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Piezoelectric performance analysis of co-axial electrospun PVDF/TPU nanofiber mats

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ABSTRACT

Energy demand is increasing daily due to the improvement in technology and energy-dependent devices such as wearable electronics. Nanogenerators have been studied intensively by researchers for the last 20 years. Nanofiber and polymer-based devices are the most popular ones among the others. A core-shell nanofiber structure by electrospinning was proposed in this study for the first time in literature due to high elastic properties of TPU and good piezoelectric performance of PVDF. PVDF/TPU nanofiber mats with the ratio of the core to the shell structure were changed. The diameter of the nanofibers and piezoelectric performances were characterized and compared with each other. As a result, a maximum voltage of 2.1 V, maximum current of 28.28 μA, a maximum power of 31.31 μW, and a maximum power density of 184.17 μW/g.cm-2 were obtained from the maximum ratio of 3:1 Poly (vinylidene fluoride): Thermoplastic polyurethane (PVDF:TPU).

I. INTRODUCTION

Electrospinning technology has significant advantages in the preparation of nanofiber materials. Nanofibers have many effects such as small size, surface and interface, quantum size, and quantum tunneling effects. It has very desirable features. As is well known, electrospinning products are obtained as non-woven mats. Researchers have focused on methods to produce oriented nanofiber bundles and threads [1, 2]. Oriented nanofiber bundles and nanofiber thread have significant value in many application areas, including filters [3–7], sensors [8–10], biomedical materials [11–13], tissue engineering scaffolds [14–17], drug delivery [14, 18–21], and wound dressings [22–26], nanogenerators [8, 27–36]. Issues such as the simplicity of the mechanism to be used, the ability to easily change the process parameters, and the wide choice of polymers to be used have made this method a common practice today and enable multiaxial electrospinning [8, 37–39].

PVDF/TPU core-shell nanofiber electrospun piezoelectric mats have garnered significant attention in the field of flexible and wearable electronics due to their exceptional piezoelectric properties and mechanical flexibility. Polyvinylidene fluoride (PVDF) and thermoplastic polyurethane (TPU) are combined to form a unique core-shell structure, where the PVDF core provides excellent piezoelectric response, while the TPU shell enhances the mechanical robustness and flexibility of the nanofiber mat. The piezoelectric effect allows these mats to generate electrical charges in response to mechanical stress. It enables them to convert mechanical energy into electrical signals, making them ideal for diverse applications, such as energy harvesting from body movements and

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biomechanical sensing. The core-shell structure ensures efficient charge separation and transfer, leading to enhanced piezoelectric performance and stability. These nanofiber mats hold great promise for the development of next-generation flexible electronics, wearable sensors, and smart textiles [40]. Given the fact that most piezoelectric systems operate between microwatts and milliwatts, the most common application of piezoelectric energy conversion is to provide energy for low-power electronics, including integrated electronics, implantable biomedical devices, wireless sensor nodes, and portable electronics [41–44].

Due to its polar crystalline structure, PVDF can produce potential with motions, making it suitable for piezoelectric and sensor applications. TPU can potentially enhance mechanical properties in the case of a mixture of other polymers [45–48]. PVDF/TPU fibrous materials from solution mixture were studied for various applications such as triboelectric, energy harvesting [49, 50] and sensor applications [51, 52] piezoelectric [45, 47, 53, 54] nanogenerator [55] for lithium-ion batteries [56–59] oil-water separation [60]. The core-shell structure of PVDF:TPU nanofibrous as tactile sensors was studied by Zhou et al. [61] due to elasticity and piezoelectric effects.

In this study, core-shell nanofiber production by electrospinning from TPU (core)/PVDF (shell) with different thicknesses of the shell structure is proposed to use the advantage of the elasticity of TPU while piezoelectric behavior of PVDF has the possibility to generate electricity from mechanical movements. The proposed core-shell structure enables more interaction between PVDF nanofibers while the TPU contributes to the mechanical strength and elasticity and recovers during the piezoelectric stimuli. The resulting nanofiber mats were characterized in terms of piezoelectric performance and compared with each other. SEM analyses were carried out for further characterization.

II. EXPERIMENTAL METHOD

2.1 Materials and Preparation Techniques

Poly (vinylidene fluoride) (PVDF) (Kynar 761, from Huaian Ruanke Trade Co. Ltd.), thermoplastic polyurethane (TPU) (from Ravathane 130 A70), dimethyl formamide (DMF, Tekkim Kimya), and acetone (from Sigma-Aldrich) were purchased to prepare a polymer solution for nanofiber production.

The appropriate amount of polymers was dissolved in DMF:acetone (1:1) at room temperature to obtain a 15% polymer solution for electrospinning. Polymer solutions were left to stir overnight in an ambient atmosphere with a magnetic stirrer for a homogenous solution. The electrospinning process was carried out by dosing the polymer through a syringe by means of a micropump, nanofibers were spun from bottom to top on a grounded rotating collector (at 250 rpm) with a diameter of 10 cm and a width of 15 cm. For the core-shell nanofiber production, a coaxial nozzle was used. As depicted in Figure 1, the shell polymer (PVDF) from the outer side and the core polymer (TPU) were fed from the inner side. A high voltage of 16 kV was applied to the metal coaxial nozzle.

Three different feed ratios of the polymer solutions were carried out: (PVDF:TPU) 1:1, 2:1, 3:1. While TPU solution was fed at 2 ml/hour for all the situations, PVDF solution was fed at 2, 4, 6 ml/hour for 1:1, 2:1, 3:1, respectively.

A total polymer solution of 20g (approximately 3g polymer) was used for each process. Approximately 30 cm*10cm nanofiber mat was produced. The areal densities of the mats are calculated approximately as 0,17 g/cm^2.

Nanofiber mats were cut into 5cm*5cm pieces, and both sides of the nanofiber mats were silver coated by magnetron sputtering, and the aluminum tape was stuck to each side as an electrode. Contacts were taken by a cable soldered on copper tape stuck to aluminum tape.

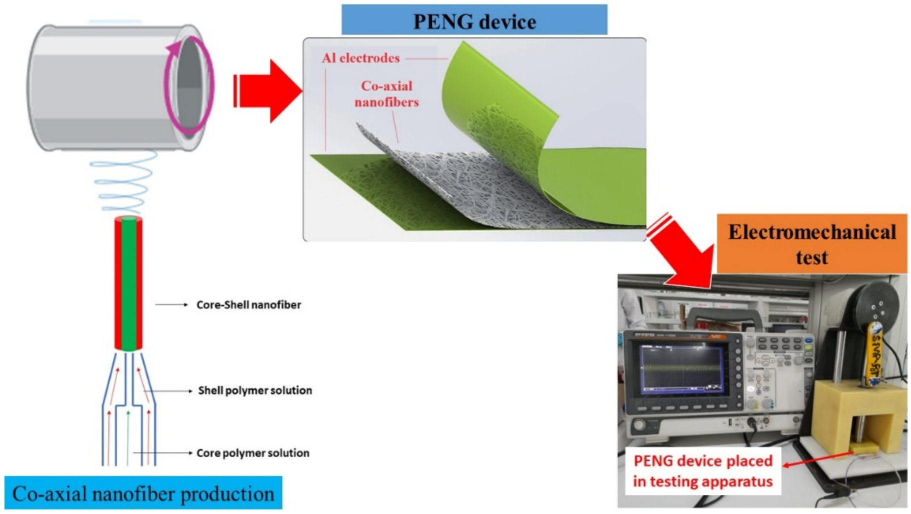

Figure 1. The depiction of core-shell nanofiber production, PENG device [10] produced thereof, and piezoelectric characterization set-up.

2.2 Characterization of materials

FTIR analyses of PVDF, TPU, and the three ratios of PVDF:TPU nanofibers are carried out. The core-shell structure was characterized using a scanning transmission electron microscope (STEM, Tescan MAIA3 XMU). A transmission electron microscope grid was placed on the collector during the electrospinning and a very thin nanofiber layer was collected on the grid prior to testing. Piezoelectric tests were carried out by a compression test device designed by Bedeloglu et al. that continuously applied 35 Pa pressure according to ANSYS simulation calculations [49, 62] and the electrical data was saved by an oscilloscope (GW-Instek 1102B) connected to the piezoelectric devices. A photograph of the characterization set-up is shown in Figure 2. Eight samples from each group were characterized, and the mean data was presented.

Figure 2. The photograph of piezoelectric characterization set-up.

III. RESULTS AND DISCUSSIONS

3.1 FTIR Analysis

Figure 3 shows the infrared spectrum of electrospun PVDF, TPU, and three PVDF/TPU nanofiber ratios. For TPU, the peak at 1704 cm^{-1} corresponds to the stretching vibration of C=O. 1108 cm^{-1} is attributed to C-O-C in the flexible chain in TPU. For PVDF nanofibers, 1400 cm^{-1} corresponds to C-F vibration. The peaks at 878 cm^{-1} and 840 cm^{-1} are related to the β phase in PVDF and the peak at 763 cm^{-1} is attributed to the bending vibration of the α phase [52, 63]. For the core-shell structures, characteristic peaks for both PVDF and TPU are shown evidencing the successful production of PVDF/TPU core-shell nanofibers.

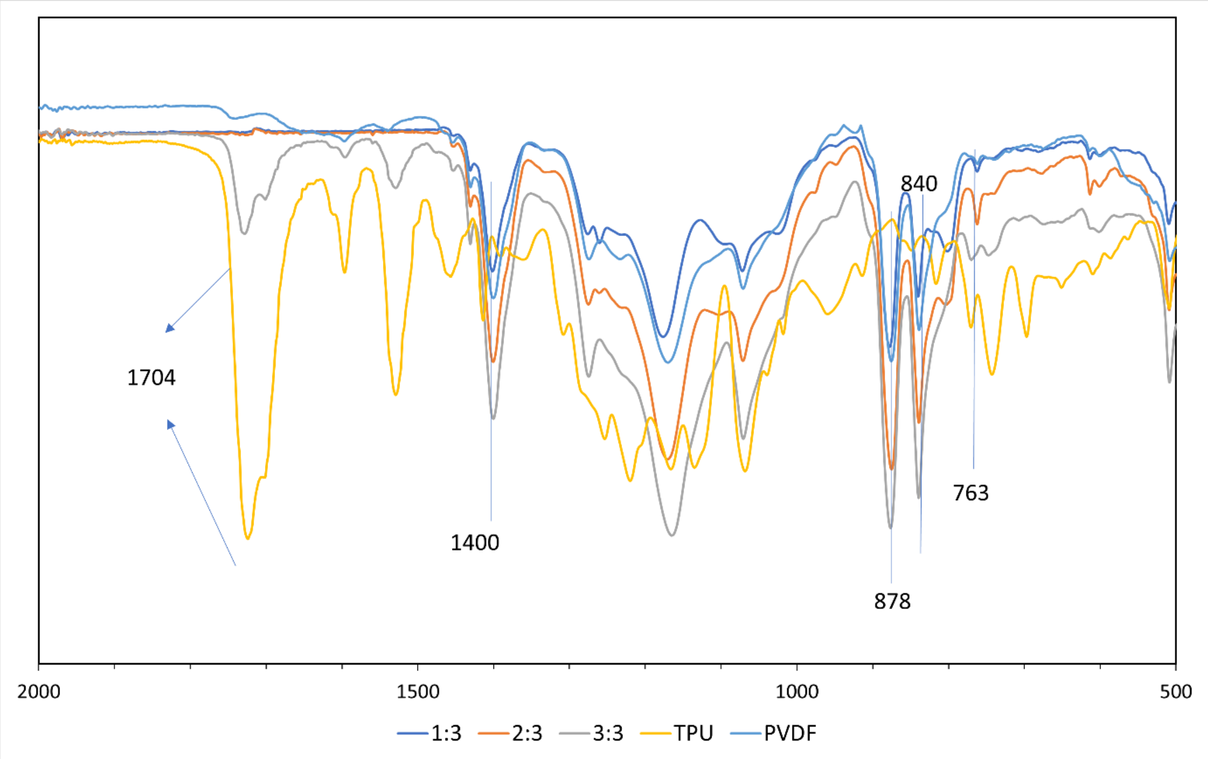

Figure 3. FTIR spectra of electrospun PVDF, TPU and TPU/PVDF nanofibers.

3.2 SEM Images

Scanning Transmission Electron Microscopy (S-TEM) images taken on copper grid by means of Scanning Electron Microscopy of single nanofibers produced by electrospinning are presented in Figure 4. STEM images were taken at 50 kX magnification with 30 kV voltage. The core-shell structure is clearly seen in the STEM images. Core (TPU) nanofiber diameter was 265 nm at 1:1 ratio, however increased by the increasing ratio. The rising ratio of PVDF drastically increased shell (PVDF) nanofibers. This can be explained as the applied voltage was constant. Since the applied voltage value remained the same with the increase in the polymer solution fed, the polymer solution was exposed to less electric field.

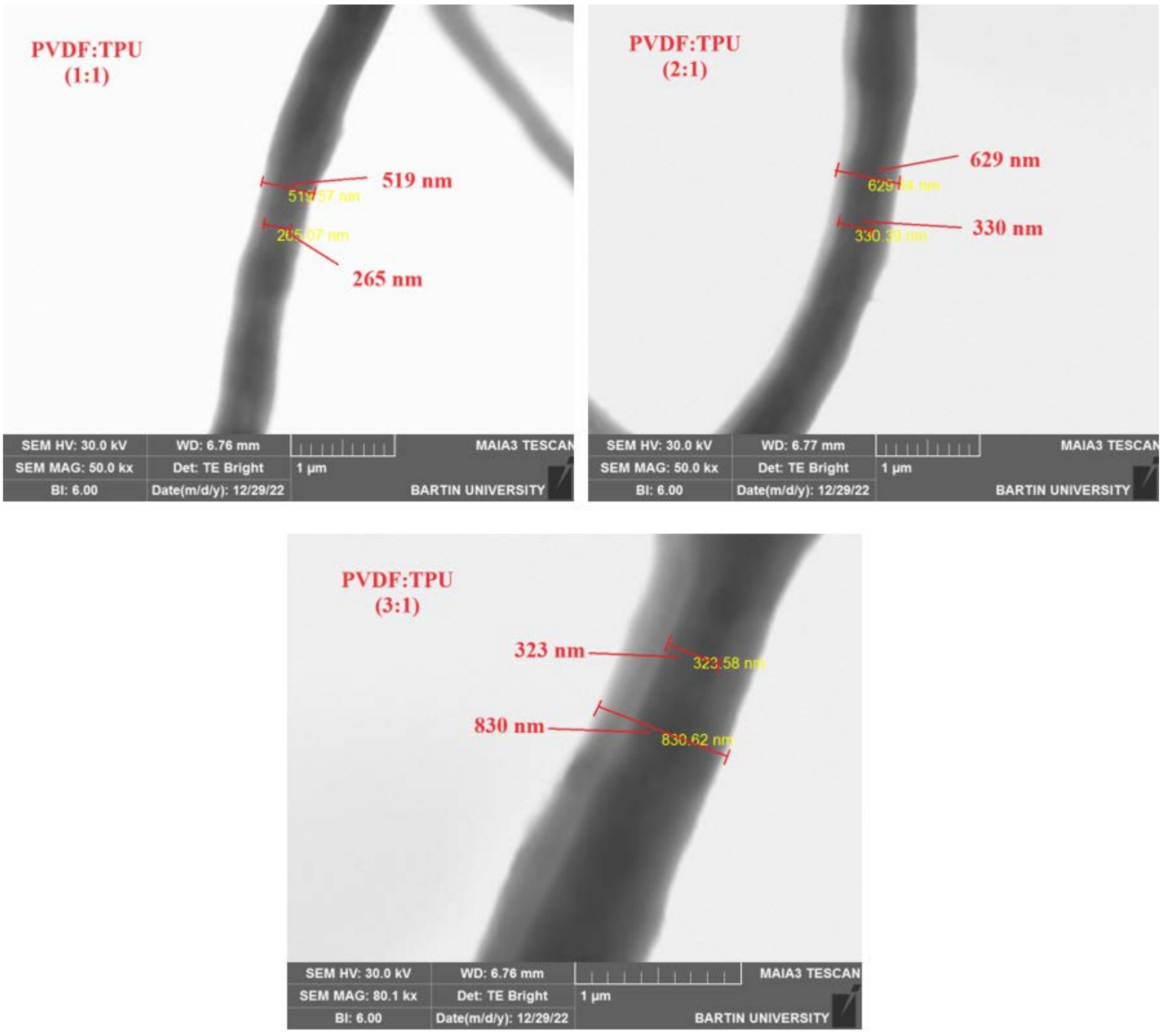

Figure 4. STEM image of the core-shell nanofibers.

3.3 Piezoelectric Characterization

The mean current values were 28.84, 25.76, 28.28 μA; the mean voltage values were 1.43, 1.66, 2.10 V; the mean power values were 20.09, 21.84, 31.31 μW; and power density values were 118.17, 128.47, 184.17 μW/cm^2 for the PVDF:TPU ratios 1:1, 2:1, 3:1, respectively.as given in Table 1.

As seen from the graphs presented in Figure 3, increasing the PVDF to TPU ratio improved the voltage and power outputs however current values were the highest at 1:1 and lowest at 2:1. The voltage increased 50% from 1:1 to 3:1 ratio, thus the power value increased with a similar trend. The 3:1 ratio maximized voltage and power outputs, however, the current value was not the maximum at this rate.

Table 1. Piezoelectric values of the produced nanogenerators

Core-shell ratio	**Current** (μA)	**Voltage** (V)	**Total Power** (μW)	**Power density** (μW/cm^2)
1:3	28.84	1.43	20.09	118.17
2:1	25.76	1.66	21.84	128.47
3:1	28.28	2.10	31.31	184.17

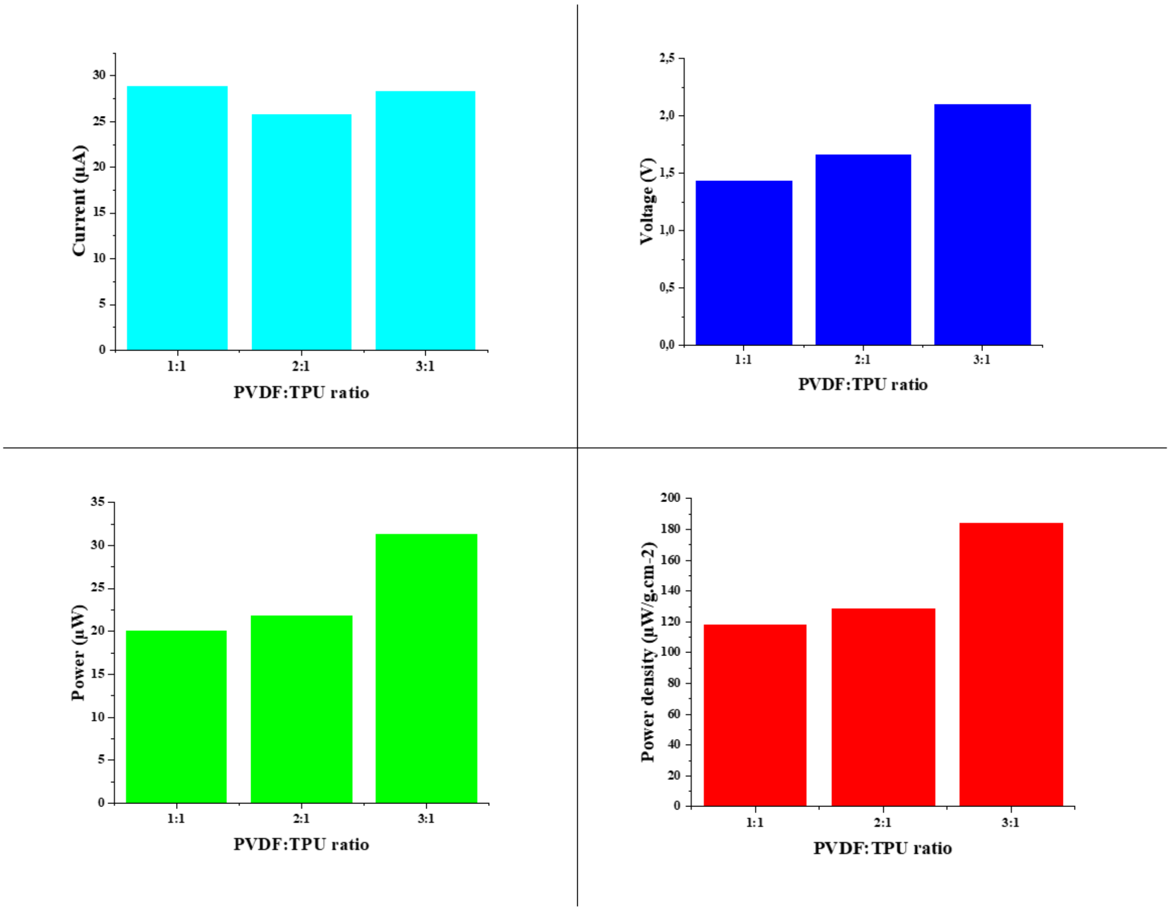

Figure 5. Current, Voltage, Power and Power density graphs of the core-shell nanofiber pieozoelectric devices.

IV. CONCLUSIONS

This study investigated the effect of the changing core-shell nanofiber structure from PVDF as shell and TPU as core and the impact of the changing core-to-shell ratio on the piezoelectric performance. This exciting approach was enabled by the feeding ratio of the different polymers during the electrospinning process. Regarding piezoelectric performances, core-shell nanofiber diameters and the three types of PVDF:TPU (1:1, 2:1 and 3:1) ratios were compared to each other regarding piezoelectric performances. The highest voltage, power and power density values were gathered from the maximum PVDF to TPU ratio at 3:1 by the rise of 50% from the 1:1 ratio. It can be understood from this study that TPU nanofibers have a significant change in the piezoelectric performance of the nanogenerators produced. By the addition of nano-additives [10, 62, 64], the nanogenerator energy harvesting efficiency can be improved for further studies. The proposed core-shell structure approach may enable to provide power for the portable microelectronic devices with flexibility and lightweight property.

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