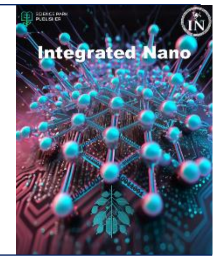
Integrated Nano, 2024, Vol. 1, Iss. 1, 41-47

DOI: 10.62184/in.jin010420243

Review Article

SCIENCE PARK PUBLISHER



Integrated Nano

Short Review about Challenges and Advanced Solutions of Higher Performance Piezoelectric Nanofibers Mats

Nader Shehata[1,2,3,*]

[1] Department of Engineering Mathematics and Physics, Faculty of Engineering, Alexandria University, Alexandria 21544 Egypt

[2] Department of Physics, Kuwait College of Science and Technology, Al-Asimah, 13133, Kuwait

[3] School of Engineering, Ulster University, Belfast, Northern Ireland, BT15 1AP, United Kingdom

Received: 27, 12, 2023; Accepted: 03, 09, 2024; Published: 29, 09, 2024

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Abstract

Piezoelectric nanofibers mats have been received an incremented interest in both research and commercial products for wide energy harvesting applications. Such nanofibers, with diameters less than one micron, can convert the mechanical excitations into electric signals with an improved efficiency according to formed internal electric dipoles along with higher surface-to-volume ratio, compared to bulky polymeric piezo-films. This paper introduces a brief review about the main challenges of piezoelectric nanofibers mats from different aspects including materials and processes. Then, the paper briefly discusses some recent solutions to overcome the challenges facing the piezoelectric polymeric nanofibers through materials additives and processes enhancement which can develop the piezosensitivity of the organic nanofibers.

Keywords: Piezoelectricity, Nanofibers, Scaling-up, Spinning processes, Energy harvesting

1. Introduction to Piezoelectric Nanofibers

The demand for sustainable and efficient energy management systems has been growing steadily, particularly in sectors such as agriculture, energy harvesting, and vibrational actuators [1]. In recent years, the growing demand for sustainable and renewable energy sources has led researchers to explore innovative methods of energy harvesting. One such method gaining attention is energy harvesting from footsteps. The concept involves converting the mechanical energy generated by human footsteps into usable electrical power. This approach has the potential to provide a sustainable and environmentally friendly solution for powering various applications, such as wearable devices, remote sensing systems, and smart infrastructure. Harvesting energy from footstep vibrations not only utilizes a readily available source but also offers the advantage of being applicable in various settings, including public spaces, industrial environments, and even everyday activities. Researchers have explored various techniques using piezoelectric materials such as Lead Zirconate Titanate (PZT) and polyvinylidene fluoride (PVDF), which exhibit the ability to convert pressure into electricity. Several studies have demonstrated the effectiveness of this approach, with outcomes like 40V generated from footsteps [2], improved voltage through parallel and serial connections [3], and even the ability to charge batteries using a relatively small number of steps [4]. Alternative methods include systems that use water pressure [5], controlled slip shoe designs [6], and integration

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with other renewable sources like solar panels and wind turbines to enhance overall energy generation [7].

The piezoresponse of a nanomaterial is dominant over macro-sized material of same kind due to its large surface to volume ratio, porosity, robustness, and easy reusability with excellent thermal property. Materials showing piezoactivity includes crystalline type, ceramic types and polymer type contents. Among these crystalline type materials are brittle and same time the ceramic type materials have toxicity, so polymer type piezomaterials with remarkable properties are our field of focus. Some of the remarkable properties of polymers are their flexibility, light-weight, mechanical stability and elasticity. In addition to pure polymers, nanocomposites prepared from the combination of either crystals or ceramics with pure polymers can greatly enhance the piezoresponse. Quartz and Rochelle salt are examples of crystals that exhibit high piezoelectric behaviour. Barium titanate, bismuth titanate, and zinc oxide single crystal are ceramics that exhibit adequate piezo behaviour [8]. Although the piezoelectric response of ceramic materials is higher than piezoelectric organic ones, the organic/polymeric piezoelectric structures offer a more environmental-friendly materials with no embedded lead element. Moreover, the organic films can be more mechanically-flexible and better stretchable for wider applicability in bendable wearable electronics and smart textile [9]. Therefore, there is an urgent need to overcome some technical drawbacks of piezoelectric organic/polymeric nanofibers to be competitive with the commercial ceramic piezoelectric sensor nodes. The following two sections show briefly some challenges of piezoelectric nanofibers mats and some recent techniques as solutions to some barriers against polymeric nanofibers against being commercially-used on a wider scale.

2. Challenges of Piezoelectric Nanofibers

There are different challenges against the development of piezoelectric nanofibers mats to be competitive to commercial ceramic piezoelectric nodes. In this short review, we will focus on mainly three obstacles: First challenge is the piezoresponse itself of piezoelectric nanofibers which is still relatively low, compared to the traditional ceramic piezo membranes. The highest piezoelectric coefficient (d_{33}) of some organic nanofibers including graphene embedded PVDF nanofibers has a range up to 39.7 pC/N [10], which is still less than the ceramics piezoelectric films with a range up to 640 pC/N [11, 12]. That leads to a less piezoelectric sensitivity, which means a lower generated voltage at applied mechanical stress, that can be reached by polymeric nanofibers compared ceramic films according to different factors of crystallinity degree, beta sheets formation, and concentration of aligned electric dipoles [13].

A second main challenge is the scaling-up capability of nanofibers to be compatible with the mass production and commercialization need. This is a general challenge not only for piezoelectric nanofibers, but also for all functionalized nanofibers mats. The traditional process of generating nanofibers is electrospinning, which depends on higher intensity electric field correlated to high voltage power supply between both sides of emitter (needle) and collector. When the repulsive force within the charged solution becomes higher than surface tension, then a polymeric jet is ejected from the tip of a metallic needle and then being accelerated toward a metallic collector side which is electrically grounded [14]. However, the productivity rate of the nanofibers from electrospinning is extremely low, of range 0.01 to 2 g/h, compared to other processes [15]. There are other processes which can offer a higher scaled-up piezoelectric fibers such as solution-blown spinning and melt-blown spinning which both depend on mechanical impact on the organic solutions [16, 17]. However, the mechanically-based spinning processes may not generate higher piezosensitivity of nanofibers, compared to electrospun piezoelectric nanofibers, due to the absence of electric field that may be more effective in alignment of dipoles and forming higher concentration of beta sheets, compared to mechanical impact [18]. In addition, the melt-blown spinning depends on extruding mechanism for the embedded polymers, which can negatively effect on the properties of the polymers along with possible phase changes with reduced piezosensitivity. Also, the melt-blown spinning can generate microfibers scale, larger than 10 microns, which is less effective in piezo generation compared to nanofibers [19].

One more challenge is related to the biocompatibility of the used piezoelectric polymers. Although most of organic nanofibers have no embedded lead, compared to most of piezoelectric ceramics, some polymeric nanofibers can be considered partially biocompatible. The used solvents to dissolve the organic piezoelectric initial precursors of the form of whether pellets or powder are not biocompatible such as Dimethylformamide (DMF) and chloroform. Therefore, the formed piezoelectric nanofibers mat includes a partial weight of non-biocompatible elements within the formed nanofibers mat [20]. Therefore, there is a need to use a more biocompatible solvents for PVDF, along with investigating more piezoelectric bio-synthetic materials.

3. Recent Solutions

To overcome the first challenge of relatively lower piezoresponse property of nanofibers, there are different recent solutions to increase the piezoelectric coefficients. Addition of materials in-situ within the organic solution is one promising technique to enhance the piezoresponse capacity. There are different recent research articles focusing on nanocomposite films consisting of flexible piezoelectric polymers with added nanofillers, such as ZnO and TiO_2, that function as heterogeneous nucleation sites for the β-phase, and consequently considered as attraction centers where PVDF chains are adsorbed over the outer surface of the nanoparticle [21]. In another way, a nanofiller is positioned in between isolated polymer backbones, that can develop micro-capacitor structures with a better charge accumulation inside the nanofibers [22, 23]. ZnO has significant spontaneous polarization as a common piezoelectric material including an asymmetric crystalline structure, and the films created from it also have better piezoelectric characteristics, with proved enhancement of piezosensitivity of PVDF nanofibers according to added with various ZnO forms, including nanoparticles, microrods, and nanorods [24-26]. The piezoelectric energy conversion efficiency of graphene-silver embedded with PVDF nanocomposites, is found to be 15% according to the plasma-coupled piezoelectric characteristics, with a high phase content and a 44.5% crystallinity using the special interface structure of silver nanoparticles [27, 28]. Another promising filler that has been embedded within PVDF is the carbon nanofibers (CNFs), according to the synergistic interaction between CNFs and additional electrical polarization processes. The CNFs/PVDF specimen containing 0.5 wt.% CNFs had a maximum voltage up to 5.80±0.17 V as well as a short-circuit current of 1.2 ±0.1 μA [29]. Multi-walled Carbon nanotubes (MWCNTs) are a viable alternative for their integration in PVDF due to their large surface area due to their tube-like shape, high mechanical tensile strength (5-200 GPa), high electrical conductivity (103-105 S cm^{-1}), and high electron mobility (104-105 cm^2 V^{-1} s^{-1}). When CNTs are added to PVDF, the polymer's electrical conductivity is improved, which improves the mobility of electrons inside the polymer matrix [30]. Research on the piezoelectric performance of an electrospun nanocomposite made of Poly (vinylidene fluoride)/ Potassium Sodium Niobate PVDF/KNN and various CNT concentrations was just published by Bairagi et al. [31]. Additionally, it is stated that the conductivity of CNT enhances the nanofibers' ability to stretch during the electrospinning process. Moreover, some recent research work of adding PZT ceramic nanoparticles to be embedded within PVDF nanofibers to enhance the piezoelectric response, with piezoelectric coefficient d_{33} up to 104.8 pC/N and generated voltage of 9.9 mV/N, through then hybrid integration of both piezoresponse-driven resources of both ceramic and polymer [32]. In another recent additive mechanism, some elastomers have been integrated within PVDF nanofibers to enhance the mechanical stretchability of the formed nanocomposite mat. Surprisingly, recent literature found that there is an optimum concentration of elastomer, such as thermoplastic polyurethane (TPU) up to 15 wt.%, which enhances the piezoresponse of PVDF along with improving the mechanical stretchability and maximum allowed strain due to the stretched zigzag chains of PVDF according to the added elastomer [33]. The results showed an enhanced piezoresponse sensitivity of more than 0.7 V/N at the case of

PVDF/TPU nanofibers, compared to 0.55 V/N within pure PVDF nanofibers [34]. Another technique of enhancing the piezoelectric performance of PVDF nanofibers is through post-treatment technique via making an additional process or impact on the already-synthesized nanofibers mat. Annealing the PVDF electrospun nanofibers mats up to 100°C is found to increase the remanent polarization up to 0.42 μC/cm^2, compared to less than 0.42 μC/cm^2 at no annealing post-treatment step with an enhanced d_{33} coefficient up to 16.2 pC/N [35]. Furthermore, PVDF/ poly(trifluoroethylene) (TrFe) nanofibers, which is one of the most promising piezoelectric copolymer of PVDF, has an increased piezoelectric constant up to 48.5 pm/V when annealed for 2 hours at 135 °C in vacuum environment, compared to 29.1 pm/V at non-heated nanofibers case [36].

To overcome the second challenge of scaling-up productivity, there are different ideas to enhance the productivity of the generated piezoelectric nanofibers and any nanofibers mats, in general. One example of scaled-up electrospinning process is the needless electrospinning mechanism such as Nanospider™ needle-free technology that has been presented by Elmarco Company has been used [37]. In this technique, the electrospinning with no nozzles have been developed to improve the nanofibers mats' productivity rate of nanofibers, compared to the traditional electrospinning. This mechanism depends on a matrix of small holes in a metallic drum immersed in the polymeric solution to form a multi-jet emitter, which reduces the possibility of sparks that can happen in the case of the traditional needle-matrix design of the normal electrospinning [38]. Another recent technique for scaled-up the generated piezoelectric nanofibers mats is to mix between electrospinning and solution-blown spinning in one process called electro-blown spinning (EBS). Elnabawy et al. illustrated the EBS process as a hybridization approach to produce nanofiber mats with higher scaling up with minimum beads and uniform mean diameter. Both effects of electric field and airflow merged driving forces showed a remarkable improvement in the produced structure as well as solution jet stability [39].

Regarding the fabrication of greener and more bio-friendly piezoelectric nanofibers, there are different trials to generate biocompatible and biodegradable nanofibers mats with an incremented piezosensitivity. For a more biocompatible PVDF, it is highly recommended to use Dimethyl sulfoxide (DMSO) rather than other solvents such as DMF. DMSO is considered a green solvent because it is not only has relatively low intrinsic toxicity, but is also biodegradable, with the capability of generating non-toxic mats [40]. Other natural piezoelectric polymers have been used to fabricate nanofibers mats, though the piezosensitivity may not be comparable to the response of PVDF ones. Collagen is an organic piezoelectric biocompatible and biodegradable polymer that is a functional protein found in mammals. The piezoelectric coefficient of collagen is relatively low with a range between 0.2 pC/N to 2 pC/N. However, the partial hydrolysis of the collagen polymer, extracted from the connective tissue of animals generates gelatin is found to have a much enhanced d_{33} coefficient up to 20 pC/N [41]. In another biodegradable material, spider silk has been extensively received a wide attention due to their remarkable mechanical properties. This naturally abundant spider-based silk material shows piezoelectric coefficient in the range 0.36 pm/V and the cocoon silk-based biomaterial exhibits a piezoelectric response of 1 pC/N. However, silk fibroin polymer results in a higher piezoelectric coefficient (d_{33}) up to 38 pC/N [42].

4. Conclusion

In this short review, some challenges have been represented against the development of piezoelectric nanofibers to be commercially competitive, when compared to ceramic piezoelectric mats. In addition, some recent and updated solutions have been discussed to improve both piezoelectric response and scalability of piezoelectric nanofibers mats.

Author Information

Corresponding Author: Nader Shehata*
E-mail: nader.shehata@alexu.edu.eg, n.shehata@kcst.edu.kw
ORCID iD: 0009-0009-3840-3044

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