO NWs) were fabricated. The zinc oxide was included in the PVDF and nylon 11 during electrospinning. It was found that ZnO was aligned along the fiber axis, which resulted in the alignment of the nanowires with the polymer chains. It leads to the formation of the highly polar crystalline β-phase of PVDF and δ'-phase of nylon. The addition of ZnO to the polymers also improved their tensile strength and elastic modulus and thermal stability. The maximum open-circuit voltage, short-circuit current, and transferred charge quantity of the composite membranes attained 330 V, 10 μA, and 150 nC, respectively. The maximum power density of the TENG attained 3W/m^2 under an external load of 10~20 MΩ.

PVDF is commonly used in TENGs. In this study, liquid metal (LM) galinstan nanodroplets were introduced into electrospun PVDF-co-hexafluoropropylene (PVDF-HFP) nanofibers for the improvement of the TENG performance[22]. The TENG is comprised of PVDF-HFP/2% LM nanofiber membrane as a negative layer and thermoplastic polyurethane as a positive layer. The voltage and power density attained high values of 1680 V and 24 W/m^2, respectively. It is important to note that increased amounts of LM weakens the mechanical property of PVDF. However, 2% LM PVDF demonstrated an efficient balance between mechanical characteristics and electrical generation. The great output of the TENG is due to the improvement of surface potential, charge trapping capability, and capacitance. Furthermore, LM droplets enhanced the dielectric constant, caused a low dielectric loss, caused a higher content of γ-phase crystalline in PVDF-HFP, and induced the secondary polarization inside PVDF-HFP, which also contributed to the high output.

In this study, the liquid solid TENG is based on functionalized graphene oxide (F-GO) and PVDF mem-

branes[23]. The membranes were fabricated with different amounts of F-GO using the blade coating method. The functionalization of GO with 1H,1H,2H,2H-perfluorooctyltriethoxysilane (FOTS) improved the miscibility of F-GO with fluorine group in PVDF, which in turn induced β-phase configuration of PVDF. Therefore, the dielectric and polarization characteristics and hydrophobicity of the membranes were enhanced, resulting in the great performance of the TENG. The current and voltage attained 18.1 μA and 16.5 V, respectively. Furthermore, the maximum TENG power density reached 2.08 W/m^2, which can light up directly 30 LEDs with one droplet.

In this study, liquid-solid TENG based on PVDF was fabricated with improved polarity and adequate functional groups on PVDF[24]. The surface polarity of PVDF was improved through chemical bonding with silica nanoparticles and grafting with negatively charged 1H, 1H, 2H, 2H-Perfluorooctyltriethoxysilane (FOTS) with epitaxial growth method, which formed FOTS/SiNPs/PVDF (FSiP) membrane. The membrane has an excellent level of hydrophobicity and dielectric constant due to the induced fluorine-bearing silane chain and increased interfacial polarization. The output reached a current and voltage of 5.79 μA, and 28.3 V under a flowrate of 5.0 ml/s, respectively. The power density reached a maximum value of 420 mW/m^2 and cal light up to 120 LEDs.

In another study, liquid-solid TENG is fabricated based on poly (vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) and ionic liquid (PIL)[25]. Multiple PIL membranes are prepared with different amounts of IL with the establishment of a nanoporous structure via the evaporation phase inversion method. The nanoporous PIL membranes with high uniformity and hydrophobicity are synthesized using blade coating, and then they are deformed to form circular tubes for PIL-TENG cells' use. The output performance was excellent thanks to fluoride atoms doping, which increased the electronegativity, and the accessibility of more ions via the creation of an electrical double layer. The nanoporous PIL-TENG of 10 wt.% ionic liquid showed the best output by attaining a maximum voltage, current, and power density of 16.95 V, 2.56 μA, and 26.1 mW/m^2, respectively.

3. Conclusion

There is a continuous increase in demand in the area of portable and wearable devices for application in sensors and Internet of Things. TENG is an excellent device that is easy to fabricate, highly efficient, and cost effective. It can be used efficiently to power smaller devices used for various applications. PVDF is an excellent material with high mechanical strength and high negative polarity. The fluorine group present in the repeating unit gives rise to polymorphism among which polar β-phase with electrostatic induction. This review discussed the application of PTFE and PVDF membrane in TENG. The future direction of research in TENG should focus on replacing synthetic polymers with naturally derived polymers. Furthermore, searching for more biocompatible and stretchable materials is necessary for the TENG's implementation in everyday life.

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