Development of an Elastic Piezoelectric Yarn for the Application of a Muscle Patch Sensor

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ABSTRACT: In this paper, an elastic poly(vinylidene fluoride-co-trifluoroethylene) piezoelectric yarn for the application of a muscle patch sensor is presented. The electrospinning method is used to fabricate the piezoelectric yarn, and different parameters were used to control the orientation and structure of piezoelectric fibers. We further develop a post-alignment process to reorganize the orientation of fibers and to reshape fiber microstructures. Two unique microstructures of piezoelectric fibers that have an excellent elastic performance were identified. This piezoelectric yarn is composed of skewed and crimped fibers that align along the elongation direction, and it can be cyclically stretched up to 65% strain with good linearity, durability, and repeatability. Its mechanical behavior is superior to randomly distributed and fully straightened piezoelectric fibers, and it is suitable for long-term use of larger strain sensing. Our study demonstrated that this piezoelectric yarn can be stretched for more than 12 h under a repeated 1 Hz cyclic deformation. Using this elastic piezoelectric yarn, a muscle patch sensor that can be attached to the skin over human muscles for real-time monitoring is developed. The concentric, eccentric, and isometric contractions of biceps and triceps can be measured simultaneously to study their contraction behaviors. To further verify whether this patch sensor can be used under intense exercise conditions, the contraction behavior of a soleus muscle during stationary jumping and running is monitored to demonstrate sensor performance. Finally, this patch sensor is sewed onto a chest band, and it is verified that both breathing movement and heartbeat can be monitored.

1. INTRODUCTION

For the past decade, wearable devices that can be attached to the human body for various functions have become an emerging research and commercial field for linking human daily activities to portable devices and for improving work efficiency, health monitoring, personalized health care, and diagnosis. Comprehensive reviews can be found in reports published by Kenry et al.,1 Stoppa and Chiolerio,2 and Heo et al.3 Peake et al. also provided a detailed review of commercial products on the market.5 It is believed that wearable devices can play an important role in data collection for applications of big data,5 machine learning,6 and artificial intelligence.7

Among reported wearable devices, physical sensors that are lightweight, flexible, and stretchable for direct wearing on human bodies make up one of the most active fields. This type of sensor needs to have a high degree of deformability to provide a good conformability for attaching on the skin surface and a good comfortability for long-term wearing. These physical sensors usually are mechanically deformable sensors that use the geometrical deformations of sensing materials to perform measurements. Detection mechanisms can be separated into four categories: piezoresistance, capacitance, triboelectricity, and piezoelectricity.

The piezoresistive-based physical sensor is one of the most studied methods. The approaches to make these resistive-based sensors stretchable are based on nonwoven fibers,8,9 twisted fibers,10 or embedding in an elastomer-like polydimethylsiloxane.11–14 Their applications include hand gesture,8,9,12,14 breathing,10,14 arterial pulse wave,11,13,14 joint movement,12 and so forth. Capacitance-based physical sensors usually are applied for pressure sensing that measures the thickness change between electrodes, such as arterial pulse waves on wrists.15,16 Another application is to measure the overlapping areas between two electrodes for skin-motion detections.17 On the other hand, triboelectricity has also been applied for the measurement of joint movement.18,19

The fourth type of physical sensor is based on piezoelectric polymers, including poly(vinylidene fluoride-co-trifluoroethylene) [P(VDF-TrFE)],20–24 polyvinylidene difluoride,25 poly- (l-lactic acid),25 and poly(l-lactic acid) and poly(n-lactic acid) composites.26 These sensors usually are designed as stretchable sensors for body movement detection or pressure sensors for weight measurement from feet. Although the typical elasticity of piezoelectric polymers is several GPa and is the lowest among piezoelectric materials, their elastic modulus still is much higher than human skin or muscle. Thus, efforts have been made to...
increase its stretchability and applicable strain. Reported methods include using fiber-based structures,\(^\text{20–22,25}\) curved film-structure,\(^\text{23}\) coil structure,\(^\text{24}\) and multilayered films.\(^\text{27}\) Among these methods, twisted electrospun \(\text{[P(VDF-TrFE)]}\) nanofiber ribbons can provide the largest deformations of up to 740\% strain.\(^\text{22}\) The toughness and strength for large deformation were improved considerably. Nevertheless, a large hysteretic effect resulted, and a certain level of plastic deformation could be induced.

Among these approaches, using randomly distributed nano-fibers or microfibers to increase deformability and compliance is a common method.\(^\text{8,9}\) Fibers, however, can start to align along the direction of deformation, making the output signal no longer repeatable after long-term use, where its characteristic curve can drift considerably. On the other hand, highly aligned fibers also have been used.\(^\text{12,20,25}\) This type of sensor, however, cannot be very compliant. It can be very fragile under a high level of strain, and a permanent plastic deformation can result, making it unable to return to its original length after releasing. Last, a twisted structure can sustain a large deformation but can have a highly nonlinear characteristic curve.\(^\text{21,22}\) A further challenge for electrospun piezoelectric fibers is that dipoles have a preferred orientation perpendicular to the axial direction, and it makes its piezoelectric coupling effect to be lower in the axial direction. Jiang et al. shows that the piezoelectric output charges of aligned \(\text{P(VDF-TrFE)}\) nanofibers in the axial direction can be nearly 2 orders lower than in the thickness direction.\(^\text{28}\) Thus, electrodes for sensors made by piezoelectric fibers are usually placed on the top and bottom surfaces. This design can limit sensor deformability and stretchability as metallic electrodes are stiffer and fragile.

To reduce the influence of dynamic modifications of fibers during repeated large deformations, we studied the correlation between orientations and structures of piezoelectric fibers and sensor performance. Our study showed that, using a combination of skewed and crimped piezoelectric fibers aligned along the elongation direction, an elastic piezoelectric yarn that had a good linearity, repeatability, and durability was created. The axial piezoelectric response was improved by poling in the axial direction, and silver wires can be connected at two ends with minimal influence on the deformation of the piezoelectric yarn. Using the developed piezoelectric yarn, we further developed a muscle patch sensor to record contractions of human muscles during different activities. In this paper, experimental studies on the piezoelectric fibers and their sensing performance are presented. The feasibility and application of the muscle patch sensor also are discussed.

2. EXPERIMENTAL SECTION

2.1. Electrospinning Setup of the P(VDF-TrFE) Piezoelectric Yarn. The \(\text{P(VDF-TrFE)}\) piezoelectric yarn was fabricated via the standard electrospinning process and collected by a rotating drum collector. Figure 1A shows an illustration of the electrospinning setup, which includes a syringe pump (KD Scientific, KDS 100) and a glass syringe with a 27-gauge stainless steel blunt needle (spinneret), a rotating drum collector, and a high-voltage DC power supply (Matsusada Precision Inc., AU40N7.5). The spinneret-to-collector distance \((d)\) was set at 5 cm to reduce the level of whipping instability during the process. This effect was because electrical charges injected in the electrospinning jet can induce instability in the air and created a randomly distributed fiber mat on the collector.\(^\text{29,30}\) Thus, combining a short spinneret-to-collector distance and the rotating speed of the drum collector, the orientation and structure of electrospun fibers can be controlled.

Three rotating speeds were used to create fibers with different structures and orientations, including 500, 1100, and 1700 rpm. The corresponding linear velocities were 3.93, 8.64, and 13.35 m/s. Based on the structure of the as-spun piezoelectric fibers that will be discussed in Figure 2, fibers collected under the conditions of 500, 1100, and 1700 rpm are labeled as CuTw fiber (curly twisted), CrSk fiber (crimped and skewed), and CrSkRe fiber (crimped and skewed—reduced), respectively. The average diameter of electrospun fibers was 1.51 \(\mu\)m with a standard deviation (STD) of 0.23 \(\mu\)m. The collected piezoelectric fibers were rolled up to form a piezoelectric yarn. These piezoelectric yarns also were labeled as CuTw yarn, CrSk yarn, and CrSkRe yarn, respectively.

To further enhance the elastic performance of the piezoelectric yarn, a post alignment process was developed. A cyclic stretching process was applied to reorganize the orientation of fibers and to reshape the fiber microstructures. In the following sections, the effects of the alignment process on the orientation and structure are studied. Its contributions to the elastic performance and signal output are also discussed. The optimized alignment process was a 2.5 h 65\% strain alignment process, an Al— was added to the piezoelectric yarn for clarification, such as Al—CuTw yarn, Al—CrSk yarn, and Al—CrSkRe yarn.

2.2. Experimental Setup of the Muscle Patch Sensor. After analyzing the elastic performance, linearity, and repeatability of piezoelectric yarns, it was found the aligned Al—CrSk yarn provided the most reliable performance. Using this yarn, we developed a muscle patch sensor that can be directly attached to the skin over a muscle, as shown in Figure 1B. Figure 1C,D shows images of the fabricated device and aligned piezoelectric yarn, respectively. The Al—CrSk yarn was sandwiched between two 0.1 mm thick thermoplastic polyurethane (TPU) sheets, and two silver threads were used as electrodes and connecting to a Bayonet Neill—Concelman (BNC) cable. To measure the level of muscle forces, the fabricated muscle patch sensor was placed perpendicular to the direction of muscle contractions as the resultant circumference change of muscles is proportional to the level of generated muscle forces. An increase of muscle
Figure 2. SEM micrographs of electrospun piezoelectric fibers, where (A–C) are as-spun fibers, (D–F) are fibers under a 50% tensile strain, and (G–I) are fibers released from a 50% strain to 0% strain. (A,D,G), (B,E,I), and (C,F,I) are CuTw fibers (500 rpm), CrSk fibers (1100 rpm), and CrSkRe fibers (1700 rpm), respectively. The rotation direction of the drum collector is in the vertical direction. (Scale bar = 20 μm).

3. RESULTS AND DISCUSSION

3.1. Orientation and Structure of the As-Spun Fibers Can Be Controlled. Figure 2 shows experimental results of as-spun P(VDF-TrFE) fibers using SEM. The corresponding rotating direction is in the vertical axis. Figure 2A–C are micrographs of as-spun fibers collected at a 500, 1100, and 1700 rpm rotating speed. This shows that, at a low rotating speed of 500 rpm (Figure 2A), fibers had a curly twisted structure, and these were named CuTw fibers. The orientation had a low-level alignment along with the rotating direction of the drum collector. At a higher rotating speed of 1100 rpm (Figure 2B), a cramped and skewed structure was observed and a higher level of alignment along the rotating direction was observed. Nevertheless, the orientation of fibers was skewed at an angle with respect to the rotating direction. The average angle was 15.2° with a STD of 7.4°. This type of fiber was named CrSk fibers. The cramped and skewed structure of fibers was reduced considerably under a higher rotating speed of 1700 rpm, as shown in Figure 2C, and these were named CrSkRe fibers. The radius of curvature was increased significantly and the majority of P(VDF-TrFE) fibers were almost aligned with the rotating direction. Very few fibers had a large skew angle, and the average skew angle was 6.8° with an STD of 7.4°. This result demonstrated that the orientation and structure of piezoelectric fibers can be controlled and adjusted using the rotating speed of the drum collector with a shorter spinneret-to-collector distance of 5 cm.

3.2. As-Spun Fibers Have Distinct Orientation and Structure under a Large Deformation and after Releasing. To study the response of as-spun fibers under a large deformation, a 50% tensile strain was applied. The orientation and structure of fibers under this large deformation and after releasing were studied. Figure 2D–F shows the SEM micrographs of CuTw, CrSk, and CrSkRe fibers stretched at a 50% strain in a vertical direction, respectively. Corresponding micrographs of fibers released from a 50% strain are shown in Figure 2G–I. The experimental results demonstrated that fibers were all straightened along with the applied tension in the vertical direction. It was found that the curly twisted structure of CuTw fibers (500 rpm) became very small under tension, and most fibers got straightened (Figure 2D). This tightened structure rebounded a bit after releasing the tension, as shown in Figure 2G. Nevertheless, the curly twisted structure did not return to its original structure (Figure 2A), and its orientation became less randomly distributed and had a higher degree of alignment along the tension direction. The straightening effect also was observed on CrSk fibers (1100 rpm) at 50% strain, as shown in Figure 2E. The majority of the crimped fibers were straightened by the application of tension, and skewed fibers also were aligned along with the tension direction. After releasing (Figure 2F), the radius of curvature of the crimped structures became much larger, and the angle of skewed fibers also was reduced considerably. The measured average skew angle was 3.0° with an STD of 3.8°. The average skew angle was reduced 5.1 times by one 50% strain stretching process. As for CrSkRe fibers (1700 rpm), all fibers were nearly straightened under 50% strain, and all the crimped structures almost disappeared (Figure 2F). This stretching process makes these fibers more aligned along with the tension direction after releasing, and the crimped...
structure was reduced considerably (Figure 21). The average skew angle became 2.1° with an STD of 1.9°. The average skew angle was reduced by 3.2 times after one stretching process.

Based on this experimental study, it was found that the stretching process can modify the orientation and structure of piezoelectric fibers significantly. In the macroscopic scale, these microscale remodeling processes resulted in a plastic deformation, making the overall length become longer after each cyclic stretching process. This dynamic remodeling process can cause considerable drifting of the characteristic curve and nonlinearity performance. This effect is a common issue for fiber-based physical sensors, particularly for the application of large deformations and long-term use. This can be a big obstacle for commercializing fiber-based physical sensors for practical applications.

In the following sections, we present our study on the alignment process to reorganize the orientation and structure of piezoelectric fibers to have a good elastic performance and repeatability for overcoming the constant dynamic remodeling issue of fiber-based physical sensors.

3.3. Mechanical Performance of As-Spun Piezoelectric Yarns. Figure 3A shows tensile test results of three piezoelectric yarns, where each yarn was stretched until it broke off. As the cross section was not quantified easily, the force–strain curve was used to study the mechanical performance. Arrows indicate the vertical axis used for each condition. Note that there was a nearly instantaneous increase of stress with a steep slope during the initial stage for all three conditions. Its level was highest for CrSkRe yarn (0.3% strain at 32.5 mN) followed by CrSk yarn (0.9% strain at 6.7 mN), and the smallest one is CuTw yarn (1% strain at 1.0 mN). This effect corresponded to the portions of fibers that were initially aligned in the direction of tensile force. After this region, the CuTw yarn showed a plateau force with an increase of 1.8 mN between 3.2 and 80% strain, suggesting that a lot of curly twisted fibers were straightened and contributed to this nearly flat region. After 80% strain, the force magnitude started to increase with the strain hardening effect and broke at 235.9% strain under 15.4 mN force. In contrast, the CrSkRe fiber shows a second elastic region from 1.2 to 24.2% strain followed by a third elastic range and broke at 311.2% strain with 248.7 mN force. This result suggests that the CrSkRe yarn had a characteristic of glassy polymer. The CrSk fiber has behavior similar to CrSkRe, but it only has a second elastic region up to 73% strain followed by a strain softening effect and broke at 202.6% strain under 20 mN force.

These results suggest that the resultant strain of piezoelectric yarns in the macroscopic scale is contributed not only by the material strain but also by the structure strain. The structure strain contributes a portion of the large deformation, and it is the reason that the yarn does not need a high-level force to create deformations. This structural effect also is the major reason that most of fiber-based sensors cannot have a good linearity, repeatability, or durability as its structural deformation is not repeatable for each cycle.

3.4. Effect of the Alignment Process. Here, we present our study on using a postalignment process to organize the orientation of fibers and reshape their microstructures so that the resultant macroscopic behavior can be nearly elastic with a good repeatability. Figure 3B–D shows measured characteristic curves of CuTw, CrSk, and CrSkRe yarns, respectively. Gray lines are the first five cycles of as-spun fibers. It was found that the characteristic curves drifted to more positive values during each cycle, and CuTw yarn and CrSkRe yarns drifted more significantly than the CrSk yarn. Note also that the characteristic curve of the CuTw yarn was not stable throughout the stretching cycle. This effect could have been induced by the curly twisted structure of piezoelectric fibers that were dynamically modified during the stretching process. Furthermore, this result verified that the remodeling process of piezoelectric fibers can alter the characteristic curve significantly. This effect could become a considerable issue for commercialization.

To improve the repeatability and linearity, an alignment process was conducted. A 1 Hz cyclic tensile strain in the range between 0 and 10 to 12% was applied continuously for 12 h, and the signal response was monitored during the process. Black lines shown in Figure 3B–D are signal responses of the first five cycles after 2.5 h of the stretching process. This demonstrated that the drifting effect was reduced, except for the CrSkRe yarn. Furthermore, the level of hysteresis was also reduced after 2.5 h of the alignment process for all three types of yarns. To study the influence of the alignment process to the reduction of the hysteresis curve, the difference between the stretching and releasing cycles was measured for each strain value, and the overall STD was calculated. Figure 3E shows the variations of STD values during the 12 h alignment process. This analysis demonstrated that the CrSk yarn can reach a plateau in 2.5 h and its STD value was much lower than those of the other two yarns. The level of hysteresis of the CuTw yarn and CrSkRe yarn did reduce over time, but it could not reach a stable condition after 12 h of the alignment process. This result suggests that the random orientations and curly twisted structure of the CuTw yarn can keep remodeling for 12 h under a 10% cyclic tensile strain. On the other hand, the as-spun CrSkRe yarn has fibers
with high-level alignment that could experience breakage of fibers during the alignment process.

This study verified that the orientation and structure of piezoelectric fibers can be adjusted using a cyclic tensile strain. Nevertheless, the remodeling process of random piezoelectric fibers, such as CuTw fibers, cannot reach a steady state after a 12 h alignment process. On the other hand, piezoelectric fibers with more organization and alignment after the electrospinning process also can have a hard time to reach a steady state, like CrSkRe fibers. This effect could be because the part of fibers that had been straightened were ruptured under tension, making the macroscopic response not repeatable. In contrast, fibers that were straightened with a small skew angle or had a cramped structure can sustain a large deformation and maintain macroscopic elasticity with a good repeatability. The aligned CrSk fibers demonstrated this capability.

3.5. Mechanical Performance of Aligned Piezoelectric Yarns. To investigate the performance of piezoelectric yarns under a high mechanical strain, a 1 Hz cyclic tensile strain between 0 and 65% was applied. The measured maximum pressure applied on the pressure sensor at 65% strain was 3.93 MPa. Figure 4A shows tensile test results of CuTw, CrSk, and CrSkRe yarns that been aligned for 2.5 h, which are labeled as Al–CuTw, Al–CrSk, and Al–CrSkRe yarns, respectively. The force–strain curve of Al–CuTw yarn, Al–CrSk yarn, and Al–CrSkRe yarn were further reduced after 1.67 and 1.38 h, respectively. Furthermore, the Al–CrSk yarn reached a plateau near the end of the 2.5 h alignment process, which suggests that the Al–CrSk yarn can provide stable sensing performance for long-term use after a 65% large strain alignment process.

Figures 5A–C,D–F,G–I are SEM micrographs of the corresponding orientations and structures of Al–CuTw fibers, Al–CrSk fibers, and Al–CrSkRe fibers under three different conditions after 2.5 h 65% alignment process. The three different conditions are (1) right after the alignment process (Figure 5A,D,G), (2) under a 50% tensile strain (Figure 5B,E,H), and (3) after releasing of the 50% tensile strain (Figure 5C,F,I).

Figure 5A shows the micrograph of Al–CuTw fibers right after the alignment process. The strain applied to these fibers was 0%. It was found that the curly twisted structure was straightened partially, and the radius of curvature of the curly twisted structure became much smaller than the as-spun fibers shown in Figure 2A. This curly twisted structure can be tightened more under a 50% tensile strain, as shown in Figure 5B, where many of the fibers were straightened along with the direction of tensile force. After releasing the 50% strain, the straightened structure recovered partially to a different state, as shown in Figure 5C. This observation suggests that the elastic behavior of the Al–CuTw yarn was controlled by the modification of the curly twisted structure. Because of the dynamic remodeling of this structure, however, the orientation and structure can be different for each stretching cycle. This effect contributed to the drifting effect of the Al–CuTw yarn, making it an unreliable piezoelectric yarn.
Figure 5 shows a micrograph of the Al–CrSk yarn right after the alignment process. The crimped structure of the as-spun fibers shown in Figure 2D was aligned along with the direction of tensile force. The radius of curvature of the crimped structure was increased slightly. The average skew angle with respect to the direction of tensile force was 3.1° with a STD of 4.7°. These crimped structures straightened to almost straight structures of crimped and skewed fibers under a 50% tensile strain and could recover after releasing the tensile force, where Figure 5E,F show the corresponding micrographs. The average skew angle was 1.72° (STD = 1.34) and 2.87° (STD = 2.04), respectively. This result suggests that the fibers with a skewed angle and crimped structure can have a more repeatable response during a large stretching process, and there can be a reliable microscopic deformation to provide an elastic property for the piezoelectric yarn in the macroscopic scale.

Figure 5G shows the micrograph of the Al–CrSkRe fibers after the alignment process. It was found that many fibers were broken, when compared to the as-spun fibers (Figure 2C). The unbroken fibers were all straightened in the direction of the tensile force with a very small skew angle. The average skew angle was 1.83° with an STD of 1.57°. These fibers can be straightened more under a 50% tensile strain, as shown in Figure 5H, where broken fibers were observed. The average skew angle was further reduced to 1.75° with a smaller STD of 1.55°. After releasing 50% strain, the structure of the straightened fiber can be retained with a small skew angle. Nevertheless, more broken fibers were found. The average skew angle was 1.38° with an STD of 1.53°. This result suggests that Al–CrSk fibers are not suitable for a large deformation application. This is because their fibers have a high level of alignment along with the stretching force, and fibers can continue to break off one after another during each stretching process. It cannot be applied for long-term applications as its characteristic curve can drift gradually over time.

3.6. Elastic Performance of the Aligned Al–CrSk Yarn. After studying three types of piezoelectric fibers with distinct orientations and structures at the microscopic level, it was found that all piezoelectric yarns made of these three piezoelectric fibers could sustain a tensile strain as high as 65%. Nevertheless, only Al–CrSk fibers could maintain their microstructure after a 2.5 h 65% cyclic strain alignment process. They can provide long-term use with excellent repeatability and a small hysteretic effect. This unique performance was enabled by the micro-structures of crimped and skewed fibers with a small skew angle. To verify the performance of the Al–CrSk yarn, we applied different levels of tensile strains to study its elastic characteristic curve after the alignment process.

Figure 6A shows signal responses of aligned Al–CrSk yarn under a small cyclic strain, including 5, 10, and 15%. The measured signal by the current amplifier was integrated with respect to time to retrieve sensor charge output. It was found that the Al–CrSk yarn can have a complete loop with a small hysteretic effect under 10 and 15% tensile strain but not under 5% strain. This effect could be due to the applied tension under 5% being insufficient and fibers possibly bending during the contraction cycle, resulting in an incomplete hysteretic loop. Figure 6B shows signal responses of the Al–CrSk yarn under a high cyclic strain, including 30, 35, 45, 50, 55, and 60%. It was found that the aligned Al–CrSk yarn can have a repeated hysteretic loop under high tensile strain after the alignment process. This also suggests that this type of fiber can have good elasticity for large deformation measurement. Note that tensile strain higher than 65% was not studied as the orientation and structure of fibers can be modified further under such conditions. The sensor performance could drift again unless fibers are aligned with respect to a higher mechanical strain.

3.7. Verification of Piezoelectric Performance. To verify the crystalline structure, an X-ray diffractometer (Malvern Panalytical, X’Pert³ MRD XL) was used to conduct a 2θ scan on the CrSk piezoelectric yarn before and after the alignment process. Figure 7A shows the scanning results. The peak appears between 19 and 20°, suggesting that they both were β-phase piezoelectric fibers, and aligned fibers had a stronger β-phase response. The average diameters of fibers decrease from 2.06 μm
These results suggest that the alignment process can assist β-phase formations. Furthermore, to confirm that the polarization process can increase the piezoelectric response in the axial direction, one silver wire served as a signal line and the other was grounded. Then, their roles were switched to verified dipole orientations. Figure 7B,C shows the measured signal output (red lines) versus displacement field (black lines) under a 35% cyclic strain. It shows that the signal output had a 180° phase difference when switching the direction. This result suggests that dipoles can be skewed toward the axial direction through the polarization process. Finally, we also verified the performance of a muscle patch sensor after sealing Al−CrSk piezoelectric yarn between two TPU films. Figure 7D shows the correlation of the sensor output under 63% cyclic strain for five cycles. It shows that the muscle patch sensor still had a good linearity and repeatability, and the hysteretic loop was reduced because of the small viscoelastic behavior of TPU films.

3.8. Finite Element Analysis on the Crimped and Skewed Piezoelectric Fibers. Having identified that crimped and skewed piezoelectric fibers can provide a good elasticity, their microscopic deformation versus signal output was investigated by the finite element method. Figure 8A shows von Mises stress of skewed piezoelectric fibers being stretched from 0% (left) to 50% strain (right). This shows that the level of induced stress was higher with a smaller skewed angle. This effect also was evident in the charge output during the stretching process, as shown in Figure 8B. Note that the charge output displayed good linearity with fibers in different skewed angles. Identical analysis also was conducted for crimped fibers. The simulated von mises stress from 0% (left) to 50% strain is shown in Figure 8C. It shows that the stress level was highest around the antinodal points of crimped fibers, which are the locations that are nearly parallel with the stretching direction. Note that fibers with a larger radius of curvature have a higher level of stress. The corresponding charge output over the stretching process also had the same characteristics with good linearity, as shown in Figure 8D.

These finite element studies demonstrate that both crimped and skewed piezoelectric fibers have a good linear response, and they can be used to measure large deformations. Furthermore, fibers with smaller skewed angle and larger radius of curvature can have a higher sensitivity and signal output, verifying that aligned Al−CrSk fibers can provide a better sensor performance than as-spun CrSk fibers. Finally, this result also suggests that the unique structure and orientation of crimped and skewed piezoelectric fibers are the origin of the excellent elastic performance of the Al−CrSk piezoelectric yarn.

3.9. Study Contraction Profiles of Biceps and Triceps Using the Muscle Patch Sensor. After confirming the elastic performance of the Al−CrSk yarn under different levels of...
tensile strain, we used it to develop a muscle patch sensor to measure the variation of the muscle circumference during contraction and extension exercises. Thus, contraction profiles of different muscles can be monitored and studied, and the feasibility of the muscle patch sensor was verified.

Figure 9A,B shows two images of the experimental setup to simultaneously measure the contraction profiles of biceps and triceps using two muscle patch sensors, which are labeled by red and blue arrows, respectively. Both muscle patch sensors were attached perpendicularly to the direction of muscle contraction. Figure 9A shows the action of the upward phase of exercise, negative peak also was observed from the triceps signal. On the contrary, a pair of inverse signals was observed during the downward phase of exercise, which is shown between 23 and 32 s in Figure 9C,D. This experimental result demonstrated that the muscle patch sensors can monitor contraction behavior of muscles. Using a pair of muscle patch sensors, the functions of biceps and triceps were observed, showing that they were one of agonist-and-antagonist muscle pairs of the upper arm. A Supporting Information Video S1, video-Fig9CD.mp4, shows the recording of this experiment, where the top yellow line and the bottom blue line on the oscilloscope are signals of the triceps and biceps, respectively.

Based on the experimental study, it was found that the signal output of the muscle contraction is a biphasic profile using a current amplifier as the interface circuit for the muscle patch sensor. For a concentric contraction, the muscle patch sensor will pick up a negative peak followed by a positive peak biphasic signal. On the contrary, a biphasic signal with a positive peak followed by a negative peak will be picked up during an eccentric contraction.

As the length of the muscle was also changed during the concentric and eccentric contractions, the correlation between the signal profiles and muscle actions can be complicated. To verify the correlations between signal output of the current amplifier and muscle contractions, we studied the signal profile during the isometric contraction of one tricep. The subject’s arm was maintained at the posture shown in Figure 9B. The experimental result is shown in Figure 9E, where the black line represents the measured signal using the current amplifier. The first negative peak was the onset of the isometric contraction, and the force was maintained for about 20 s. The second positive peak represents the force releasing process. This result demonstrated that the applying and releasing processes of a muscle force are correlated to the signal with a negative peak and a positive peak, respectively. While the muscle force reached and maintained a complete tetanus state, the output signal of the current amplifier remained zero. Integrating the signal of the current amplifier with respect to time, the charge output of the muscle patch sensor can be found and is shown as the gray line in Figure 9E. The gray line shows that the contraction force continuously increased during the onset of isometric contractions and was maintained at a plateau during the biceps holding at its maximum force output. Then, the signal returned to zero during the force releasing process at the end of the cycle. This result demonstrated that the contraction profiles can be monitored and studied using the muscle patch sensor. A Supporting Information Video S2, video-Fig9E.mp4, shows the recording of this experiment, where the top yellow line and the bottom blue line on the oscilloscope are signals of the triceps and biceps, respectively.

3.10. Performance of the Muscle Patch Sensor in an Intense Condition: Soleus Muscle. To study the performance of the muscle patch sensor under a more intense condition, we attached it to the soleus muscle of a subject’s right leg. Then, its activity during stationary jumping and running was investigated. Figure 10A–D shows corresponding images of these two experiments, where white arrows indicate the location of the muscle patch sensor. It was also attached perpendicularly to the direction of muscle contractions. Figure 10E shows the experimental result of monitoring stationary jumping. The small positive peak represents the onset of jumping shown in Figure 10A, and the large negative peak represents the process of the maximum force output to jump, as shown in Figure 10B.
characteristic signal output was observed for each jump (Figure 10E), and the amplitude also changed with different jumping strengths. A Supporting Information Video S3, video-Fig10E.mp4, shows the recording of this experiment. The same experiment also was conducted for stationary running, and this is shown in Figure 10F. The small negative peak represents the moment the foot was stepping on the ground (Figure 10C), and the large negative peak represents the leg being lifted away from the ground (Figure 10D). A Supporting Information Video S4, video-Fig10F.mp4, shows the recording of this experiment. These two experiments demonstrated that the muscle patch sensor can be used for more intense exercise, and a muscle under different exercises can have different characteristic signals.

3.11. Using the Muscle Patch Sensor to Measure Breathing and Heartbeat.

The muscle patch sensor also was used for breathing and heartbeat measurement. Two ends of the muscle patch sensor were sewed on an elastic chest band, as shown in Figure 11A. Figure 11B shows the measured characteristic signal of chest wall movement during tidal breathing. The positive and negative peaks represent inhale and exhale cycles during each breathing step, which correspond to the stretching and shortening of the piezoelectric yarn. Fast Fourier transform (FFT) was used to identify the frequency of tidal breathing, and it is shown in Figure 11D. It was found that the tidal breathing was at 0.285 Hz (17.1 times/min). Furthermore, it was found that the heartbeat can be detected while holding one’s breath. Figure 10C shows the measured result, where the large positive and negative peaks are signals of inhale and exhale steps, respectively. During the breath holding period, the signal of heartbeat was observed, and the FFT result shows that the heart rate was 1.175 Hz (70.5 bpm), as shown in Figure 11E. A Supporting Information Video S5, video-Fig11C.mp4, shows the recording of this experiment. These two experiments demonstrated that the muscle patch sensor can be used for more intense exercise, and a muscle under different exercises can have different characteristic signals.

4. CONCLUSIONS

In this paper, we present our study on the orientation and structure of electrospun P(VDF-TrFE) fibers for creating a piezoelectric yarn that can have good elasticity and repeatability and low hysteretic effect. It was found that randomly oriented piezoelectric fibers had an inevitable dynamic remodeling behavior during each stretching process, which can cause a considerable drifting effect on the characteristic curves. Our study showed that this dynamic remodeling can still play an important role after a 12 h cyclic stretching process. This is a common effect for fiber-based physical sensors, and it is a serious issue in commercializing this type of sensor. To overcome this microscopic remodeling process, we increased the rotation speed of the drum collector to modify the orientation and structure of the collected fibers. We further introduced an alignment process to allow fibers to reorganize their orientation along the stretching direction and to stabilize their microstructure. It was found that nearly fully aligned and straightened fibers also were not applicable for large deformations and long-term use as these fibers can rupture easily under a large deformation. Based on our studies, we found that using a higher rotating speed of the drum collector at 1100 rpm and a 2.5 h 65% strain alignment process, a piezoelectric yarn composed of straight fibers with a small skew angle and crimped fibers with a large radius of curvature can be created. This piezoelectric yarn monitored simultaneously to study a patient’s or an athlete’s heart function.
provides a good elastic characteristic with a small hysteretic effect. It does not show a plastic deformation under a large strain, as high as 60% strain, and it can be repeatedly stretched for 12 h under a 1 Hz cyclic strain (43,200 cycles).

Having a reliable piezoelectric yarn sensor for the application of large deformations, we developed a muscle patch sensor that can be attached to the skin over human muscle for monitoring the muscle activities. It was demonstrated that the concentric and eccentric contractions of biceps and triceps can be distinguished using a current amplifier as the interface circuit. These two types of muscle contractions showed opposite biphasic peaks and can be used for studying their contraction behaviors. We also demonstrated that the profile of isometric contraction of triceps can be measured by integrating the signal output of the current amplifier. The performance of the muscle patch sensor under a more intense condition was also studied. We demonstrated that this sensor can be used for studying the contraction profiles of a soleus muscle during stationary jumping and running. It also was demonstrated that the soleus muscle had different characteristic profiles under different exercises. These studies suggests that the muscle patch sensor could be applied for athletic training, personal health monitoring, and rehabilitation. Finally, we integrated the muscle patch sensor into a chest band and verified that it can be used for measuring breathing and heartbeat, which could be applied in studying a patient’s heart function. In conclusion, we developed a method to fabricate an elastic piezoelectric yarn that can be applied for large deformation. Using this piezoelectric yarn sensor, a muscle patch sensor that can be used for monitoring human muscle activities was developed, and its feasibility to measure muscle contractions was verified.

5. MATERIALS AND METHODS

5.1. Preparation of the P(VDF-TrFE) Copolymer Solution. The piezoelectric P(VDF-TrFE) powder in a 75/25 ratio was purchased from KUREHA in Japan. The solvent used to dissolve the P(VDF-TrFE) powder was dimethylacetamide (DMAC) and methyl ethyl ketone (MEK). A solvent mixture of DMAC and MEK in a 2:3 ratio first was prepared. Then, the P(VDF-TrFE) powder was dissolved in the solvent mixture to reach 20% by weight. The powder was mixed with stirring in the solvent mixture for 12 h before the electrospinning process.

5.2. Electrospinning Setup. A commercial environmental controlled tabletop electrospinning machine (Falco Tech Co. Ltd., FES-COS) was used for this study. The humidity was maintained below 50% during the spinning process. Before the electrospinning process, the P(VDF-TrFE) copolymer solution was loaded into a glass syringe, and a syringe pump (KD Scientifc, KDS 100) was used to push this solution through the needle at a speed of 1 mL/h. A high-voltage supply (Matsusada Precision Inc., AU40N7.5) was used to apply a ~15 kV DC voltage on the needle to serve as the spinneret, and the drum collector was grounded. The surface of the drum collector was wrapped with a 50 μm thick aluminum (Al)-coated polyethylene terephthalate (PET) film for collecting the electrospun fibers. These fibers were spun onto the same location of the rotating drum collector for 20 min to make them into a thin strip. Then, the collected piezoelectric fibers on the Al coated PET film were placed in a FeCl3 solution to lift off the collected fiber strip.

The orientation and structure of the collected piezoelectric fibers under different electrospinning conditions, tensile strain, and alignment processes were investigated using SEM (Hitachi TM-300). Each condition was studied and imaged at least three times for verification. The skew angle of the piezoelectric fibers was quantified by randomly selecting 20 fibers from three different SEM images taken from different portions of a piezoelectric yarn sample.

5.3. Experimental Setup for Studying Elastic and Sensing Performances of the Piezoelectric Yarn. To study the sensing performance of the piezoelectric yarn, two silver threads were connected to two ends of the piezoelectric yarn with a separation of 1 cm, and they were connected to a BNC cable for EMI shielding. A current amplifier (Stanford Research System SR570) was used as the interface circuit, and a digital oscilloscope (Tektronix MDO3014) was used to record data. The sensitivity of the current amplifier was 50 pA/V. A one-dimensional 1 Hz cyclic tension was applied to stretch the piezoelectric yarn between 0% and up to 10−12% or 65% strain. The commercial MATLAB program was used for this signal processing and data analysis. The actual strain applied on the piezoelectric yarn was measured simultaneously by a laser position sensor (KEYENCE LK-G3S). The resultant force applied on the piezoelectric yarn was measured by a pressure sensor (QTC1074), and a poly(methyl methacrylate) level with a fulcrum at center was used to transfer tensile force into a compression force for applying on the pressure sensor.

5.4. Fabrication of the Muscle Patch Sensor. The fabrication process is summarized in the following: First, an Al−CrSk yarn was sandwiched between two 0.1 mm thick TPU sheets using thermal bonding. Two silver threads were placed on top of the yarn with a 1 cm separation, and silver paste was used to make electrical contact with the piezoelectric yarn before sealing between the two TPU sheets. The sealed piezoelectric yarn was polarized axially through these two silver threads at −1 kV for 3 h. This step was to enhance the piezoelectric response in the axial direction, and two silver wires can be connected at two ends of the yarn without interference in mechanical deformation of the muscle patch sensor. This fabricated muscle patch sensor can be attached on the skin of a muscle under detection by using two 3M double-coated tissue tapes on two ends. Thus, the piezoelectric yarn can stretch along with the muscle for real-time monitoring. The detected muscle contraction profile was picked up by a current amplifier (Stanford Research System SR570) and recorded by a digital microscope (Tektronix MDO3014). The sensitivity of the current amplifier was 50 pA/V.

5.5. Finite Element Analysis. To understand the sensing performance of cramped and skewed piezoelectric fibers, finite element simulation was conducted using COMSOL Multiphysics software. Two fibers of 20 μm length and 1 μm diameter were built in the model. The piezoelectric strain constants were \( d_{31} = 23 \text{ pm/V}, d_{32} = 8 \text{ pm/V}, \) and \( d_{33} = -33 \text{ pm/V}. \) Young’s modulus and Poisson’s ratio were 3 GPa and 0.3, respectively. The permittivity \( \varepsilon_r = 109.5 \text{ pF/m} \) and density was 1780 kg/m³. One end of the fiber was fixed, and the other end was stretched axially and sinusoidally between 0 and 50% strain while setting it free in the radial directions. The charge output during the periodic stretching process was analyzed versus applied strain. Skewed fibers with 3, 5, and 15° skew angles and cramped fibers with radii of curvature in 12.5, 6.3, and 4.2 μm were studied and compared.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.0c03309.

http://pubs.acs.org/journal/acso0f
Using two muscle patch sensors to measure the concentric and eccentric contractions of biceps and triceps simultaneously, where the top yellow line and the bottom blue line on the oscilloscope are signals of triceps and biceps, respectively (MP4).

Using two muscle patch sensors to measure the isometric contractions of biceps and triceps simultaneously, where the top yellow line and the bottom blue line are signals of triceps and biceps, respectively (MP4).

Using a muscle patch sensor to measure the force contraction of the soleus muscle during stationary jumping (MP4).

Using a muscle patch sensor to measure the force contraction of the soleus muscle during stationary running (MP4).

Sewing a muscle patch sensor on a chest band to measure the breathing and heartbeat (MP4).

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Notes

The authors declare no competing financial interest.

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