



OPEN Self-assembly of polyvinylidene fluoride (PVDF) and carbon nanotubes into flexible piezoelectric nanofiber transducers

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We report a scalable fabrication strategy for flexible piezoelectric nanofiber transducers through a single-nozzle, self-assembling electrospinning process. Unlike traditional coaxial methods, this approach exploits the surface energy differential between polyvinylidene fluoride (PVDF) and multi-walled carbon nanotubes (MWCNTs) to drive the spontaneous formation of a core-shell architecture. The resulting 80 nm diameter fibers feature a predominantly crystalline β -phase PVDF shell, with molecular chains oriented along the [001] fibers axis, encapsulating a perfectly aligned MWCNT core that serves as an integrated internal electrode. Quantitative X-ray diffraction analysis confirms a five-fold increase in β -phase content relative to as-received PVDF powder, with a total crystallinity of 52% in the electrospun nanofibers. This unique coaxial configuration enables the demonstration of both direct and inverse piezoelectric effects in a single-step manufactured nanofiber device, without any post-fabrication electrical poling. The manufactured device yields an effective piezoelectric voltage coefficient of $g_{\text{eff}} = 0.22 \text{ V.m.N}^{-1}$, comparable to values reported for fully poled bulk PVDF films, and an effective piezoelectric strain coefficient of $d_{\text{eff}} = 2.8 \text{ pC.N}^{-1}$ under electrical excitation. The flexible, continuous nanofiber format, ten times thinner than spider silk, opens exciting possibilities for self-powered sensing, smart textiles, artificial skin, and structural health monitoring applications.

Keywords Piezoelectricity, PVDF, MWCNTs, Co-axial structure, Nanofiber transducer

Piezoelectric devices are pervasive in modern technology, spanning applications as diverse as accelerometers in mobile phones and automotive airbag systems, fiber-optic signal converters in transoceanic telecommunication cables, and transducers for medical ultrasound imaging. Across this broad spectrum of applications, countless sensors and actuators exploit the ability of piezoelectric materials to reversibly interconvert mechanical strain and electric polarization. The vast majority of the piezoelectric materials currently in use are ceramics, including zinc oxide (ZnO), lead zirconate titanate (PZT), and barium titanate (BaTiO_3), due to their excellent piezoelectric properties^{1–4}. Nonetheless, they all suffer from the mechanical stiffness and brittleness common to ceramics; furthermore, the fabrication of piezoelectric ceramics is relatively complex and costly, challenges that are further compounded by the relentless miniaturization of modern devices, which increasingly demand functional components at the micrometer and nanometer scales. The geometries accessible through ceramic processing are additionally constrained, being largely limited to thin films and polyhedral forms.

These challenges inherent to piezoelectric ceramics have stimulated interest in exploring piezoelectric polymers, particularly polyvinylidene fluoride (PVDF) and its copolymers. Piezoelectric polymers are stretchable, flexible, chemically inert, inexpensive, and easily bulk processable into almost any desired geometry^{5–12}. A high-performance flexible piezoelectric polymer nanofiber device would constitute an enabling platform technology with transformative potential across applications currently inaccessible to ceramic-based systems, including strain-sensing smart textiles, piezoelectric vascular implants, and flexible electronic devices with embedded acoustic transducers. Nevertheless, piezoelectric polymers exhibit piezoelectric coefficients that are significantly inferior to those of their ceramic counterparts, and their integration into micrometer- and nanometer-scale device architectures remains technically challenging.

Considerable effort has therefore been expended over the years to improve the piezoelectric response of semi-crystalline PVDF through chemical modification and the incorporation of functional additives. However,

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one of the most promising approaches has been to manufacture PVDF nanofibers using the electrospinning process^{13–17}.

Electrospinning produces continuous nanofibers by applying an electric potential to a nozzle from which a polymer melt or solution is extruded. This electric potential acts as a driving force to stretch the polymer into a continuous fiber with a controllable diameter $\sim 10\ \mu\text{m}$ to $\sim 10\ \text{nm}$, through judicious control of processing parameters^{18,19}, as illustrated schematically in Fig. 1. The mechanical stretching of PVDF during the electrospinning process not only improves the polymer's crystallinity but also increases the relative volume fraction of the piezoelectric β phase. These two structural changes combine to greatly enhance the piezoelectric behavior of this nanostructured PVDF compared to the bulk, establishing electrospinning as a powerful and scalable route to high-performance piezoelectric polymer nanostructures.

While this interesting manufacturing process may allow for improved piezoelectric properties and a uniquely valuable form factor (long continuous nanofibers of piezoelectric material), assembling this nanostructured material into a useful piezoelectric device remains elusive. Although some authors report piezoelectric devices created by sandwiching a membrane/mat of randomly oriented electrospun fibers between two large metal electrodes^{20–22}, all such attempts have yielded unsatisfactory results: the random orientation of the fibers relative to the electrodes largely negates the material property improvement obtained by nanostructuring the PVDF. Indeed, the crystalline orientation of the β phase induced in PVDF electrospun fibers is such that the greatest piezoelectric polarization will be radially toward the center of the fiber²³. This implies that any attempt at placing electrodes on either side of a fiber or collection of fibers (such as a membrane/mat) is bound to yield disappointing results, as the piezoelectric polarization vectors within the fibers will largely cancel out between the electrodes.

Therefore, a major remaining obstacle to realizing a flexible piezoelectric polymer nanofiber device is the difficulty in integrating electrodes^{24,25} that can harness the nanostructural advantage of these electrospun semi-crystalline nanofibers.

Recent studies have highlighted piezoelectric nanofiber-based architectures as promising platforms for self-powered sensing and energy harvesting in wearable and structural applications^{26–29}. The incorporation of functional fillers such as carbon nanotubes (CNTs) and barium titanate (BTO) has been shown to enhance β -phase content, dielectric properties, and overall piezoelectric performance. Electrospun PVDF/BTO (barium titanate) nanofibrous membranes showed robust tactile-sensing performance, including long durability, a fast response time and high sensitivity across a 0–5 bar pressure range, while also enabling motion monitoring when attached to the body³⁰. In parallel, core-shell P(VDF-TrFE)/PEDOT: PSS (poly(3,4-ethylenedioxythiophene polystyrene sulfonate)) nanofibers improved piezoelectric output by tuning shell thickness, reaching about 8.76 V and 547 nA, with further gains when devices were connected in series or parallel, and they were also successfully used for motion detection^{31,32}. Extending this concept to structural materials, aligned core-shell nanofibers embedded in flexible epoxy laminates formed a distributed piezoelectric pixel network capable of identifying the exact location of mechanical stimuli³³. Together, these works show that piezoelectric nanofiber composites can

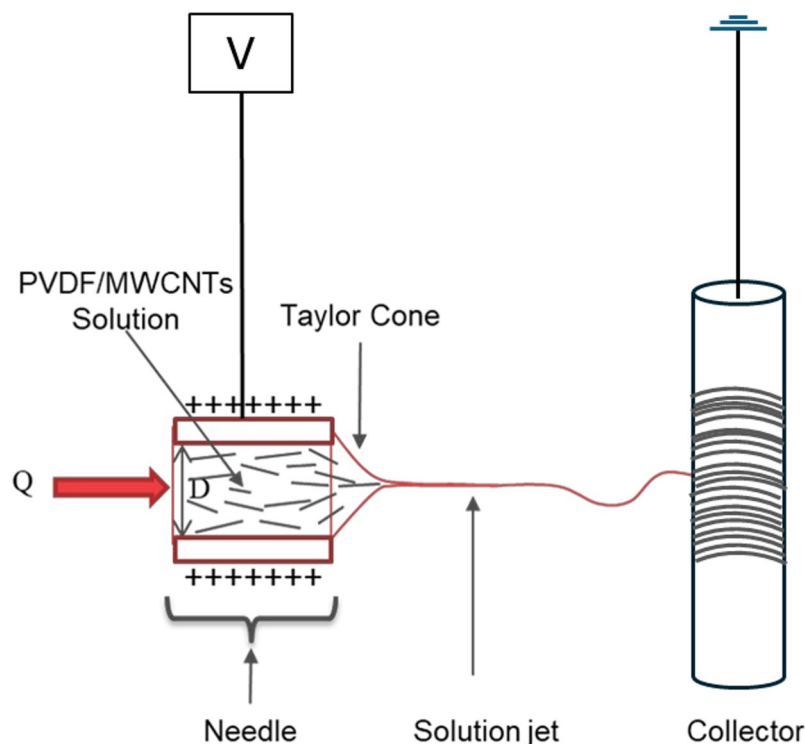


Fig. 1. Schematic showing a principle of solution electrospinning technique.

combine mechanical flexibility, self-powered operation, and spatially resolved sensing, making them attractive for wearable electronics, health monitoring, and robotic skin applications³⁴. Broader efforts in electrospun PVDF-based energy harvesting have further explored structural design and filler engineering as complementary strategies. Mokhtari et al. demonstrated that twisting MXene and silver nanoparticle-filled P(VDF-TrFE) nanofibers into a bioinspired coil structure yields a power density of 6.6 mW cm^{-3} at 50% strain with a conversion efficiency of 17%, illustrating how structural geometry can amplify material-level enhancements³⁵. Raman et al. showed that graphene nanoribbon-filled PVDF nanofibers achieve an open-circuit voltage of 83.3 V and a triboelectric power density of 6.37 W m^{-2} , driven by enhanced interfacial polarization and dielectric response³⁶. Sukumaran et al. further demonstrated that both conductive and insulating nanofillers promote β -phase formation through distinct mechanisms, with BN-filled PVDF reaching a triboelectric power density of $281 \text{ } \mu\text{W cm}^{-2}$, highlighting the importance of filler electrical character in governing charge retention³⁷.

Nevertheless, PVDF fibers obtained by electrospinning preferentially crystallize in the orthorhombic piezoelectric β phase with the [200] direction perpendicular to the length of the fiber. This implies that the piezoelectric polarization is most likely perpendicular to the length of the fiber. The ideal geometry for leveraging the piezoelectric effect therefore consists of a radially symmetric coaxial structure, in which an inner central electrode is enrobed by PVDF, which is in turn coated by an outer electrode, as shown in Fig. 2. This structure would then act as an arbitrarily long piezoelectric PVDF nanofiber device. Such a coaxial structure would constitute an arbitrarily scalable, continuous piezoelectric nanofiber device in which the electrode geometry is inherently matched to the electrical polarization direction of the active material.

In this manuscript, we report a novel strategy for realizing a long, continuous, flexible, and scalable piezoelectric nanofiber device by exploiting the interfacial tension between multi-walled carbon nanotubes (MWCNTs) and PVDF to drive the spontaneous self-assembly of a coaxial nanofiber architecture during a single-step electrospinning process. To the best of our knowledge, this represents the first demonstration of a coaxial piezoelectric nanofiber device fabricated entirely through self-assembly from a single precursor solution, without recourse to specialized coaxial nozzles or multi-step electrode integration processes.

Experimental section

Materials

Polyvinylidene fluoride (PVDF) powder (Arkema Kynar 741) was used as-received. Multi-wall carbon nanotubes (MWCNTs) with an average length of $1.5 \text{ } \mu\text{m}$, an average diameter of 9.5 nm and a specific surface area of $250\text{--}300 \text{ m}^2/\text{g}$ were also used as-received (Nanocyl product no. NC7000). These MWCNTs were selected for their metallic nature, which is essential for their function as the inner conductive core electrode. The solvent *n*, *n* dimethylformamide (DMF) and acetone were used as received.

Solution preparation and electrospinning process

0.6%wt. MWCNTs were dispersed in a 7:3 mixture (by weight) of DMF and acetone using a Qsonica Q700 ultrasonic probe to ensure homogeneous nanotube dispersion and to disrupt nanotube agglomerates. 14.4% wt. PVDF powder was gradually dissolved into this mixture while stirring at 40°C until a homogeneous solution was obtained.

This solution was loaded into a syringe and extruded through a hollow stainless steel needle with diameter of 0.361 mm (27 AWG) at a controlled flow rate of 1 mL/h using a syringe pump. An electrically grounded rotating cylindrical collector (diameter of 7.5 cm) with a rotation speed of 400 rpm was placed 20 cm from the needle to promote preferential fiber orientation along the collector rotation direction. A high voltage of 25 kV was applied between the metallic needle and the grounded collector using a high-voltage power supply (SL40*50, Spellman), corresponding to an applied electric field of $1.25 \times 10^5 \text{ V/m}$. The collector speed of 400 rpm was

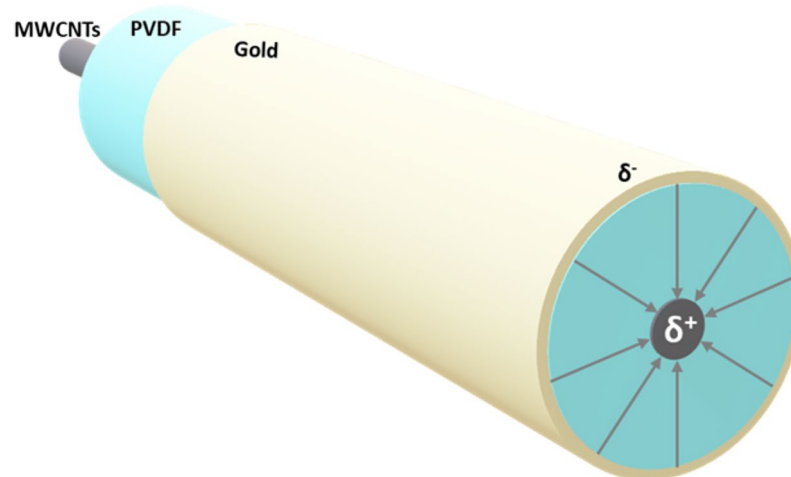


Fig. 2. Schematic representation of a PVDF nanofiber device, showing direction of piezoelectric polarization. The inner electrode consists of a MWCNT core and the outer electrode consists of a PVD gold thin-film.

deliberately selected to simultaneously achieve preferential fiber orientation and ensure compatibility with TEM grid collection, as higher speeds render reliable and reproducible fiber deposition on TEM grids impractical.

Device assembly and electrode integration

Outer electrode

A thin gold layer was deposited onto the top surface of the nanofiber membrane by physical vapor deposition (PVD) using a shadow mask to precisely define the active electrode area. PVD was selected for its ability to partially penetrate the porous fibrous network, maximizing electrical contact with the outer surface of the nanofibers while preserving their structural integrity. The shadow mask ensured that gold deposition was strictly confined to the defined surface area, preventing any overlap with the cross-sectional regions reserved for inner electrode contact.

Alternative conductive contact strategies, including aluminum foil, silver paste, or digitally patterned conductive ink, are equally viable for the outer electrode depending on the target application and fabrication constraints.

Inner electrode

Electrical contact to the conductive MWCNTs core was established at the cross-section of the nanofiber membrane, a region spatially distinct from the gold-coated surface, where the fiber cross-sections naturally expose the MWCNTs core. Silver paste was applied to this exposed cross-sectional area covered with conductive copper tape to form a reliable ohmic contact with the inner MWCNTs electrode. The spatial separation between the outer gold electrode (membrane surface) and the inner MWCNTs electrode (membrane cross-section) is the key design feature that prevents short-circuit between the two electrodes.

Protective encapsulation

Following electrode integration, a thin layer of polydimethylsiloxane (PDMS) was applied over the active area of the device. This encapsulation layer serves exclusively as a mechanical protective coating, shielding the fragile sub-100 nm diameter nanofibers from physical damage during repeated mechanical stimulation. PDMS was selected for this purpose due to its high flexibility, electrical insulating properties, and chemical inertness with respect to both PVDF and the electrode materials. The PDMS layer lies outside the electrode measurement path and does not contribute to the piezoelectric voltage output.

It should be noted that PDMS encapsulation was selectively applied exclusively during direct piezoelectric effect measurements, where repeated mechanical stimulation poses a risk of physical damage to the nanofiber network. No encapsulation was required for the inverse piezoelectric characterization, as laser Doppler vibrometry measurements are entirely contactless and impose no mechanical load on the membrane.

Structural characterization and discussion

The morphology of the electrospun PVDF/MWCNTs nanofibers was characterized using a Hitachi SU-8230 field emission scanning electron microscope (FEG-SEM). Representative images are presented in Fig. 3. The higher magnifications (10k x and 25k x) were deliberately chosen to enable accurate fiber diameter measurement and reliable detection of fiber edges, a necessity given the sub-100 nm fiber dimension. The SEM images reveal a uniform nanofiber morphology characterized by smooth, continuous fibers free of beads or defects; indicative of well-optimized processing parameters. Fiber diameter analysis was performed on 100 individual fibers using ImageJ image analysis software, yielding a mean diameter of 80 ($\pm$ 13 nm), with the narrow distribution confirming the reproducibility and consistency of the fabrication process.

The internal nanostructure of the electrospun fibers was examined using a Thermo Scientific Talos F200C transmission electron microscope (TEM) operated at an accelerating voltage of 200 kV. Representative TEM images are shown in Fig. 4, unambiguously revealing that the MWCNTs constitute the core of the nanofiber and are completely enrobed by a continuous PVDF shell, confirming the formation of a well-defined coaxial nanostructure.

This spontaneous core-shell self-assembly is thermodynamically driven by the marked difference in surface tension between the two constituents; PVDF exhibits a relatively low surface tension of 30.3 mN m⁻¹^{38,39}, whereas MWCNTs present a significantly higher surface tension of 45.3 mN m⁻¹⁴⁰. In accordance with the principle of interfacial energy minimization, the lower surface tension PVDF spontaneously migrates to the fiber surface during the electrospinning jet elongation process, while the higher surface tension MWCNTs are driven toward and concentrated along the fiber core. This thermodynamic driving force provides a robust and reproducible self-assembly mechanism that reliably yields the coaxial architecture required for optimal piezoelectric device performance, without recourse to specialized coaxial nozzles or multi-step fabrication procedures.

To further support the coaxial architecture evidenced by TEM imaging, electrical conductivity measurements were performed using a two-point method following ASTM D4496 over a gauge length of 15 mm. Surface and core conductivities were measured independently on three samples each fabricated under identical conditions. Direct probe contact on the fiber mat surface yielded a consistent conductivity of 10⁻¹⁵ S/cm across all three samples, confirming the insulating character of the PVDF shell. Measurements performed on cryogenically fractured cross-sections via silver paste contacts revealed a core conductivity of 10⁻⁴ S/cm, equally consistent across all three samples, confirming the presence of a continuous and electrically percolating MWCNT network along the full fiber length. The identical values obtained across independent samples speak to the remarkable reproducibility of the self-assembly process. The eleven-orders-of-magnitude contrast between surface and core conductivity values provides compelling independent evidence of the coaxial structure, while the moderate core conductivity confirms the optimal MWCNT loading; excessive conductivity was found to induce short-circuit conditions destabilizing the electrospinning process.

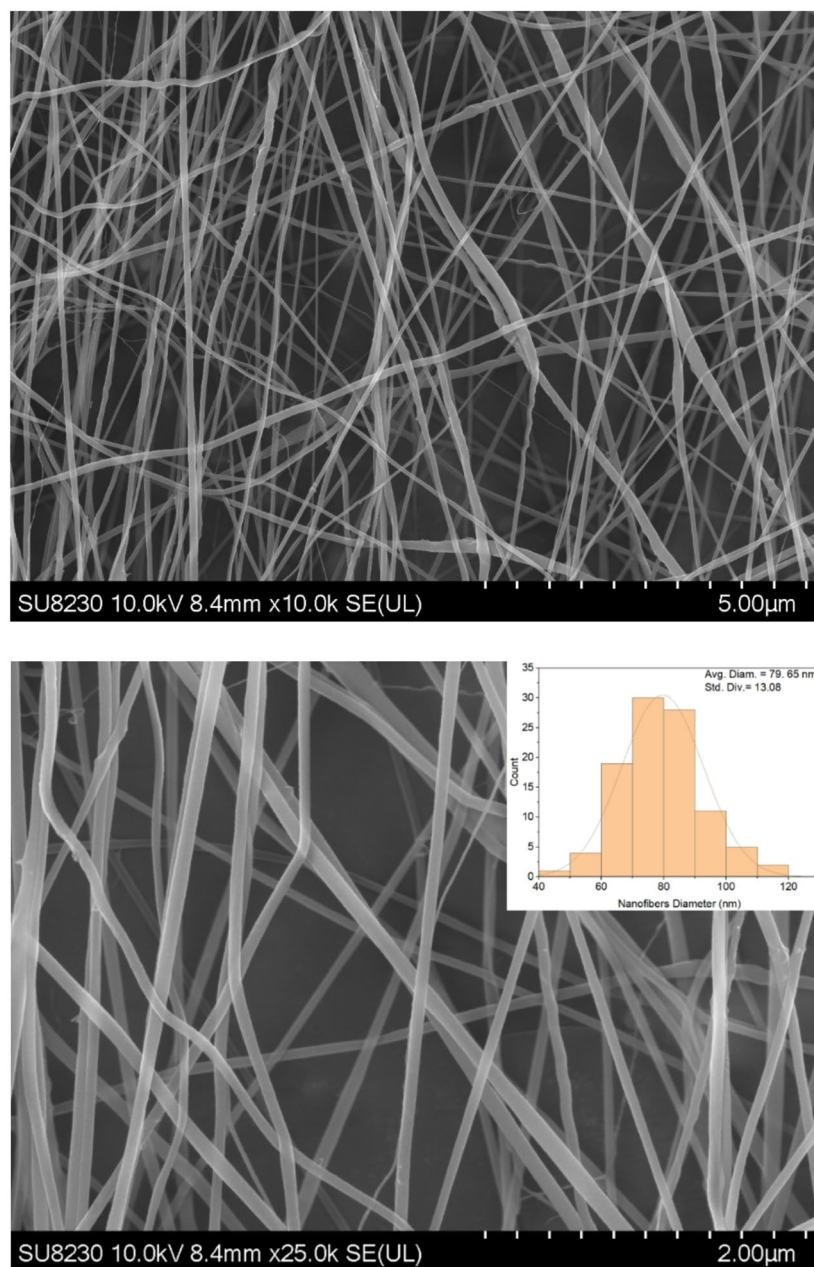


Fig. 3. Representative SEM image of PVDF/MWCNTs nanofibers produced by electrospinning process.

Having characterized the geometry and structure of the nanofibers with SEM and TEM, X-ray diffraction (XRD) was further employed to study the crystalline structure of PVDF. A Panalytical X'pert Pro MPD X-ray diffractometer using Cu-K α radiation $\lambda = 1.54 \text{ \AA}$ was used at room temperature. Figure 5 shows a representative X-ray diffractogram of as-received PVDF virgin powder and electrospun PVDF/MWCNTs nanofibers. All diffracted peaks were fitted to Voigt functions, and the relative volume fractions of crystalline phases were quantified using structure factor calculations (Table 1).

All diffraction peaks were fitted using Voigt functions after background subtraction, and phase fractions were calculated from the integrated peak areas corrected by the corresponding structure factors. The α phase of PVDF exhibits characteristic reflections at $2\theta \approx 17.6^\circ$ (100), 18.3° (020), and 19.9° (110), consistent with its monoclinic crystal structure^{41,42}. In contrast, the electroactive β phase (orthorhombic)⁴⁰ is identified by the strong reflection at $2\theta \approx 20.6^\circ$ corresponding to the overlapping (110)/(200) planes. The γ phase, which would appear near $2\theta \approx 20.2^\circ$ and 18.5° , is not observed within the resolution of the present measurements, indicating that its contribution is negligible.

Quantitative phase analysis shows that the as-received PVDF powder is predominantly composed of the α phase, with a total crystallinity of approximately 40%. However, the coaxial PVDF/MWCNTs nanofibers have greater PVDF crystallinity (52%), in which the piezoelectric β phase dominates (53% of the crystalline content).

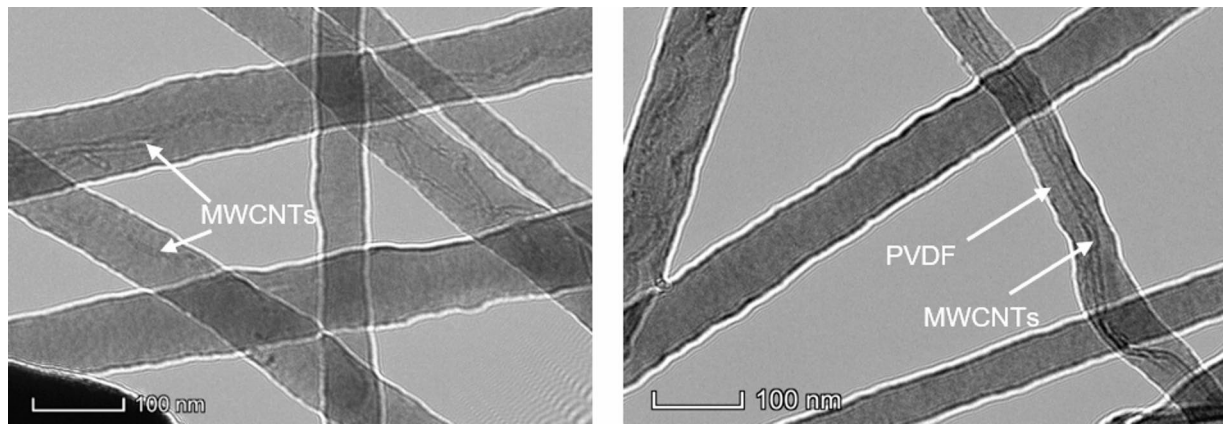


Fig. 4. Representative TEM image of nanofibers obtained by electrospinning technique showing a core of MWCNT (identified with arrows) enrobed by PVDF.

The net effect is that the amount of β phase increased approximately five-fold as a result of the electrospinning process. These results are in-line with those obtained in the past using similar electrospinning process parameters in the absence of MWCNTs²³; this suggests that the incorporation of MWCNTs does not have a measurable effect on the crystalline structure of PVDF. It is worth noting the absence of a peak at $2\theta = 35.04^\circ$ for the nanofibers in Fig. 5, which would correspond to the β phase (001) plane. This indicates that the crystallographic direction [001] runs along the length of the fiber axis, thereby confirming that the molecular chains of the β phase are oriented along the length of the fiber axis^{23,44–46}. The strong (200) peak confirms that the [200] direction is therefore perpendicular to the fiber length, consistent with a piezoelectric dipole moment (polarization vector) that is radially symmetric about the center of the fiber, as hypothesized in Fig. 2.

Piezoelectric behavior

A co-axial nanofiber mat-based piezoelectric device was assembled and characterized to obtain representative device-level performance, as illustrated in Fig. 6. In this assembly, a 15 mm width of parallel nanofibers were cross-sectioned with a scalpel to a length of 15 mm. The exposed ends were then painted with silver paste to electrically connect to all the embedded MWCNT inner electrodes simultaneously. The gold outer electrodes deposited on the membrane surface by PVD naturally formed a continuous electrical contact across the fiber network. Copper wire probes were connected to the inner and outer electrode contacts to enable voltage measurement or application across the coaxial nanofiber assembly. A schematic of this device configuration is presented in Fig. 6.

Direct piezoelectric effect (sensor mode)

The direct piezoelectric effect, in which mechanical stress is converted into an electric field, is governed by the constitutive relation:

$$E_i = g_{iJ} \sigma_J \quad (i = \{1, 2, 3\}; J = \{1, 2, \dots, 6\}) \quad (1)$$

Where E_i is the electric field generated in the i direction, σ_J is the applied mechanical stress in the J direction, and g_{iJ} is the piezoelectric voltage coefficient.

Prior to mechanical characterization, a thin PDMS encapsulation layer was applied over the active device area to protect the sub-100 nm diameter nanofibers from physical damage during repeated mechanical loading cycles. As PDMS is electrically insulating (resistivity $> 10^{15} \Omega \cdot \text{cm}$) and lies outside the electrode measurement path, its presence does not influence the piezoelectric voltage output.

A cyclic uniaxial compressive stress of 31 kPa at a frequency of 2 Hz was uniformly applied over the 15 mm $\times$ 15 mm active device area using a vibration exciter K2007E01. A Keysight DSOX3014T oscilloscope was used to record the output voltage generated between inner and outer electrodes of the nanofiber assembly (Fig. 6).

The direct piezoelectric response of the device is presented in Fig. 7. Under an applied compressive stress of 31 kPa, the device generates a peak electric field of up to $6.9 \times 10^7 \text{ V m}^{-1}$, a value that reflects both the intrinsic piezoelectric properties of the electrospun β -phase PVDF and the efficacy of the coaxial electrode architecture in harnessing the radially oriented piezoelectric polarization. The negative values of the electric field correspond to the relaxation of the fibers upon removing the applied mechanical stress (spring-back), generating a reverse polarization; a hallmark of the direct piezoelectric effect under cyclic loading that confirms the electromechanical coupling is genuine and reversible.

Using Eq. 1, the effective piezoelectric voltage coefficient (g_{eff}) of the flexible device was found to be $g_{\text{eff}} = 0.22 \text{ V.m.N}^{-1}$. This value is particularly significant given that no electrical pre-poling was performed prior to characterization; a processing step that is typically required to align piezoelectric domains and maximize the piezoelectric response in bulk PVDF. The fact that $g_{\text{eff}} = 0.22 \text{ V.m.N}^{-1}$ is achieved in the as-fabricated, unpoled

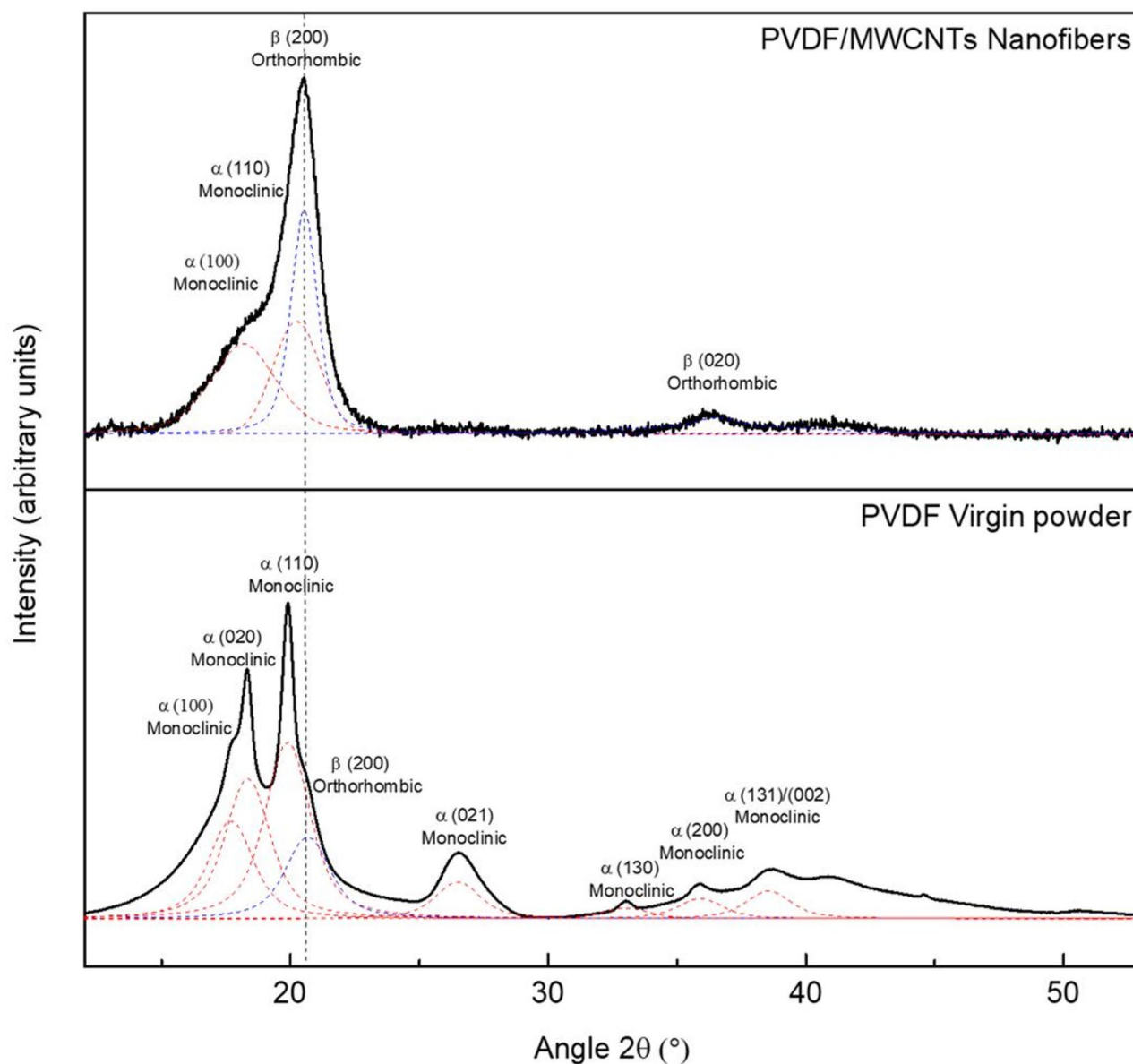


Fig. 5. Representative X-ray diffractogram of electrospun PVDF/MWCNTs nanofibers compared to PVDF virgin powder.

	PVDF Virgin Powder	MWCNTs/PVDF nanofibers
Fiber diameter (nm)	n/a	80
Degree of crystallinity (%)	40	52
Crystalline volume fraction % α phase (not piezoelectric)	89	47
Crystalline volume fraction % β phase (piezoelectric)	11	53

Table 1. Diameter, degree of crystallinity, and phase volume fractions in PVDF virgin powder and nanofibers.

state compares favorably with values reported in the literature for high-quality, fully poled bulk PVDF films ($g_{33} = 0.14 \sim 0.30 \text{ V.m.N}^{-1}$)^{7,47},

These results are particularly noteworthy given that no electrical pre-poling was performed; a processing step conventionally considered necessary to maximize the piezoelectric response in bulk PVDF. The fact that the self-assembled coaxial nanofiber architecture alone is sufficient to yield competitive piezoelectric performance in the as-fabricated state highlights the elegance and practical appeal of the approach reported in this work, and opens the door to simpler, more scalable manufacturing routes for flexible piezoelectric devices.

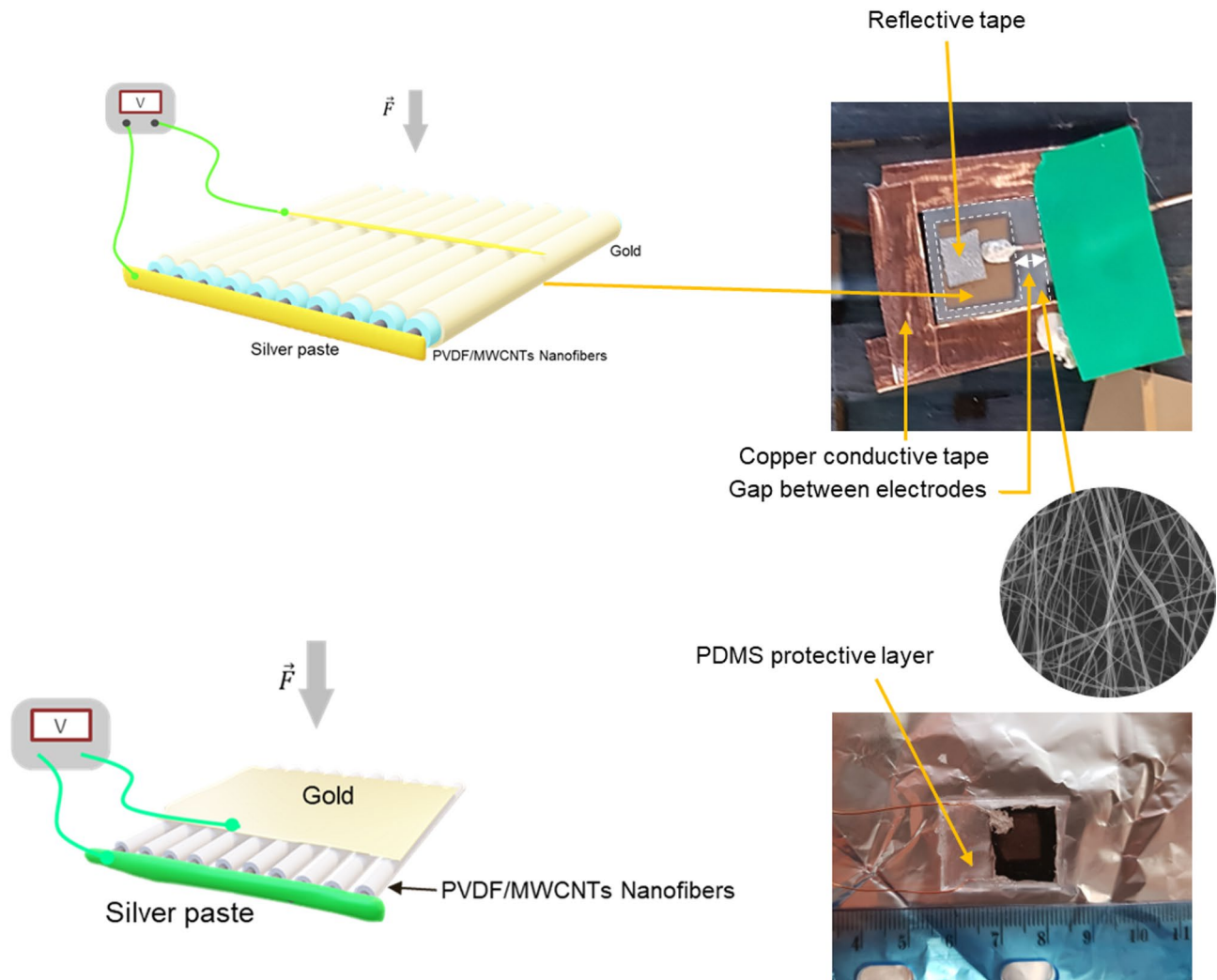


Fig. 6. Schematic showing how the direct piezoelectric effect was measured by applying a mechanical load on the piezoelectric nanofibers.

Inverse piezoelectric effect (actuator mode)

The inverse piezoelectric effect, in which an applied electric field induces a mechanical strain, is governed by the constitutive relation:

$$\epsilon_J = d_{Ji} E_i \quad (i = \{1, 2, 3\}; J = \{1, 2, \dots, 6\}) \quad (2)$$

Where ϵ_J is the strain along J , E_i is the applied electric field along i , and d_{Ji} is the piezoelectric coefficient.

The inverse piezoelectric response of the coaxial PVDF/MWCNTs nanofiber device was characterized using laser scanning vibrometry (LSV), which enables direct measurement of the mechanical displacement of the nanofiber membrane under electrical excitation. Measurements were performed using a Polytec GmbH (Germany) OFV-505 laser vibrometer connected to an OFV-2570 controller, with the laser beam oriented perpendicular to the fiber axis. An electrical signal with maximum amplitude 4.6×10^6 V/m at a frequency of 35 kHz was applied to excite the piezoelectric device. This frequency was selected on the basis of preliminary characterization as the frequency yielding the most stable and reproducible piezoelectric actuation response under the measurement conditions employed.

The applied electric field and the resulting mechanical strain are presented in Fig. 8. The mechanical displacement of the nanofiber membrane closely follows the form of the excitation signal, confirming a clear and unambiguous electromechanical coupling consistent with the inverse piezoelectric effect. A phase lag of approximately 0.3×10^{-4} s is observed between the excitation signal and the induced mechanical strain; this is attributable to the viscoelastic nature of the PVDF polymer matrix and the inertial response of the nanofiber membrane under dynamic excitation at 35 kHz.

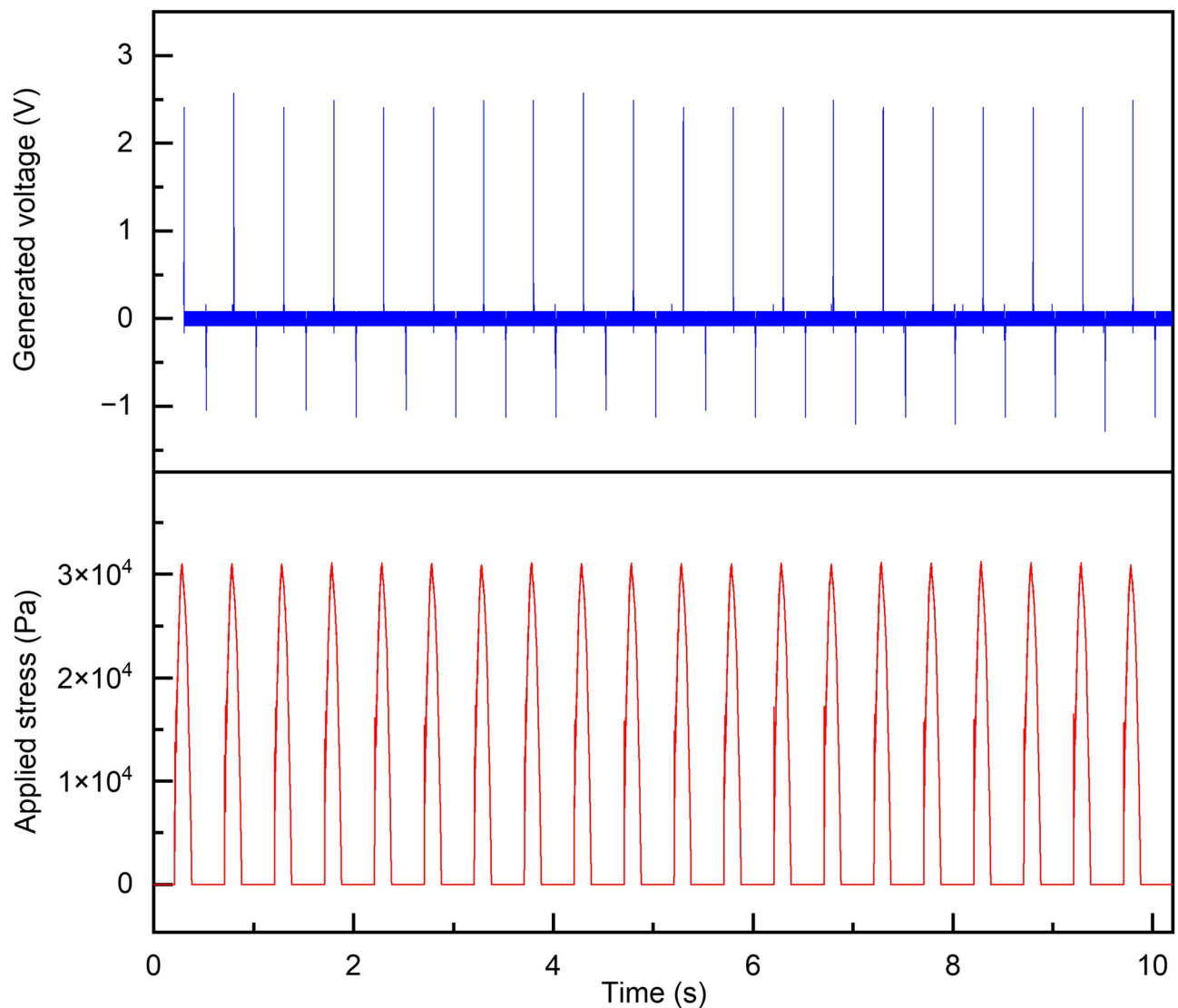


Fig. 7. Applied mechanical stress on the nanofiber assembly and the generated electric field within the nanofibers.

The effective piezoelectric coefficient (d_{eff}) of the flexible device was estimated using Eq. (2) to be $d_{\text{eff}} = 2.8$ pC/N. As with the direct piezoelectric characterization, it is important to emphasize that this value was obtained in the as-fabricated, without any electrical pre-poling treatment.

While this value is moderately lower than the quasi-static d_{33} values of $d_{33} = 10 \sim 30$ pC/N reported in the literature for high-quality pure, fully poled, bulk PVDF films^{9,11}, the comparison must be interpreted in its proper context: the literature values represent the upper bound of performance achievable in optimally processed and poled bulk material, whereas $d_{\text{eff}} = 2.8$ pC N⁻¹ is obtained under dynamic excitation of the transducer. The frequency-dependent reduction in apparent piezoelectric coefficient relative to quasi-static values is a well-established phenomenon in viscoelastic piezoelectric polymers and is consistent with the observed phase lag.

Conclusions

A flexible piezoelectric PVDF polymer nanofiber transducer with integrated electrodes was manufactured using the electrospinning process. The self-assembly of MWCNTs forms a continuous inner electrode at the core of the 80 nm diameter fibers, thereby leveraging the PVDF nanostructure. This single-step electrospinning process is easily scalable and can in principle be used to readily manufacture nanofiber devices of arbitrary length. Upon integrating of an outer electrode, the completed piezoelectric device was shown to operate reversibly as a sensor (direct effect) and actuator (inverse effect), yielding competitive piezoelectric coefficients in the as-fabricated state. The easily obtained nanostructure allows for a piezoelectric transducer with great sensitivity to be mass-produced. Unlike traditional, bulk, piezoelectric devices, the often difficult pre-poling was found to be completely unnecessary.

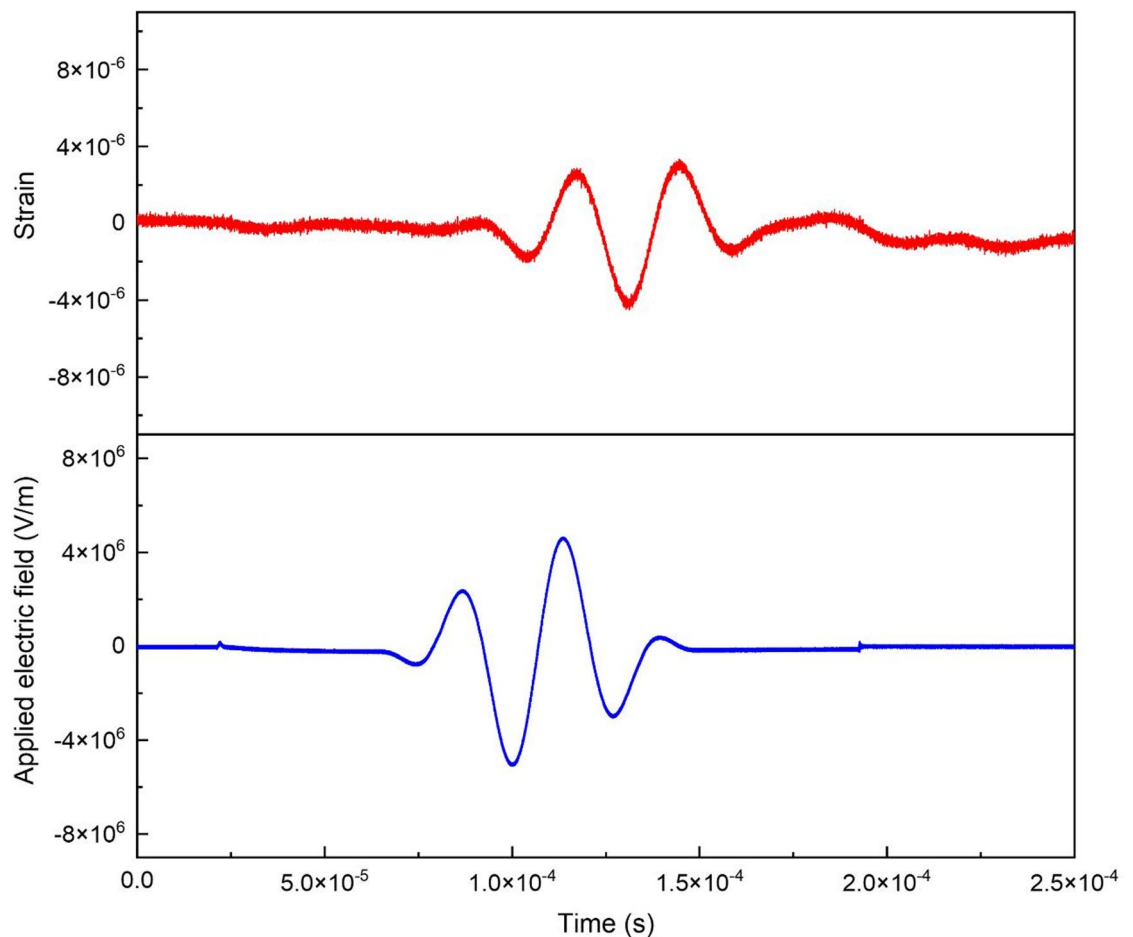


Fig. 8. Electric field applied across PVDF nanofibers and the resulting mechanical strain.

The thus obtained nanofibers are ten times thinner than spider silk ($\sim 1 \mu\text{m}$ diameter). The biocompatibility, chemical inertness, and mechanical flexibility that PVDF is known for, combined with the unusually long coaxial nanofiber form factor, suggests a wide range of exciting novel applications. In addition, multiple fibers/strands could be braided together to create a thicker “cord” consisting of multiple, independent transducers or else woven together into a “smart membrane”. For example, with the appropriate choice of outer electrodes, these nanofiber transducers could be integrated into smart textiles, form artificial skin or muscles, be embedded into aerospace composites, or incorporated into biomedical implants. In addition, given that PVDF is known to have ferroelectric and pyroelectric properties, non-volatile memory or temperature-sensing applications can also be envisioned.

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Received: 20 October 2025; Accepted: 16 June 2026

Published online: 20 June 2026

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Author contributions

Conceptualization, CMA, NRD and RJZ; methodology, CMA; validation, NRD and RJZ; formal analysis, CMA; investigation, resources, NRD and RJZ; data curation, CMA; writing—original draft preparation, CMA; writing—review and editing, NRD and RJZ; supervision, NRD and RJZ; funding acquisition, RJZ. All authors have read and agreed to the published version of the manuscript.

Declarations

Competing interests

The authors declare no competing interests.

Additional information

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