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ORIGINAL REPORT



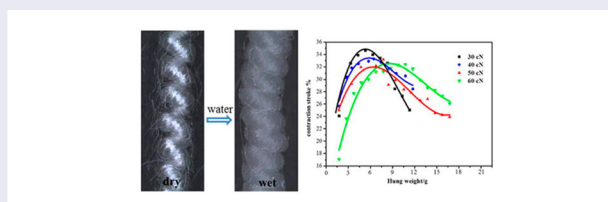
Water-responsive artificial muscles from commercial viscose fibers without chemical treatment

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ABSTRACT

Moisture change in the clothing system caused by perspiration may be used as a stimulus for smart textiles in a variety of applications. Here we report a water-responsive coil-shaped artificial muscle produced from time-proven, low-cost viscose fibers without chemical modification. Crystallinity of the viscose fiber is a key factor to the actuation performance of the final artificial muscle. The viscose fiber artificial muscle can generate a 35% contraction and a force many times greater than animal skeletal muscles of the same weight. The structural parameters and fabrication conditions were optimized to improve the performance of the coil-shaped artificial muscle.



IMPACT STATEMENT

Here we report a simple method of making water-responsive artificial muscles from commercial viscose fibers, which have a wide range of potential applications as smart building and textile materials.

ARTICLE HISTORY

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KEYWORDS

Water responsive; viscose fiber; artificial muscle

Introduction

Flexible actuators and artificial muscles are interchangeable terms for single-component material structures or devices that can reversibly contract, expand, or rotate in response to an external stimulus, similar to biological muscles. A number of artificial muscles that are actuated by the absorption and desorption of water have recently been developed [1–7]. These actuators are made from polymer composite films or carbon nanomaterials that are chemically modified to achieve hydrophilicity. Modified hydrophilic CNT fibers with hierarchical helix structures were developed to perform rapid and large contraction or rotation in response to water moisture [7,8]. Helical assemblies of modified nanowires and carbon nanotube actuators have also been reported to provide high actuation responses to electrochemical, temperature and electrical stimuli [9–17].

Perspiration is an instinctive body reaction to physical exercise, emotion, or ill health. Moisture change in the clothing system caused by perspiration may be used as a stimulus to drive textile actuators or artificial muscles. Natural cellulosic fibers, such as cotton, flax and hemp, have been used in actuators, sensors, and energy harvesting devices [18–20]. To increase the response speed of moisture-driven artificial muscles, the natural cellulosic fibers need to be chemically treated. Viscose is a regenerated cellulosic fiber widely used in apparel fabrics. Like natural cellulose, viscose fibers can absorb considerable amount of water and swell anisotropically, i.e. expanding preferentially in the radial direction. Based on this anisotropic hydro-expansion, we converted commercial viscose fibers into high-performance water moisture-cellulose artificial muscles without the need of chemical modification, which offers advantages when used in apparel.

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Materials and methods

Two types of commercial viscose yarns, a staple fiber yarn (15 tex, 955 turns/m in Z direction) and a multifilament yarn (150D/44F, twistless), were used in this investigation. The yarns were folded and then twisted on a textile twisting machine by hanging a predetermined weight at one end of the multiply yarn while the end was stopped from rotation, as illustrated in Figure S1. To obtain approximately the same linear density (150 tex) of multiply yarns, the staple fiber yarn was folded to 10-ply while the multifilament yarn was folded to 9-ply. The twist insertion rate was 1512 turns/min. When sufficient twist was inserted, the yarn started to form a spring-like coil, which was later used as water-responsive artificial muscle. The detailed experimental procedures for characterization of the coiled muscles made from viscose are provided as *Supporting Information*.

Results and discussion

Construction of artificial muscles

Compact, helical coil-structured artificial muscles were constructed by over-twisting the viscose staple fiber yarn

and the multifilament yarn. A series of left-handed helical muscles were prepared under different tensions by twisting the yarns in Z direction. Right-handed helical muscles were also prepared for comparison. Optical images of the as-prepared muscles are displayed in Figure 1. The rising angle of the helix (α) and the outer diameter (d) of the coils in the muscle, as indicated in Figure 1(B), were used to represent the helix geometry of the coils.

The tension applied to the yarn during preparation is critical to the formation of the coil structure. When the applied tension is low, a side snarl, or a loose snarl perpendicular to the general yarn direction, is formed [21]. When the tension is sufficiently high, a compact artificial muscle (cylindrical snarl) can be formed continuously. The number of twist turns required to form a compact muscle is affected by the size of the multiply yarn (measured in tex), the torsional rigidity of the yarn and the tension applied to the yarn during coil fabrication.

The length of the final muscle was determined by the original length of the yarn and the applied tension [22]. From the same length of initial yarn, more than 10cm of artificial muscles were obtained from the 10-ply spun yarn at the applied tension between 30 and 70 cN, whilst

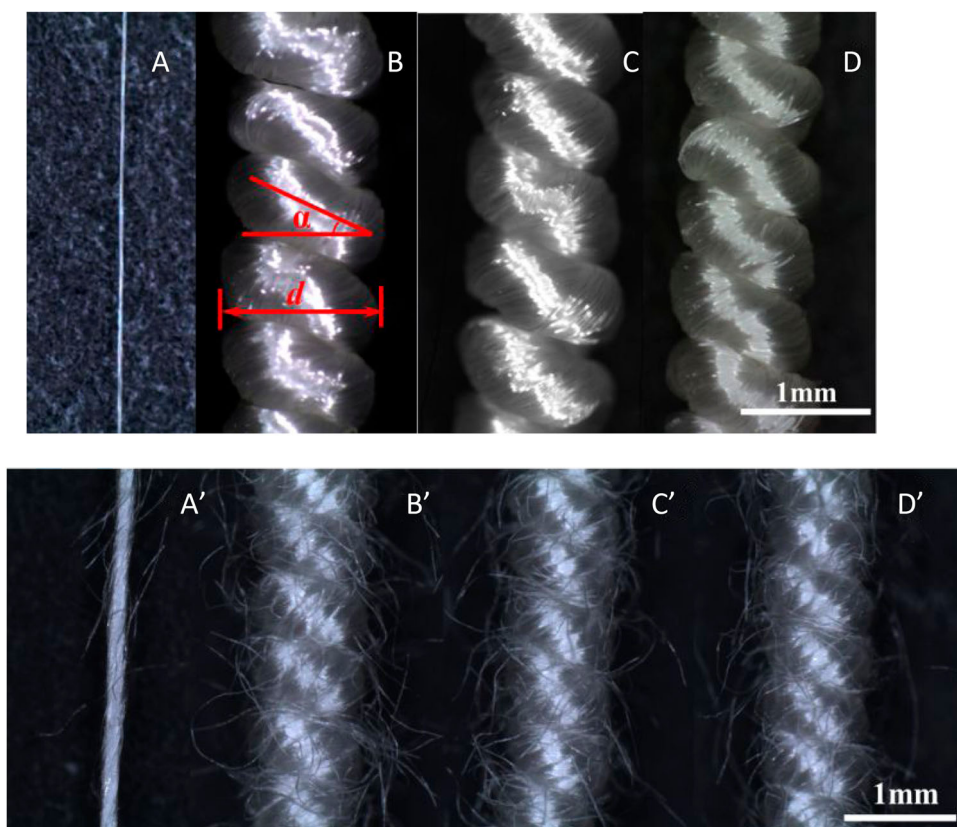


Figure 1. Images of coils formed from 9-ply viscose multifilament yarn and 10-ply staple fiber spun yarn under different tensions. (A) Original multifilament yarn; (B–D) Coils formed from 9-ply multifilament yarn at 40, 60 and 70 cN applied tension, respectively. (A') Original staple fiber spun yarn; (B'–D') Coils formed from 10-ply spun yarn under 20, 30 and 50 cN applied tension, respectively.

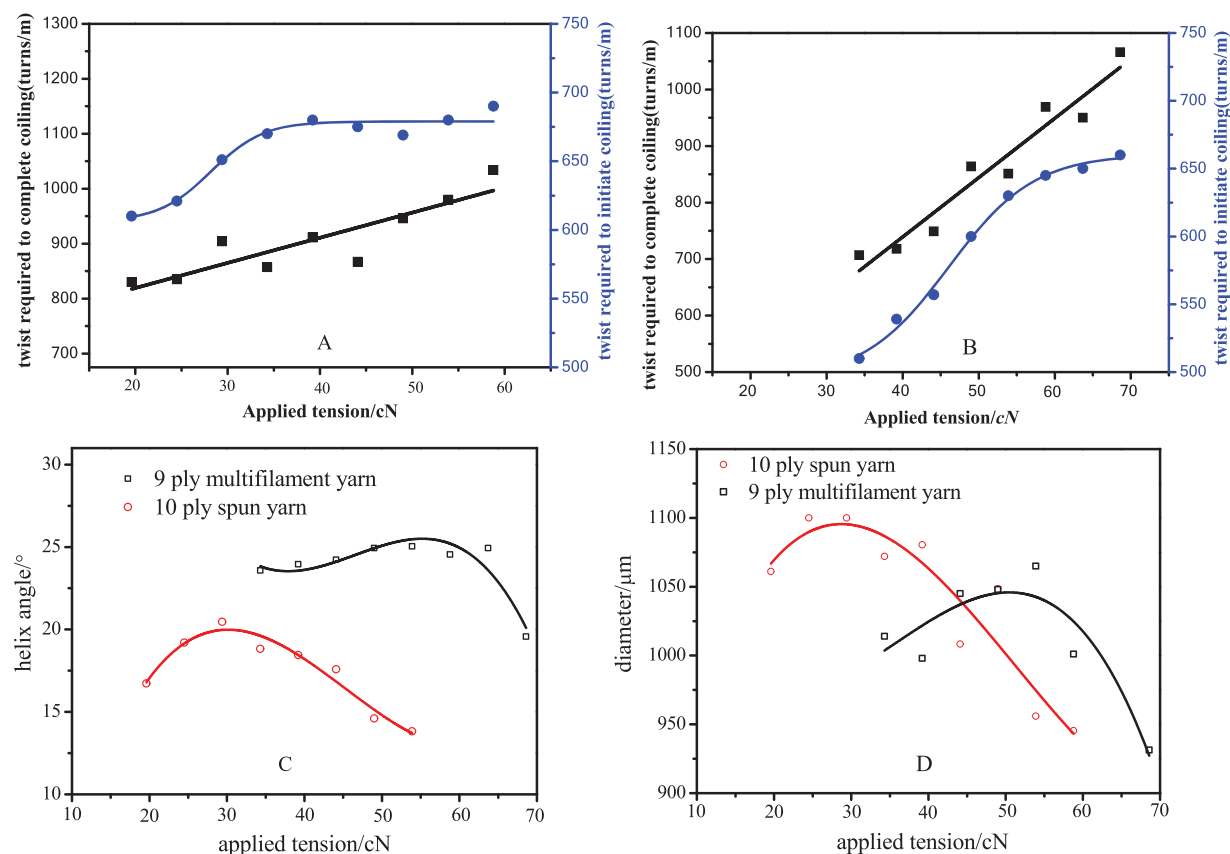


Figure 2. Effect of applied tension on the formation and geometry of coiled artificial muscles. (A) Turns of twist required to initiate coiling and to complete coiling for 10-ply staple fiber spun yarn. (B) Turns of twist required to initiate coiling and to complete coiling for 9-ply multifilament yarn. (C) Helix angle of final coiled artificial muscle as a function of applied tension. (D) Diameter of final muscle as a function of applied tension.

about 5 cm continuous muscles were formed from the 9-ply multifilament yarn at the applied tension between 50 and 70 cN. The two series of images in Figure 1 show that the helix angle and diameter of the muscles could be changed by varying the tension applied to the yarn during fabrication.

Figure 2(A,B) shows respectively the number of twist turns per meter required to initiate the formation of coil muscle and the number of twist turns per meter required to complete the formation of the muscle, plotted against the tension applied. Clearly, more twist turns were required to form a coil structure at a higher tension. At 20 cN applied tension, the 10-ply spun yarn required 610 turns/m to start coiling and 827 turns/m to complete coiling. When the tension was raised to 60 cN, the corresponding twists increased to 690 and 1034 turns/m (Figure 2(A)). At the same applied tension, lower twists were needed for the 9-ply multifilament yarn (Figure 2(B)).

On the other hand, the helix angle of the muscle formed from the 10-ply spun yarn was at a lower overall level (14.1° to 20.5°) than that formed from the 9-ply multifilament yarn (19.6° to 25°), as shown in Figure 2(C). Both the helix angle and the diameter of the coiled

muscles showed a parabolic trend, with maximum values occurring at 30 and 50 cN, respectively for the 10-ply spun yarn and the 9-ply multifilament yarn, as shown in Figure 2(D). Nonlinear relationships among ply yarn torque, tension and twist, as well as ply yarn diameter, coil diameter and the coil helix angle may be responsible for the parabolic trends in Figure 2(C,D).

Contraction of 10-ply spun yarn artificial muscle

Natural and regenerated cellulose fibers contain strongly polarized hydroxyl groups and their hydrophilic character causes the fibers to swell in diameter while contracting in length when absorbing water [23–25]. Photos of a muscle made from the 10-ply spun yarn under the tension of 40 cN before and after water actuation are shown in Figure 3(A). At its initial dry state, when a load was hung to the lower end, the muscle started to elongate, which causes gaps between neighboring turns of the coiled muscle to increase. After the muscle stopped elongating, a few drops of water were applied to the dry muscle, which caused the gaps between neighboring turns to close and the muscle to shorten, causing the weight to climb up. This gave the desired muscle action. As shown

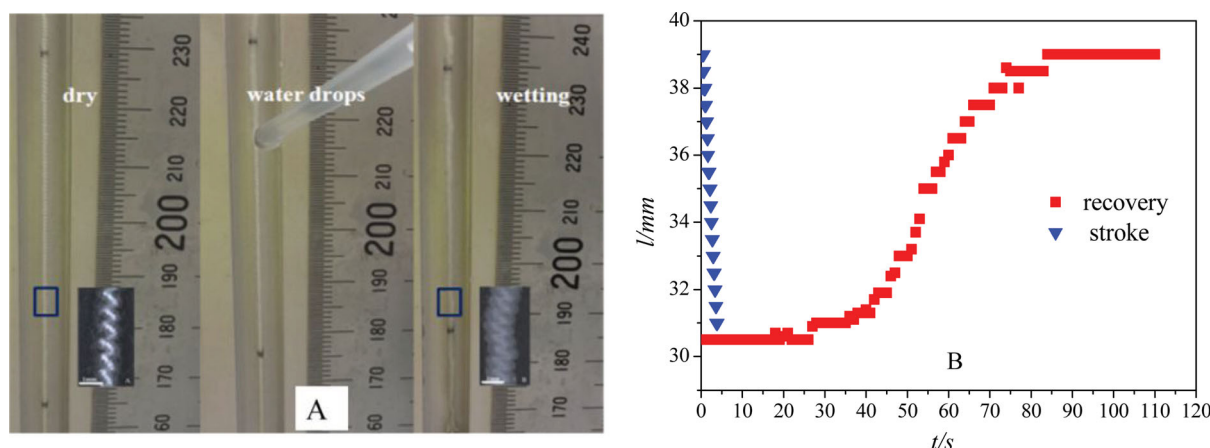


Figure 3. Contraction behavior of artificial muscle actuated by water drops. (A) Photos showing change of geometry of the 10-ply spun yarn twisted under the tension of 40 cN when wetted. (B) Trace of muscle contraction.

in Figure 3(B), with the application of a few drops of water, the muscle contracted immediately and reached its maximum contraction in about 5 s (Video S1). The length of the muscle contracted from 39 to 30.5 mm, giving a contractile strain of 0.22. Drying at room conditions took about 90 s (video S2).

At actuation, the artificial muscle increased its outer diameter by 34.3% while the number of coil turns per meter decreased by 21.3%. These changes caused the decrease of gaps between the coil turns, resulting in muscle contraction. In Figure 4(A–C), each curve represents a relationship between the stress and the contraction stroke of one muscle prepared from the 10-ply spun yarn and 9-ply multifilament yarn under a different applied tension. Clearly, the contraction stroke initially increased with the load applied to the muscle before a maximum was reached. Further increase of the applied load then caused a decrease of contraction stroke. For the muscles prepared from 10-ply spun yarn at 30, 40 and 50 cN, the maximum contract strokes achieved at a stress between 0.25 and 0.28 MPa were very similar (31%–35%) (Figure 4(A,B)). The curve for the muscle prepared from the same yarn at the highest applied tension of 60 cN displayed a shift to the right, and showed a maximum contraction of 32.8% at the contraction stress of 0.48 MPa. Figure 4(C) shows that the work capacity of the muscles increased with increasing contraction tension stress. The work capacity increased from 9.3 to 90.4 J/g as the contraction stress increased from 0.1 to 0.75 MPa for the muscle coiled under 40 cN. The maximum work capacity of 90.4 J/kg is 11 times of typical skeletal muscles (~ 8 J/kg) [26]. The 9-ply multifilament yarn displayed a much smaller contraction stroke than the muscles prepared from the 10-ply spun yarn. A maximum contraction stroke of 7.5% (based on loaded length) was achieved

at a load stress of about 0.45 MPa (Figure 4(C)). The tensile behaviors of the coiled spun yarn were studied in dry (before actuation) and wet (after actuation) states. The stress increased from 7.9 to 12.2 MPa after actuation (Figure 4(D)). After 50 cycles of actuation, there was no noticeable change in contractile stress (Figure 4(E)).

The parabolic load-contraction relationship can be interpreted as follows [27]. Under a small load, there is small or even zero space between the neighboring coils of the muscle and thus when activated, there is little or no space for the muscle to contract before neighboring coil turns contact with each other, reaching the lockup position of the muscle. Hanging a heavier load opens up a larger gap between neighboring coil turns so that the muscle could contract to a greater extent. However, when an even heavier load was used, the contraction stroke reached its peak. At this point, coil lockup is no longer the limiting factor for contraction because the energy released by wetting the muscle is not sufficient to lift the weight up to the lockup position. Therefore, further increasing the load caused a decrease of contraction stroke.

To verify the effect of the initial twist in the individual yarns to the final coiled muscle, we prepared two coiled muscles by adding Z-direction twist to the staple fiber yarn bundle in the first coiled muscle and by adding S-direction twist in the second coiled muscle, and then tested their contractions. Optical images of the coiled muscles were shown in Figure S2. The two muscles twisted to the opposite directions showed very similar contractions, 25.8% and 26.8%, respectively. Similarly, we prepared two muscles from the twistless multifilament yarns, and they demonstrated 10.7% and 12.5% contractions, less than a half of the contraction of the staple fiber yarn muscles. These results indicated that the initial

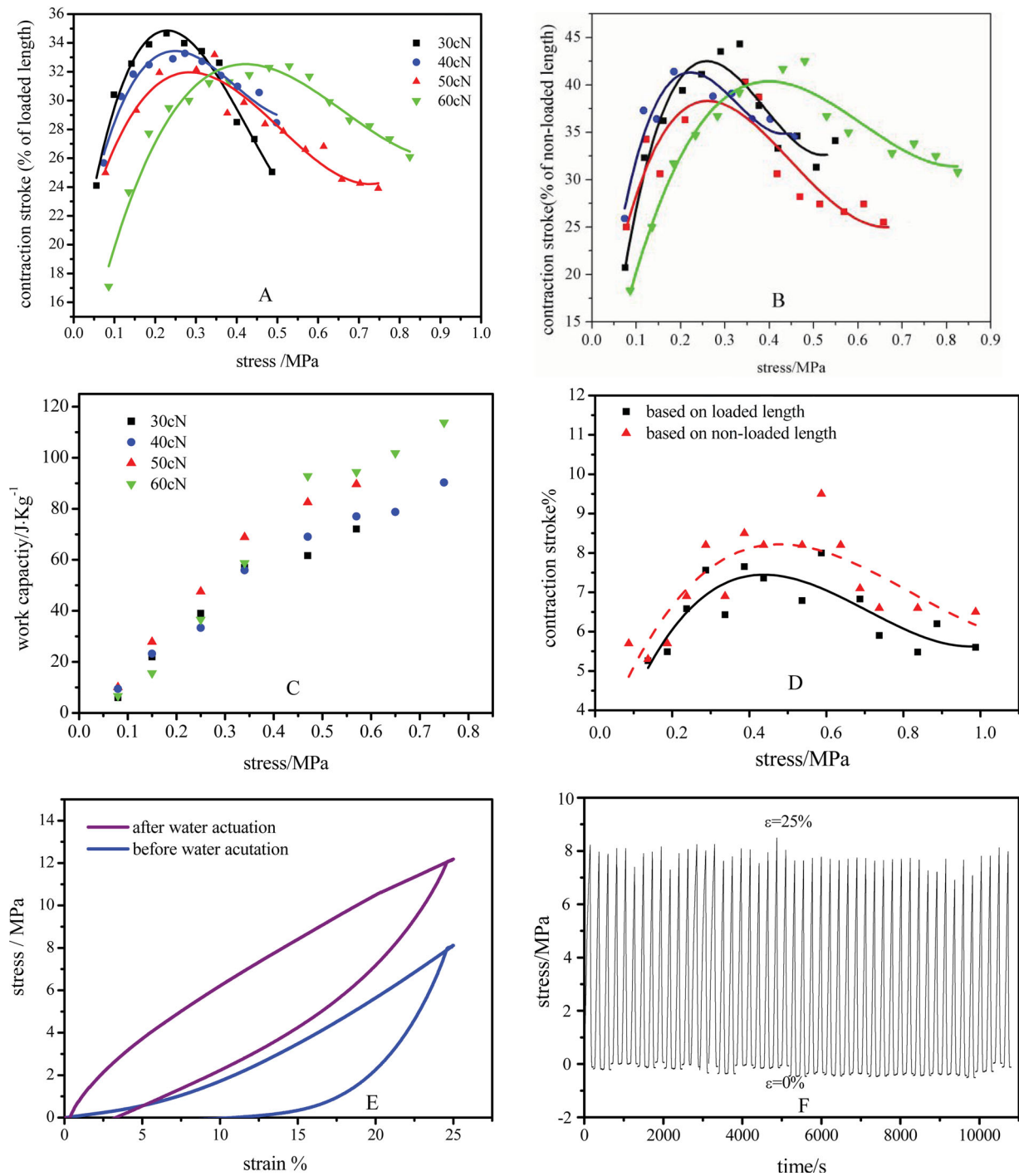


Figure 4. Effect of applied load on the contraction stroke of coiled cellulose fibers. (A) Actuation contraction stroke of muscles prepared from the 10-ply spun yarn at different applied tensions versus load stress, as a percent of loaded coiled yarn length. (B) The contraction stroke results of (A) normalized to the initial non-loaded yarn length, indicating the absolute displacement during actuation. (C) Work capacity of the 10-ply spun yarn coiled under load tension from 30 to 60 cN. (D) Actuation contraction stroke of muscle made from the 9-ply multifilament yarn as a function of hung weight. (E) Before and after water actuation Stress-strain curve of coiled 10-ply spun yarn formed under 50cN. (F) Contractive stress of 10-ply spun yarn coiled under loads of 50 cN for 50 cycles.

twist of the individual yarn in the muscles had little influence on the contractile performance. This is because, in preparing the coils, several (9 or 10) yarns were plied together and then twisted to make one coil, so the twist

in the individual yarns is 'locked up' and 'insulated' from the twist added to the ply, which is in a higher structural level. In other words, adding opposite twist to the ply does not remove the twist in the individual yarns.

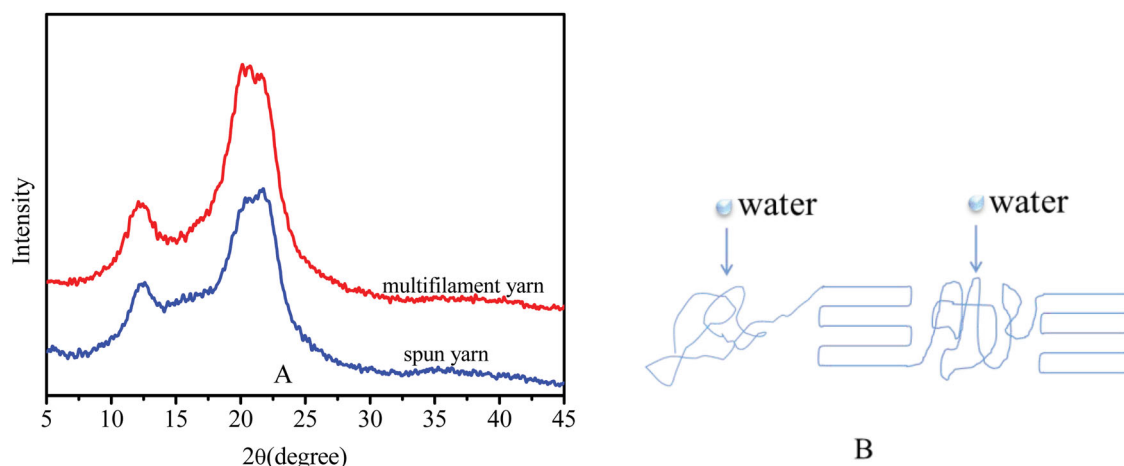


Figure 5. (A) XRD patterns of multifilament yarn and spun yarn. (B) Schematic diagram of water actuation in the amorphous region.

Figure 4(D) and Tables S1 and S2 show that the muscle made from the 9-ply multifilament yarn generated a much smaller contraction stroke and work capacity than the muscle made from the 10-ply spun yarn. This difference between the two types of artificial muscles may be explained by the crystallinity of their constituent fibers. The XRD patterns of the staple fiber spun yarn and the multifilament yarn are displayed in Figure 5(A). Typical diffraction peaks appeared at $2\theta = 12.4^\circ$ and 20.7° . The peak positions suggest that the crystal structures of both two types of fibers are cellulose III [28]. Crystallinity of the fibers was calculated from the curves using peak fitting method. The multifilament yarn showed a crystallinity of 42% while the staple fiber spun yarn showed a much lower crystallinity of 19%. It is known that crystallinity of cellulose fibers has an important impact on their water absorbency and their degree of dimensional expansion (swelling) [29,30]. In crystalline regions, the cellulose molecules are closely packed in a regular pattern. When a cellulose fiber comes in contact with water, water diffuses into the fiber, at first mostly into its amorphous domains because of their relatively high disorder [23,24], as illustrated in Figure 5(B) and Figure S3. This means that the low crystallinity staple fiber will swell considerably more than the high crystallinity multifilament. The highly twisted coil structure transforms the fiber dimensional expansion into muscle contraction, as described in our previous work [29]. This explains why the contraction capacity of the muscle made from the 10-ply staple fiber spun yarn was much larger than that of the muscle made from the 9-ply multifilament yarn.

Applications

Several applications of moisture-actuating artificial muscles made from synthetic materials have been suggested,

such as humidity switches [2] and smart windows that respond to weather conditions [4,8]. In our previous work, we demonstrated that the artificial muscles made from wool, cotton, and flax fibers could be used in smart textiles for releasing body heat [29]. Jia et al. [3] recently demonstrated applications of moisture-responsive artificial muscles made from silk yarns, including a robotic ‘caterpillar’ walking on a barbed wire, a retractable sleeve and a woven fabric that changes the spaces between weft yarns or warp yarns. Natural cotton, flax, wool and silk fibers have hydrophobic surfaces that slow down the movement of water from the surface into the center of the fibers. Therefore, they were treated chemically to increase the contraction speed of the artificial muscles. The viscose fibers used in this study are regenerated cellulose fibers that do not have a hydrophobic surface so that the artificial muscles made from the viscose fibers can generate contraction without any chemical treatment when used in the applications that were demonstrated in previous works.

Conclusion

Helical coil-structured artificial muscles responsive to water were fabricated by inserting extremely high twist to regenerated cellulose viscose fiber yarns. When actuated by water, the muscles could achieve more than 30% contraction. The fabrication method is simple and uses widely available textile machines and low-cost commercial fibers without further chemical modification. The viscose fiber artificial muscles have potential for many applications, including smart building materials and textiles.

Disclosure statement

No potential conflict of interest was reported by the author(s).

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