

# One-step and Continuous Fabrication of Coaxial Piezoelectric Fiber for Sensing Application

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## Electronic Supplementary Information

**Abstract** Although there has been rapid advancement in piezoelectric sensors, challenges still remain in developing wearable piezoelectric sensors by a one-step, continuous and environmentally friendly method. In this work, a 1D flexible coaxial piezoelectric fiber was directly fabricated by melt extrusion molding, whose core and sheath layer are respectively slender steel wire (*i.e.*, electrode) and PVDF (*i.e.*, piezoelectric layer). Moreover, such 1D flexible coaxial piezoelectric fiber possesses short response time and high sensitivity, which can be used as a self-powered sensor for bending and vibration sensing. More interestingly, such 1D flexible coaxial piezoelectric fiber (1D-PFs) can be further endowed with 3D helical structure. Moreover, a wearable and washable motion monitoring system can be constructed *via* braiding such 3D helical piezoelectric fiber (3D-PF) into commercial textiles. This work paves a new way for developing 1D and 3D piezoelectric fibers through a one-step, continuous and environmentally friendly method, showing potential applications in the field of sensing and wearable electronics.

**Keywords** Coaxial piezoelectric fiber; Melt extrusion molding; Bending and vibration sensing; Motion monitoring system

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## INTRODUCTION

In recent years, with the increasing prosperity of wearable industry, wearable electronics have greatly improved people's quality of life and have become an indispensable tool.<sup>[1–5]</sup> According to the sensing mechanism, wearable sensors are generally divided into following categories: *e.g.*, capacitance,<sup>[6,7]</sup> resistance,<sup>[8]</sup> triboelectricity<sup>[9,10]</sup> and piezoelectricity.<sup>[11]</sup> Among them, piezoelectric sensors have drawn increasing attention because of their self-power supply, easy fabrication and simple structure.<sup>[12,13]</sup> As well known, piezoelectric fiber can be easily integrated with textiles for convenient wearing owing to its light weight, high flexibility and continuous preparation compared with planar electronic devices, and have promising applications in ideal wearable electronics.<sup>[14,15]</sup>

At present, piezoelectric fiber has been rapidly developed<sup>[16]</sup> and various methods have been proposed to prepare piezoelectric fiber, such as wet spinning,<sup>[17,18]</sup> melt spinning<sup>[19,20]</sup> and electrospinning.<sup>[21,22]</sup> However, their electrodes were often obtained by additional procedures (*e.g.*, chemical deposition,<sup>[1,23]</sup> electroplating,<sup>[24]</sup> and so on), which are neither efficient nor environmentally friendly. Of note, aforementioned procedures often lead to poor stability of

piezoelectric fiber under extreme conditions due to the weak adhesion between piezoelectric fiber and electrode,<sup>[25]</sup> substantially hindering the broad-range applications of piezoelectric fiber. Therefore, developing a one-step, continuous and environmentally friendly method to prepare piezoelectric fiber is significant and urgent.

As well known, melt extrusion molding is one of the most important polymer melt processing methods owing to its facile, solvent-free, low-cost, continuous and large-scale features.<sup>[26–28]</sup> In addition, as a commercial and low-cost thermoplastic, poly(vinylidene fluoride) (PVDF) is widely used as a raw material for fabricated piezoelectric sensors due to its excellent piezoelectricity, ease of melt processing, inherent flexibility and good chemical stability.<sup>[29,30]</sup> In view of these, it will be of great significance if PVDF piezoelectric fiber can be easily fabricated *via* polymer melt extrusion molding.

In this study, a 1D flexible coaxial piezoelectric fiber (1D-PFs) was continuously fabricated by melt extrusion molding, whose core and sheath layer are respectively slender steel wire (*i.e.*, electrode) and PVDF (*i.e.*, piezoelectric layer). In view of this, the electrode can be subtly protected by piezoelectric layer from environmental damage. Moreover, such 1D piezoelectric fiber can realize bending and vibration sensing because of its short response time and high sensitivity. It is worth noting that the as-fabricated 1D-PFs can be further endowed with 3D helical structure by feasible heat setting and braided into commercial textiles as a wearable and washable

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motion monitoring system. This work proposes a one-step, continuous and environmentally friendly method to fabricate 1D and 3D piezoelectric fibers, promoting the potential applications of 1D and 3D piezoelectric fibers in the field of sensing and wearable electronics.

## EXPERIMENTAL

### Materials

Commercial PVDF (201,  $M_w=1.26 \times 10^5$  g/mol) was purchased from Arkema, France. Slender steel wire with a diameter of 0.08 mm was purchased from Sichuan Yutong Stainless Steel Co., Ltd. Mini single-screw extruder was purchased from Zhengzhou Craftsman Machinery Equipment Co., Ltd.

### Preparation of 1D Flexible Coaxial Piezoelectric Fibers (1D-PFs)

Fig. 1(a) schematically shows the preparation process of 1D flexible coaxial piezoelectric fiber. First, the cleaned slender steel wire is pierced from the die and fixed on the collecting roller. Then the dried PVDF pellets were melt extruded by a mini single-screw extruder with a screw speed of 10 r/min at 200 °C, and the extrudate was drawn at different line speeds by a collecting roller. Finally, 1D flexible coaxial piezoelectric fibers with different diameters were obtained and further assembled into a single-electrode bending and vibration sensors. For convenience, the as-fabricated 1D flexible coaxial piezoelectric fiber were labeled as 1D-PFs, where “s” represents draw ratio. For example, 1D-PF1 represents the

sample fabricated at a draw ratio of 1.

### Characterization

A field-emission scanning electron microscope (ZEISS Gemini 300) was employed to observe the morphology of 1D-PFs. Before SEM observation, the specimens were sputtered with a thin layer of gold for better imaging. In addition, a home-made test system (Fig. S1 in the electronic supplementary information, ESI) was designed to test the electrical output performance of 1D piezoelectric fibers under different dynamically cyclic pressures. Such test system includes electrometer (Keithley-6514) and voice coil motor (IVCL38231). Moreover,  $\beta$  crystal content of 1D-PFs was investigated by Fourier transform infrared (FTIR) spectroscopy. FTIR measurement was performed at 32 scans from 400  $\text{cm}^{-1}$  to 4000  $\text{cm}^{-1}$  at a resolution of 4  $\text{cm}^{-1}$ .

### Preparation of 3D Helical Piezoelectric Fiber (3D-PF) Used for Wearable Electronics

The detailed preparation process of 3D-PF is described in Fig. S2 (in ESI). 5-cm-long 3D-PF (it was selected unless otherwise specified) was braided into a 10 cm  $\times$  8 cm textile (3D-PF/textile). Moreover, 3D-PF are respectively braided into the commercial knitted glove, sweater as well as kneepad and worn on the corresponding parts of the body, which can be used as a wearable self-powered motion monitoring system (SP-MMS) to monitor the motions of the small joints (*i.e.*, index finger and wrist) and large joints (*i.e.*, elbow and knee).

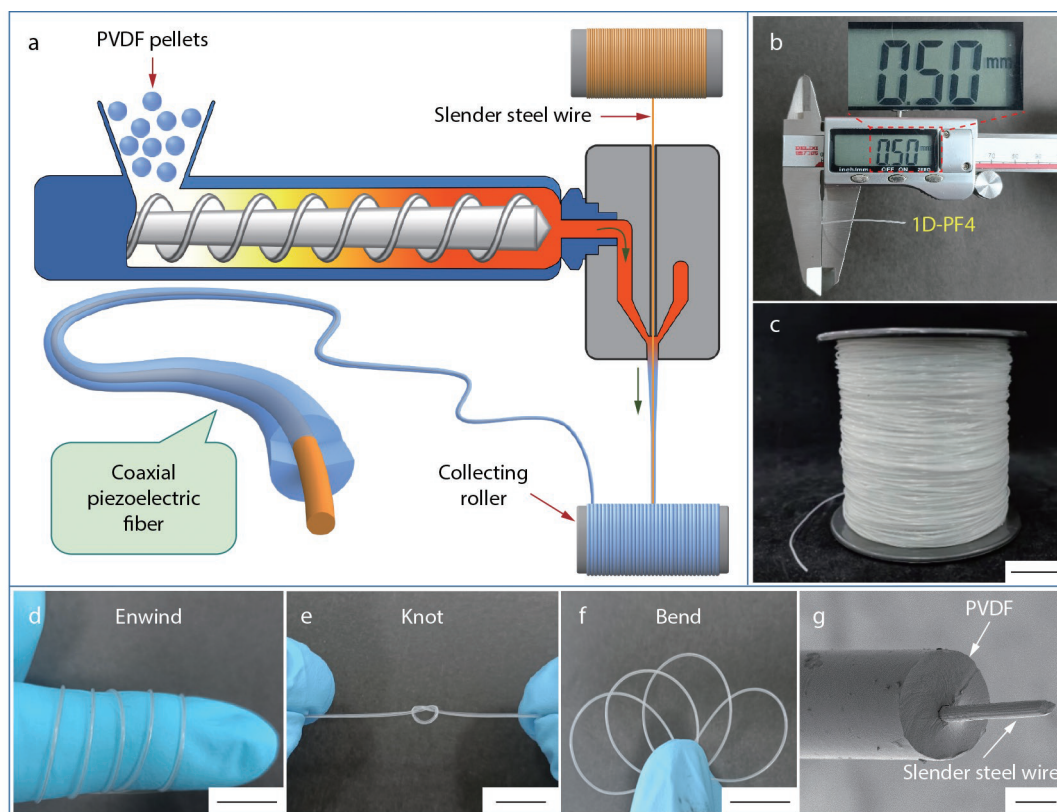


**Fig. 1** (a) Schematic diagram for the preparation process of 1D-PFs; (b) Measured diameter of 1D-PF4; (c) Optical image showing the 80-m-long 1D-PF4 collected on a roller (scale bar: 1 cm); Optical images of 1D-PF4 at (d) unwinding, (e) knotting, and (f) bending states (scale bar: 1 cm); (g) SEM image (the view of tilted angle of 45°) of 1D-PF4 (scale bar: 200  $\mu\text{m}$ ).

## RESULTS AND DISCUSSION

### Architecture of 1D Flexible Coaxial Piezoelectric Fibers (1D-PFs)

As shown in Fig. 1(b), the measured diameter of 1D flexible coaxial piezoelectric fiber with draw ratio of 4 is  $\sim 0.5$  mm, which is thinner compared with other reported coaxial piezoelectric fibers.<sup>[31–33]</sup> Moreover, the optical image in Fig. 1(c) demonstrates that the as-prepared 80-m-long 1D-PF4 can be easily unwound on a roller. It is worth noting that as long as the slender steel wire is long enough, longer 1D flexible coaxial piezoelectric fiber can be continuously fabricated *via* the method proposed in this study. More interestingly, such coaxial piezoelectric fiber with excellent flexibility can withstand complex mechanical deformations such as enwinding (Fig. 1d), knotting (Fig. 1e), and bending (Fig. 1f), implying its promising applications in wearable electronics. Moreover, the SEM image in Fig. 1(g) illustrates that such flexible coaxial piezoelectric fiber contains slender steel wire (thickness:  $\sim 80$   $\mu\text{m}$ ) inside and PVDF piezoelectric polymer (thickness:  $\sim 210$   $\mu\text{m}$ ) outside. It is worth noting that diameter and the PVDF volume in 1D-PFs can be easily controlled by adjusting the line speed of the collecting

roller, and various 1D-PFs prepared at different draw ratios are shown in Figs. S3 and S4 (in ESI). In a word, 1D flexible coaxial piezoelectric fibers can be one-step but continuously fabricated by melt extrusion molding, suggesting the scalable but controllable preparation process.

### Piezoelectric Output Performance of 1D Flexible Coaxial Piezoelectric Fibers (1D-PFs)

Fig. 2(a) shows the schematic diagram of 1D-PFs in bending mode, and output voltage of 1D-PFs was explored under the same conditions. Obviously, according to Fig. 2(b), output voltage of 1D-PFs with different draw ratios first increases and then decreases with the further increase of draw ratio. It is worth noting that 1D-PFs with the draw ratio of 3 exhibits a maximum output voltage (125 mV), and the trend of output current is consistent with output voltage (Fig. S5 in ESI). It is well known that high  $\beta$  crystal content in PVDF can enhance piezoelectric output.<sup>[34–36]</sup> Although  $\beta$  crystal content of 1D-PFs is proportional to the draw ratio (Fig. S6 in ESI), PVDF volume in 1D-PFs is inversely proportional to the draw ratio (Fig. S4 in ESI). Thus, it is understandable that 1D-PFs with the draw ratio of 3 has the optimal output voltage because of the trade-off

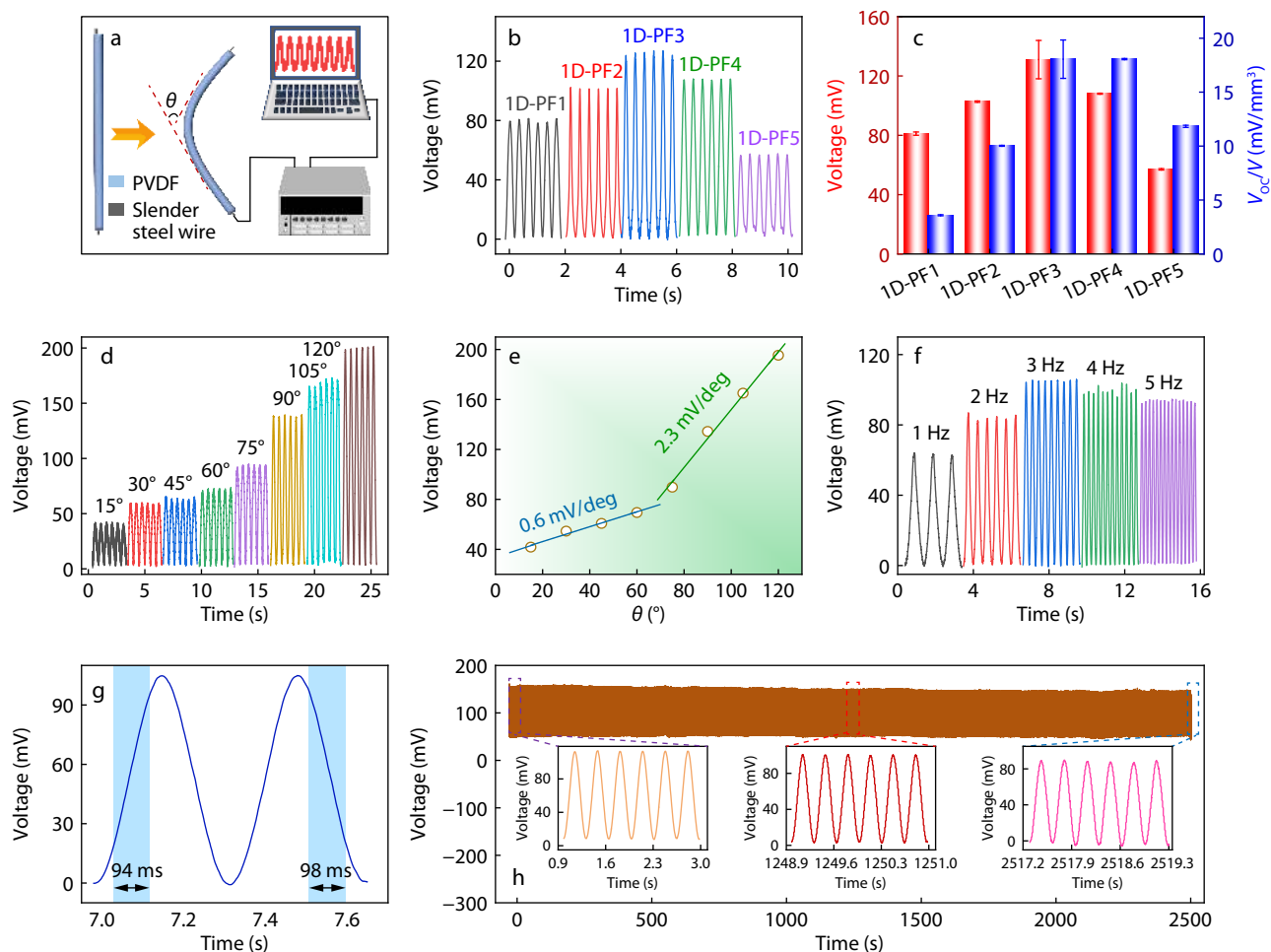


**Fig. 2** (a) Schematic diagram of 1D-PFs in bending mode; (b) Output voltage of 3-cm-long 1D-PFs at 3 Hz (bending angle from 0° to 60°); (c) Output voltage and output voltage density of 1D-PFs; (d) Output voltage of 1D-PF4 under different bending angle at 2 Hz; (e) The relationship between output voltage of 1D-PF4 and bending angle; (f) Output voltage of 1D-PF4 at different frequencies (bending angle from 0° to 60°); (g) Response and recovery time of 1D-PF4 at 3 Hz; (h) Durability of 1D-PF4 over 7500 cycles at 3 Hz (bending angle from 0° to 60°).

between  $\beta$  crystal content and PVDF volume ( $V$ ). Of note, as clearly shown in Fig. S4 in ESI, 1D-PFs fabricated at different draw ratios have different diameters, therefore the measured output voltage ( $V_{OC}$ ) cannot be directly employed as a standard to evaluate the piezoelectric output performance of 1D-PFs. In view of this, voltage density (*i.e.*, output voltage per unit volume,  $V_{OC}/V$ ) is calculated and summarized in Fig. 2(c), which is employed to directly evaluate the piezoelectric output performance of 1D-PFs fabricated at different draw ratios. Clearly, the voltage density of 1D-PFs exhibited a monotonically increasing trend with increasing draw ratio except for 1D-PF5. This should be ascribed to the fact that high draw ratio of 1D-PF5 leads to a much smaller PVDF melt volume than 1D-PFs prepared at other draw ratios, finally resulting in a smaller voltage density of 1D-PF5. Therefore, considering the higher voltage density and thinner diameter, 1D-PF4 was selected for the following section unless otherwise stated.

As illustrated in Figs. 2(d) and 2(e), the output voltage of 1D flexible coaxial piezoelectric fiber generally exhibits a monotonically increasing trend with the increasing bending angle. Herein, the sensitivity in bending mode can be defined:  $S = \Delta V / \Delta \theta$ ,<sup>[37]</sup> where  $\Delta V$  is the relative change in output voltage, and  $\Delta \theta$  is the relative change in bending angle. Obviously, in the small bending angle range ( $<60^\circ$ ), its sensitivity is 0.6 mV/deg, whereas a higher sensitivity of 2.3 mV/deg is observed in the large bending angle range ( $60^\circ$ – $120^\circ$ ). Moreover, to further understand the bend-induced response behavior, simulation analysis was carried out to theoretically explore the stress distribution and potential distribution in bending mode. As clearly shown in Fig. S7 (in ESI), when the 1D flexible coaxial piezoelectric fiber is bending under the external force, stress distribution is concentrated in the maxim-

um deformation region, resulting in a higher potential in the region of stress concentration.<sup>[32]</sup> In addition, output voltage of 1D flexible coaxial piezoelectric fiber generally increases with the increase of frequency. However, further increasing frequency will lead to the decreasing output voltage, and the maximum output voltage ( $\sim 104$  mV) occur at the frequency of 3 Hz (Fig. 2f). As well known, response and recovery time is a crucial parameter for sensors.<sup>[38]</sup> Herein, response time (*i.e.*, the rise time of voltage increasing from 10 % to 90 %) and recovery time (*i.e.*, the fall time of voltage decreasing from 90% to 10%) of 1D flexible coaxial piezoelectric fiber were respectively calculated as 94 and 98 ms (Fig. 2g), demonstrating rapid response and recovery when subjected to bending. Moreover, the change of response and recovery time of 1D-PF4 with test frequency is shown in Fig. S8 (in ESI). For the practical application of 1D flexible coaxial piezoelectric fiber, stability is one of the most important issues. As shown in Fig. 2(h), it is clear that even after 7500 cycles of bending, output voltage can maintain favorable repeatability and no obvious fluctuation is observed, which indicates that such 1D flexible coaxial piezoelectric fiber has excellent stability.

### 1D Flexible Coaxial Piezoelectric Fiber for Bending and Vibration Sensing

For some patients who cannot communicate with the doctor through language because of his illness, transferring information by bending the finger attached with the 1D flexible coaxial piezoelectric fiber in an emergency is supposed to be an efficient communication way (Fig. 3a). In this work, by using the coding principle of Morse code (*i.e.*, the combination of dot signal and horizontal one),<sup>[39]</sup> a new communication mechanism is established by combining finger bending speed with dot signal and horizontal one.<sup>[40]</sup> In other words, rapid bending of

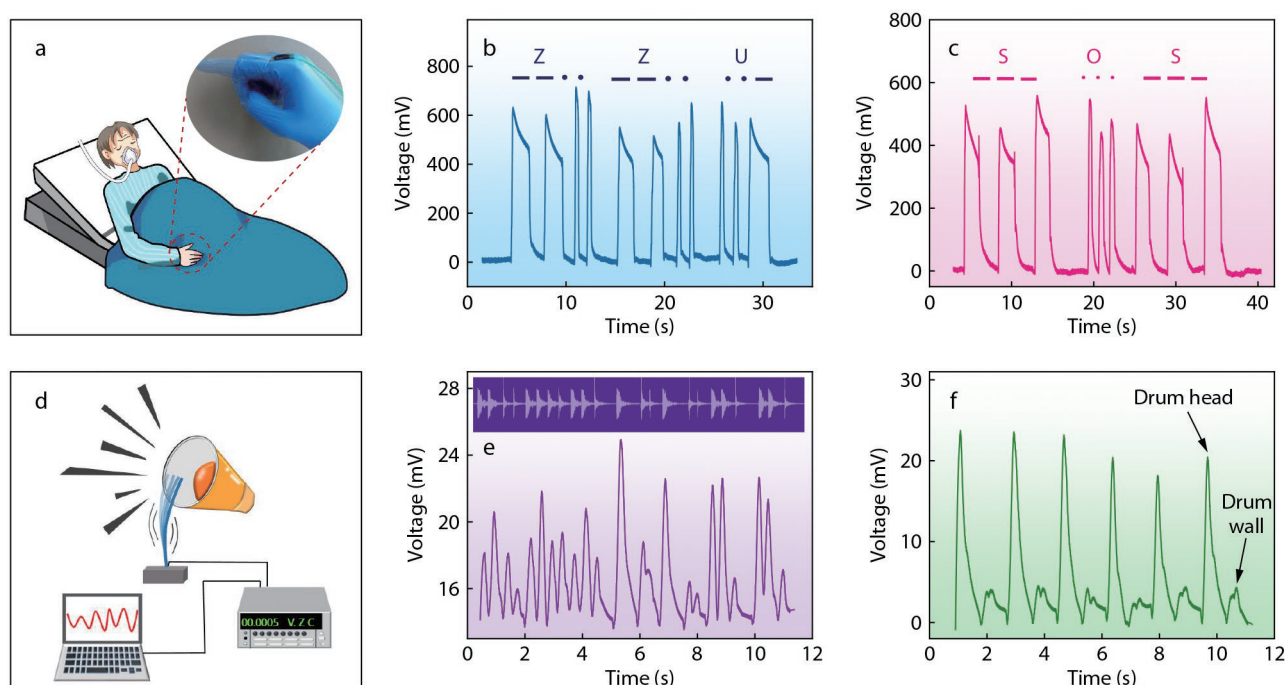


**Fig. 3** (a) Schematic diagram of information transfer based on bending mode; Generation of Morse code (b) “ZZU” and (c) “SOS”; (d) Schematic diagram of the audio information analysis based on vibration mode; (e) Response of 1D-PF4 to audio emitted from drum head; (f) Response of 1D-PF4 to the combined audio that successively emitted drum head and drum wall.

finger represents dot signal, while slow bending of finger represents horizontal one. Thus, with the help of combination of rapid and slow bending of finger, a series of useful messages can be easily sent (*i.e.*, “ZZU” and “SOS”, in Figs. 3b and 3c).

In addition, sound transmission can cause air oscillation, resulting in the periodic change of air pressure. Therefore, human can hear such oscillation when pressure waves are perceived by people’s eardrums.<sup>[41]</sup> Herein, such 1D flexible coaxial piezoelectric fiber is used to sense the acoustic-driven vibration of a loudspeaker (Fig. 3d). When the loudspeaker plays audio from the drum head, the details of volume, rhythm and dynamics are recorded immediately (Fig. 3e), and the collected output voltage exhibits a synchronous response to these original audios. Moreover, such 1D flexible coaxial piezoelectric fiber also has response to the audio from the drum wall (Fig. S9 in ESI). More interestingly, when the loudspeaker plays the combined audio that successively emitted drum head and drum wall, it is clear that output voltage of drum head and drum wall are completely different because of the different vibrations caused by different timbres (Fig. 3f). In view of these, such 1D flexible coaxial piezoelectric fiber has excellent performance when sensing the acoustic-driven vibration.

### Piezoelectric Output Performance of 3D Helical Piezoelectric Fiber (3D-PF)

It is well known that stretchability is critical for wearable

electronics.<sup>[42–44]</sup> In order to endow the as-prepared 1D piezoelectric fiber with excellent stretchability, a 3D helical structure was built by heat-setting method (Fig. S2 in ESI). Herein, output voltage of 3D-PF under different strains ( $\varepsilon = \Delta L/L \times 100\%$ ,  $L$  is the original length,  $\Delta L$  is the change in length)<sup>[45]</sup> was explored by uniaxial tensile test (Fig. 4a). Clearly, output voltage generally exhibits a monotonically increasing trend with the increase of strain (Fig. 4b). Moreover, Fig. 4(c) exhibits that there is an excellent linear relationship between output voltage and strain. Additionally, the response time and recovery time of the 3D-PF are respectively 360 and 348 ms (Fig. 4d). Furthermore, output current of 3D-PF shows no significant decrease even after 2000 stretching/releasing cycles, indicating remarkable stability and durability (Fig. S10 in ESI).

### Application of 3D Helical Piezoelectric Fiber as Self-powered Motion Monitoring System (SP-MMS)

Fig. 5(a) indicates that 3D-PF can be sophisticated braided into commercial textiles (3D-PF/textile) without any negative influence on wearability and breathability due to its ingenious helical structure. Moreover, such 3D-PF/textile can not only be folded and twisted (Figs. 5b and 5c), but also stretched (Fig. S11 in ESI). More interestingly, such 3D-PF/textile stretched to 30% can generate a voltage of ~90 mV whether or not it was suffered 500 cycles of folding or twisting (Fig. 5d), suggesting remarkable stability and durability. Apart from excellent deformability, washability is also critical for wearable electronics.<sup>[46,47]</sup> In this

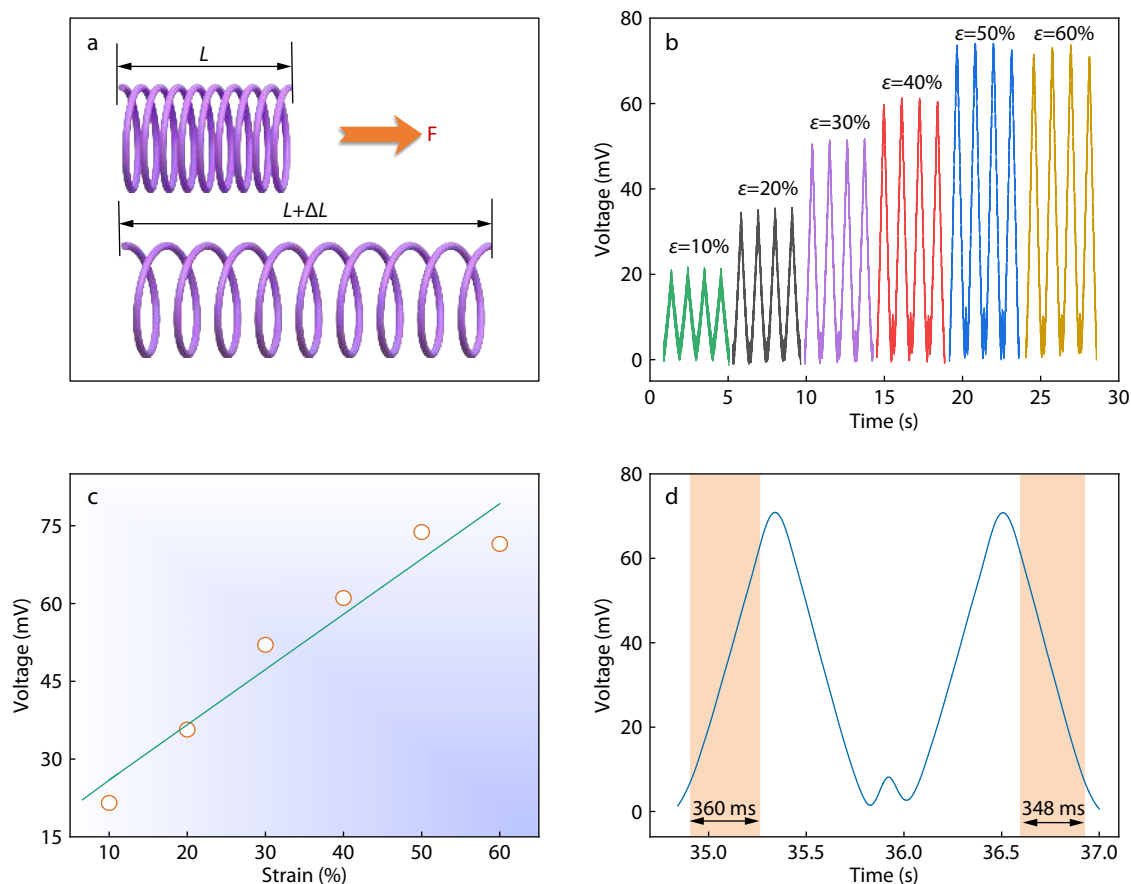


**Fig. 4** (a) Schematic diagram of 3D-PF upon stretching; (b) Output voltage of 3D-PF under different strains at 1 Hz; (c) Relationship between output voltage of 3D-PF and strain; (d) Response and recovery time of 3D-PF at the strain of 50%.

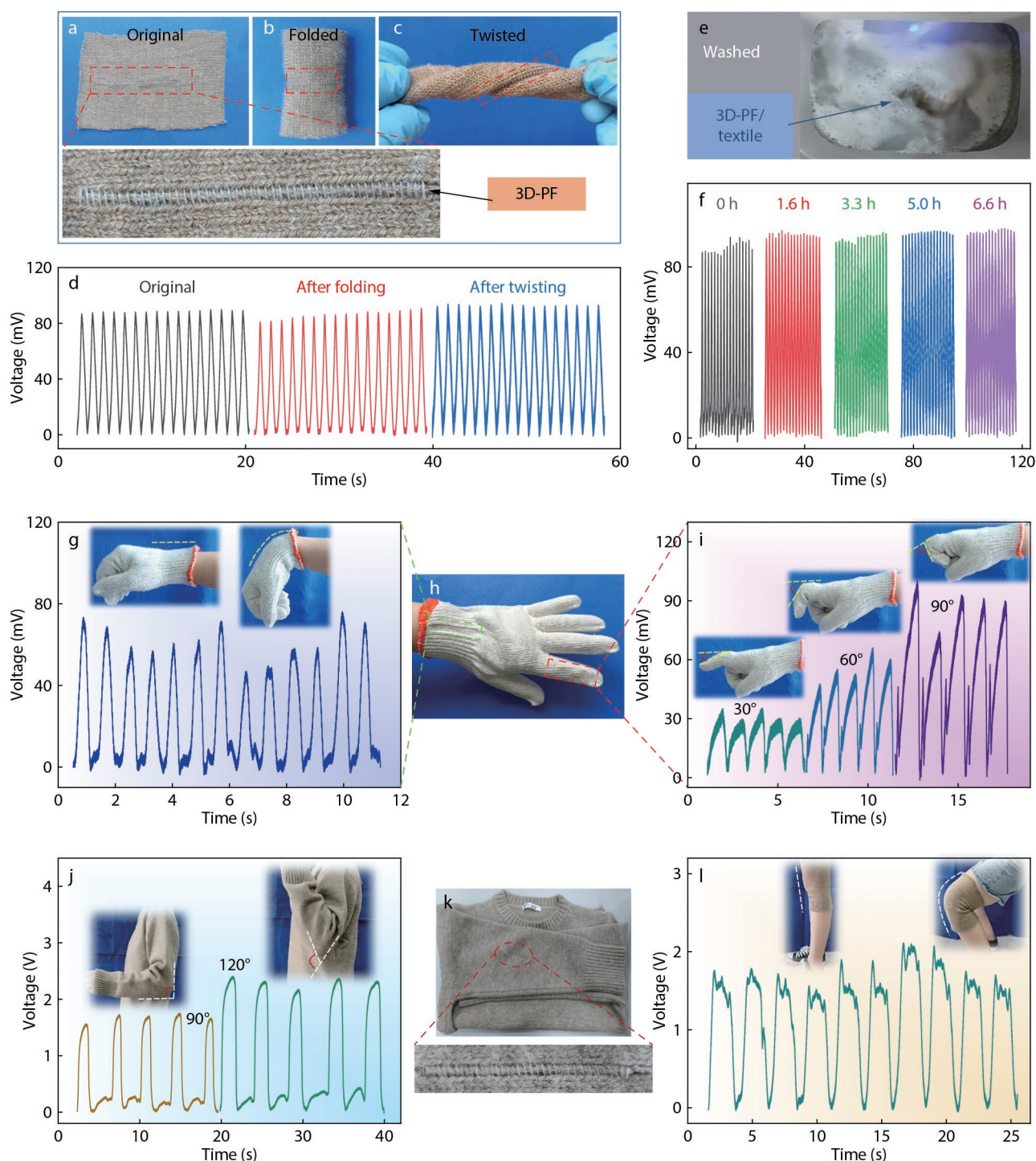


**Fig. 5** (a) 3D-PF/textile is (b) folded and (c) twisted; (d) Output voltage of 3D-PF/textile before and after being suffered various extreme deformations; (e) Machine wash test of 3D-PF/textile and (f) its output voltage after being washed for 6.6 h; (g) Wrist joint bending; (h) Motion monitoring system based on smart glove; (i) Index finger bending at different angles; (j) Elbow bending at different angles; (k) Motion monitoring system based on smart sweater; (l) Knee joint bending during squatting.

work, standard machine washing (see specific parameters in ESI) as shown in Fig. 5(e) provides the harshest test conditions for such unpackaged 3D-PF. Interestingly, such 3D-PF/textile still has the same output voltage as the original one after being subjected to 6.6 h of extreme washing (Fig. 5f). Such remarkable washability of 3D-PF/textile should be ascribed to the fact that

the electrode of 3D-PF was completely packaged and protected by PVDF. That is, the protection and waterproof properties provided by PVDF would render it with resistance against various extreme conditions. In a word, on one hand, 3D-PF/textile possesses remarkable wearability due to the fact that 3D-PF can be subtly braided into commercial textiles. On the

other hand, 3D-PF/textile also possesses excellent stability after being suffered various extreme conditions, indicating its promising application potential in wearable electronics or smart textiles in the future.

Furthermore, thanks to the wearability and excellent stability of 3D-PF/textile, a motion monitoring system is designed to monitor human motion of joints. As shown in Figs. 5(g)–5(i), it can not only monitor the bending of wrist joint, but also accurately sense the bending of index finger at different angles because of its high sensitivity. Interestingly, such motion monitoring system can also monitor elbow bending at different angles (Fig. 5j) and knee joint motions during squatting (Fig. 5l). These results suggest that the motion monitoring system can accurately sense the motion of large and small joints during human motions, and has potential applications in the field of motion tracking and health monitoring.

## CONCLUSIONS

In summary, a 1D flexible coaxial piezoelectric fiber (1D-PFs) can be one-step but continuously fabricated by melt extrusion molding, whose core and sheath layer are respectively slender steel wire (*i.e.*, electrode) and PVDF (*i.e.*, piezoelectric layer). Moreover, such 1D flexible coaxial piezoelectric fiber possesses short response time and high sensitivity, which can be used for bending and vibration sensing. Interestingly, the as-fabricated 1D flexible coaxial piezoelectric fiber can be further endowed with 3D helical structure and integrated with commercial textiles as a wearable and washable self-powered motion monitoring system (SP-MMS). The one-step, continuous and environmentally friendly fabrication method developed in this study can be facilely scaled up for low-cost industrial production of 1D and 3D piezoelectric fibers, and will promote their applications in sensing and wearable electronics.

## Conflict of Interests

The authors declare no interest conflict.

## Electronic Supplementary Information

Electronic supplementary information (ESI) is available free of charge in the online version of this article at <http://doi.org/10.1007/s10118-023-2960-0>.

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