ARTIFICIAL MUSCLES

Sheath-run artificial muscles

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Although guest-filled carbon nanotube yarns provide record performance as torsional and tensile artificial muscles, they are expensive, and only part of the muscle effectively contributes to actuation. We describe a muscle type that provides higher performance, in which the guest that drives actuation is a sheath on a twisted or coiled core that can be an inexpensive yarn. This change from guest-filled to sheath-run artificial muscles increases the maximum work capacity by factors of 1.70 to 2.15 for tensile muscles driven electrothermally or by vapor absorption. A sheath-run electrochemical muscle generates 1.98 watts per gram of average contractile power—40 times that for human muscle and 9.0 times that of the highest power alternative electrochemical muscle. Theory predicts the observed performance advantages of sheath-run muscles.

Remarkable performance has been obtained for tensile and torsional carbon nanotube (CNT) hybrid yarn muscles (1-5), whose actuation is driven by the volume change of a guest within a twisted or coiled CNT yarn. During thermally powered contraction, coiled hybrid muscles can deliver 29 times the work as the same weight human muscle (1). Changing the structural relationship between guest and host will provide major performance increases and the ability to replace expensive CNT yarn with cheap commercialized yarns.

CNT hybrid yarn artificial muscles (HYAMs) can be made by inserting twist, or twist and coiling, into a guest-filled CNT yarn. Muscles that are twisted (but not coiled), called twisted muscles, are mainly useful for torsional actuation. High inserted twist results in coiled muscles that can deliver larger strokes than can human muscles (*I*).

Polymer fiber and yarn muscles are known (6-II) that operate similarly to CNT HYAMs: Muscle volume expansion drives muscle untwist, which produces both torsional and tensile actuation. These thermally driven polymer muscles are cheap because they can be made by inserting

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extreme twist into fishing line or sewing thread. Other twisted or coiled materials have been exploited as fiber-like muscles, such as graphene oxide fiber (*12*), shape memory polymer fiber or metal alloy yarn (*13*, *14*), cotton yarn composites (*15*), carbon fiber/polydimethylsiloxane yarn (*16*), neat CNT yarns (*1*, *17–20*), and spider-silk dragline (*21*). CNT HYAMs are especially useful because guest choice results in muscles driven thermally (*1*, *4*), electrochemically (*22*, *23*), or by absorption (*2*, *3*, *24*).

The challenge is to develop a fundamentally new host-guest structure that eliminates the liabilities of CNT HYAMs. First, the ability of guest expansion to drive yarn untwist depends on the yarn's bias angle (the angle between the yarn length and the nanotube direction). Because this angle decreases to zero on going from the yarn surface to the yarn center, the input energy delivered to the guest near the yarn center is not effectively used. Second, muscle mechanical power is limited by the chemical or thermal transport times to access yarn volume.

Here, we describe a new muscle structure that addresses each of these problems. Rather than infiltrating a volume-changing yarn guest within a yarn, such as for a HYAM, this guest is coated as a yarn sheath. Because the dimensional and modulus changes of this sheath drive actuation, we call the resulting actuators "sheath-run artificial muscles" (SRAMs).

CNTs were drawn as a sheet from a CNT forest and twisted into the Archimedian yarn (fig. S1) (25, 26) used as muscle core. SRAMs were fabricated (Fig. 1A) by drawing a vertically suspended, torsionally tethered twisted yarn through a large droplet of polymer solution multiple times to achieve the targetted sheath thickness of dried polymer. The solvent used was chosen to avoid polymer infiltration into the twist-densified core yarn and provide a sharp interface between sheath and core (Fig. 1C and figs. S2A, S3, and S7, C to F). Scanning electron microscope (SEM) measurements provided the sheath/core ratio (SCR; the ratio of sheath thickness to the interior yarn diameter). To demonstrate that CNT yarns can be replaced with inexpensive yarns, we evaluated commercial nylon 6, silk, and bamboo yarns as the muscle core as well as electrospun polyacrylonitrile (PAN) nanofibers.

The nomenclature used for a sheath X on a yarn core Y of a SRAM or an X guest inside a HYAM yarn Y is X@Y. Hence, PEO-SO₃@CNT denotes a PEO-SO3 guest and a CNT yarn host, where PEO-SO₃ is a blend of poly(ethylene oxide) and a copolymer of tetrafluoroethylene and sulfonyl fluoride vinyl ether (26). The yarn-biasangle dependence of the minimum SCR needed to prevent sheath cracking for a PEO-SO3@CNT yarn is shown in fig. S7; this ratio approximately maximizes torsional stroke for the high targeted yarn bias angle. For comparative studies, the guest/ host weight ratio was essentially the same for the SRAM and HYAM, and the same mechanical load was applied during twist insertion. HYAMs were made by using the above droplet method by adding polymer solution to a low-twist yarn, partially drving the solution to a gel-like state and then adding additional twist to equal that of the SRAM. If the guest/host weight ratio is too high for a HYAM (26), the guest will extrude from the host yarn during twist insertion (fig. S12B).

"Self-coiled" yarn was fabricated by inserting further twist while the guest was in the gel state (Fig. 1B). To increase yarn stroke by increasing the spring index, twisted yarns or self-coiled yarns (Fig. 1, D and E) were coiled or supercoiled by wrapping around a mandrel. Afterward, the coiled yarn was thermally annealed (*26*). When describing a muscle, the diameter is for the dry, twisted muscle before coiling. Unless otherwise described, gravimetric work and power densities are normalized to the weight of the dry muscle. The spring index is the ratio of the difference in outer coil diameter and the fiber diameter to the fiber diameter, where a fiber's diameter is its width in its largest lateral dimension.

Torsional actuation of a one-end-tethered SRAM is illustrated in Fig. 2A. Unless otherwise noted, an equilibrium vapor pressure was delivered to muscles in flowing dry air and then removed under vacuum, using the glass tube system of Fig. 2B. For performance comparisons, a 60-mgweight paddle at the yarn end, with 0.28 kg·mm² moment of inertia, was used to characterize torsional rotation angle and speed. Also, the SRAMs and HYAMs were made from identical yarn, contained the same inserted twist, and had nearly the same host/guest weight ratio.

In Fig. 2B, we compare the time dependence of paddle rotation and speed for a PEO-SO₃@CNT SRAM and HYAM and a pristine CNT muscle that are undergoing one complete reversible cycle of ethanol vapor-powered actuation. The peak stroke and peak rotation speed for the SRAM [143°/mm and 507 rotations per minute (rpm)] are about twice that for the HYAM (76°/mm and 254 rpm) and much larger than for the pristine yarn (4.7°/mm and 36 rpm). Steady-state measurements of torsional stroke versus weight % (wt %) ethanol in the muscles (Fig. 2C) show that

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the ratio of torsional strokes for a PEO-SO₃@CNT SRAM to a PEO-SO₃@CNT HYAM peaks at 6.7 for 4.1 wt % ethanol and then gradually decreases to 1.7 for 17.5 wt % ethanol. The small hysteresis in torsional strokes for the SRAM and HYAM means that both could reliably open and close valves in response to absorbed vapor. However, the torsional stroke of the SRAM is much more sensitive to the amount of absorbed ethanol than the HYAM.

As shown in Fig. 2D, PEO-SO₃@CNT SRAMs and HYAMs reversibly actuate over 3000 cycles of ethanol absorption and desorption, despite the absence of tethering. This reversibility results because the guest acts as a torsional return spring. By contrast, the torsional stroke of the pristine yarn rapidly decreases from 27°/mm for the first cycle to ~4.7°/mm on the 27th cycle, thereafter stabilizing at this value for the next ~3000 cycles.

Our theoretical model (26) predicts the dependence of torsional stroke on the SCR by using the torque balance between sheath and core, before and after actuation. This analysis captures the two primary mechanistic contributions to SRAM torsional actuation: sheath swelling and sheath softening, which combine to partially release elastically stored torsional energy in the core yarn. In Fig. 2E, we compare the observed and predicted dependence of torsional stroke on SCR for an ethanol-driven PEO-SO3@CNT SRAM made from a 42°-bias-angle yarn. The maximum torsional stroke (143°/mm) occurs for a SCR of 0.14, which agrees with the predicted 151°/mm stroke maximum for a SCR of 0.15. A much lower SCR cannot maintain the initially inserted twist before actuation, and a much higher SCR ratio hinders twist release during actuation. As shown in fig. S6, the torsional stroke is near maximum for yarn bias angles from 38° to 43° for a PEO-SO₃@CNT SRAM that has a sheath/core weight ratio of 0.53.

Torsional stroke is sensitive to vapor type (fig. S8) and the sheath's ability to swell and soften by vapor absorption. Because ethanol produces a much larger equilibrium volume expansion in PEO-SO₃ (16.7%) than in polyvinyl alcohol (PVA) (1.3%) or nylon 6 (0.5%) (fig. S5A), the torsional stroke of a CNT core SRAM was much larger for a PEO-SO₃ sheath (143°/mm) than for PVA (22°/mm) or nylon 6 sheaths (11°/mm) (fig. S5B).

High performance resulted for ethanol-powered torsional SRAMs in which the expensive CNT yarn was replaced with a silk or electrospun PAN yarn (Fig. 2F and fig. S2B). The bias angles of these SRAMs (30° and 18°, respectively) are lower than for the CNT yarn core SRAM (42°) because higher twist broke the yarns. The lower bias angles and larger diameters of the PAN and silk core yarns provided smaller equilibrium torsional strokes (123 and 70°/mm, respectively) than for the PEO-SO3@CNT SRAM (143°/mm). However, using the invariance of the product of torsional stroke and yarn diameter when the yarn's bias angle is constant (1) and results in fig. S6 for the biasangle-dependence of torsional stroke for a PEO-SO₃@CNT SRAM, the torsional strokes of a PEO-SO3@PAN SRAM and a PEO-SO3@silk SRAM are predicted to be close to those for PEO-SO₃@CNT SRAMs that have the same core bias angle and diameter (26).

All measurements show that a SRAM has important performance advantages over the corresponding HYAM as a torsional actuator. The ratios of peak torsional speed of the SRAM to that of the corresponding HYAM are nearly the same for PEO-SO₃@CNT (1.75), PEO-SO₃@silk (1.74), and PEO-SO₃@PAN (1.79) muscles powered by ethanol vapor and close to that for water vapor-powered nylon6@CNT muscles (1.86) (Fig. 2, B and F, and figs. S2 and S9). However, greater variation arises in the ratios of peak stroke for the SRAM to that of the HYAM (1.86, 1.67, 1.36, and 1.63, respectively, for the above).

By adding sufficient additional twist to the muscles used for torsional actuation, fully coiled yarn muscles result that provide large strokes. By comparing the performance of coiled muscles made from yarns with nearly the same host and guest weight per yarn length, we demonstrate the increases in stroke, stroke rate, contractile work, and contractile power that results from transitioning from a HYAM to a SRAM.

The torsional rotor was replaced with a heavy, nonrotating weight when changing from torsional to tensile actuation. Allowing weight rotation decreases tensile contraction (fig. S10) because yarn untwist increases muscle length. When ethanol vapor-driven, a PEO-SO₃@CNT SRAM delivered a higher stroke for all loads and times than did a HYAM (Fig. 3, A and B, and fig. S11). One thousand fully reversible cycles were demonstrated. Corresponding SRAM structure changes during 0.1 Hz actuation to provide 8.5% stroke are shown in movie S1. As shown in fig. S12A, the equilibrium isometric contractile stress generated by a PEO-SO₃@CNT SRAM monotonically increases with increasing ethanol vapor concentration. By contrast, if the applied load is low and the change in sheath thickness is large, the SRAM first contracts until intercoil contact occurs and then expands as intercoil contact drives actuation (fig. S12C). Mandrel coiling greatly amplifies muscle stroke. A 70% tensile stroke was obtained for a humidity-driven cone-mandrel SRAM, and this SRAM provided faster contraction than that of cylindrical-mandrel SRAMs that were coiled and supercoiled (fig. S13).

The SRAMs provide advantages in contractile work capacity and maximum average contractile power (Fig. 3B, figs. S14 to 16 and S19, and table S2), which is the maximum ratio of contractile work to actuation time. The maximum average contractile power output was 4.44 W/g for the ethanol vapor-driven PEO-SO3@CNT SRAM and 1.51 W/g for the corresponding HYAM. The loadoptimized contractile work capacity and the maximum average power density of coiled SRAMs are higher than for coiled HYAMs at all applied loads for sorption-driven, electrothermal, and electrochemical actuation (table S2). For loads maximizing equilibrium contractile work capacities, the SRAM-to-HYAM work capacity ratio was 1.84 for ethanol vapor-driven PEO-SO3@CNT muscles (Fig. 3B), 1.73 for electrothermally driven PEO-SO3@CNT muscles (Fig. 3D), and 2.15 for electrothermally driven PU@CNT muscles (Fig. 4D), where PU is an elastomeric polyurethane (26). These SRAM-to-HYAM work capacity ratios will approximately equal the ratio of energy conversion efficiencies for sorption-powered muscles in which the equilibrium gravimetric sorption of guest in SRAM sheath and in HYAM core are equal, and for thermal muscles in which the



Fig. 1. Muscle fabrication and structure for torsional and tensile actuation. (A) Schematic lateral and cross-sectional views of a twisted CNT yarn and a SRAM, made by coating a twisted CNT yarn with a polymer sheath. (B to E) SEM micrographs for PEO-SO₃@CNT muscles. (B) A SRAM made by self-coiling a sheath-coated twisted yarn. (C) The surface of a twisted SRAM, which was broken by untwisting in liquid N₂, showing the distinct boundary between sheath polymer and CNT core. (D) A mandrel-coiled twisted SRAM. (E) A SRAM that was self-coiled and then supercoiled around a mandrel. Scale bars, (B) to (E), 35, 15, 200, and 200 µm, respectively.

differences in heat lost during high-rate contractile work are negligible.

The SRAM-to-HYAM power density ratio (table S2) is higher for ethanol vapor-driven PEO-SO₃@CNT muscles (2.94) than for electro-thermally driven PEO-SO₃@CNT muscles (1.69) and PU@CNT muscles (2.06). This is likely because the power density ratio for the vapor-driven muscle is enhanced by both the larger

equilibrium work capacity of the SRAM and the more rapid vapor absorption, and the latter diffusion-based enhancement term disappears when actuation is from electrothermally heating the CNT yarn.

Because the rate of cooling is faster for the SRAM than for the HYAM and the rate of cooling has the greatest impact on full cycle performance, the high-frequency work capacity during





electrothermal actuation is much higher for a SRAM than a HYAM. The PEO-SO₃@CNT SRAMs electrothermally operated in air and in room-temperature water to produce 2.6 W/g (for 3.2% stroke at 9 Hz) and 9.0 W/g (for 5.5% stroke at 12 Hz), respectively, of full-cycle contractile power (fig. S15, C to F), which is much higher than the typical contractile power of human natural muscle (0.05 W/g, *5*). When operated in air, this SRAM muscle provided a stoke of 8.0% at 2 Hz, corresponding to a power density of 1.2 W/g. Shown in movie S2 is the electrothermal actuation of a coiled PEO-SO₃@CNT SRAM in water at 12 Hz to generate a 5% stroke and a full-cycle contractile power of 4.2 W/g.

We next predicted the stress dependence of tensile stroke and contractile work capacity for ethanol-powered actuation of coiled PEO-SO₃@CNT SRAMs and HYAMs (fig. S21) (26). This analysis used the above theoretically derived torsional strokes of twisted, noncoiled muscles, the relationship between torsional stroke (ΔT) and tensile stroke for noncontacting coils if muscle stiffnesses were constant, and the dependence of PEO-SO₃ modulus on ethanol absorption (fig. S4A). Remarkable agreement was obtained between theory and experiment for the stress dependence of equilibrium stroke and contractile work capacity without using a fitted parameter. The observed ratio of the maximum contractile work capacity of the SRAM to that of the HYAM is 1.70, which is close to the predicted 1.52 (fig. S22).

Electrochemically powered artificial muscles have key advantages over thermally powered muscles: (i) Their efficiency is not limited by the Carnot efficiency, and (ii) they have a natural latching state, meaning that stroke can be maintained without the input of substantial electrical energy. A conventional electrochemical CNT yarn muscle is a HYAM, in which the yarn guest is the electrolyte.

A CNT@nylon6 SRAM was made with the process shown in Fig. 4A, right. Similar to a process used to make coiled CNT yarns for energy harvesting (27), a stack of CNT sheets was formed into a cylinder (Fig. 4A, left). A nylon yarn was placed in the center of the cylinder. Initially, twist is inserted only into the CNT cylinder. However, once the CNT cylinder collapses to form a sheath on the nylon 6 yarn, torque automatically transfers from this sheath to the yarn, enabling the yarn to become fully coiled.

The electrolyte-filled CNT sheath of the SRAM and the electrolyte-filled volume of the HYAM provide electrochemical actuation because of volume changes produced by electrochemical double-layer injection of anions and cations. For the used electrolyte of 0.2 M tetrabutylammonium hexafluorophosphate (TBA·PF₆) in propylene carbonate, the calculated van der Waals volume (28) of the TBA⁺ cation (~293 Å³) is much larger than for the PF₆⁻ anion (69 Å³). Potential scans (Fig. 4B) for the SRAM and HYAM show that muscle contractions increase on both sides of the potential of zero charge and that the contraction is proportional to the volume of the injected ion. These contractions are largest for the SRAM.





Fig. 3. Isobaric tensile actuation of self-coiled, sorption-powered, and electrothermally powered SRAMs, HYAMs, and pristine CNT yarns. (A) Tensile stroke versus time for a PEO-SO₃@CNT SRAM and HYAM and a pristine yarn when actuated by ethanol absorption by using the illustrated configuration and 33 MPa stress. Sorption was from a near-equilibrium ethanol concentration in dry air and desorption was by means of dynamic pumping. Before coiling, the diameters of the PEO-SO₃@CNT SRAM and HYAM and the pristine yarn were 43, 47, and 38 μ m, respectively. (B) Tensile stroke and contractile work capacity

versus applied stress for the sorption-actuated muscles of (A). (**C**) The time dependence of tensile stroke for a PU@CNT SRAM and HYAM and a pristine CNT yarn when electrothermally actuated by using the illustrated configuration, 42 MPa stress, and 0.25 W/cm power, which provided temperatures of 85°, 93°, and 97°C, respectively. (Left) The device structure. Before coiling, the diameters of the PU@CNT SRAM and HYAM and HYAM and the pristine yarn were 65, 71, and 51 µm, respectively. (**D**) Tensile stroke and contractile work capacity versus applied stress for the electrothermally actuated yarns in (C).

Because the electrical energy required for actuation increases with increasing amount of electrochemically accessible CNTs, the contractile work per weight of CNT is an important performance metric. For slow square-wave switching at 10 mHz between 0 and -3 V (Fig. 4C), the load-maximized contractile work capacity is slightly higher for the CNT@nylon6 SRAM (2.35 J/g) than for the CNT HYAM containing the same CNT weight per yarn length (2.01 J/g). However, for more practically applicable actuation rates (fig. S17), the ratios of SRAM to HYAM work capacities for similar tensile loads are much more impressive. For an applied square-wave frequency of ~0.3 Hz, this ratio is ~3.4 for all applied loads. At the highest measured frequency (5 Hz) and the highest applied load, this ratio is 14.6. The electrochemical actuation of a coiled CNT@nylon6 SRAM to provide 14.3% stroke at 0.25 Hz, while lifting a heavy load, is shown in movie S3.

The frequency dependences of work capacity for a coiled CNT@nylon6 SRAM and a coiled CNT HYAM are shown in Fig. 4D for squarewave voltages between 0 and –3 V. For 1 Hz cycle frequency, the tensile stroke, work per cycle, and average contractile power density for the SRAM were, respectively, 4.7%, 0.99 J/g, and 1.98 W/g, as compared with 0.90%, 0.11 J/g, and 0.22 W/g for the HYAM. The high performance obtained for the SRAM at relatively high frequencies expands the application possibilities for electrochemical artificial muscles.

The contractile energy conversion efficiencies were obtained for optimized voltage scan rates between 0 and -2.7 V. This peak efficiency increased from 2.96% at 80 mV/s scan rate for the CNT yarn muscle to 4.26% at 130 mV/s scan rate for the SRAM (Fig. 4E). Using a higher potential scan rate for both muscles (200 mV/s) (fig. S18), which increased stroke rates, provided a SRAM efficiency (3.8%) that is 2.7 times the HYAM efficiency (26).

Because the SRAM technology enables replacement of expensive CNT yarns with inexpensive, commercially available polymer yarns whose sheath responds to targeted ambient variables, they are attractive for intelligent structures (29). Relevant for possible use in comfort-adjusting clothing, SRAMs were knitted into a textile that increased porosity when exposed to moisture, and flat-coil SRAMs were demonstrated (figs. S23 to S25). Analyte-powered sensors that intelligently respond in the body to open and close valves that release drugs in response to antigens (*30*) or biochemicals such as glucose (*31*) are other possibilities. A CNT-free SRAM that linearly contracts with increasing glucose concentration was demonstrated (fig. S26), which could squeeze a pouch to release a drug (fig. S20).

The 5.2-, 9.0-, and 9.0-fold advantages at 1 Hz of the SRAM over the corresponding HYAM in electrochemical stroke, contractile work-per-cycle density, and average contractile power density, respectively (Fig. 4D), are important for electrically powered robotic devices in which stroke should be maintained without consuming substantial electrical energy. Electrothermal PEO-SO₃@CNT SRAMs operated in air and in room-temperature water to produce 2.6 W/g (at 9 Hz) and 9.0 W/g (at 12 Hz) of full-cycle contractile power, respectively, compared with the 0.05 W/g typical of human



Fig. 4. Fabrication and electrochemical tensile actuation of coiled CNT@nylon6 SRAM and coiled CNT HYAM yarns in 0.2 M TBA·PF₆/PC electrolyte. (A) Illustration of (left) cone spinning for fabricating CNT yarns and (right) its modification for making SRAM yarns. SEM micrographs of a coiled pristine yarn, a coiled CNT@nylon6 SRAM yarn, and a noncoiled nylon 6 yarn are shown. (B) Tensile stroke of the SRAM and HYAM during a cyclic voltammetry scan at 20 mV/s, under 22 MPa isobaric stress. (Inset) Actuator stroke at this load for this muscle versus interelectrode voltage scan rate. (C) Tensile stroke and contractile work capacity versus load when applying a 10-mHz square-wave voltage between 0 and -3 V. The spring indices of the 95-µm-diameter

muscle (5). SRAM performance and realizable low cost suggests their use for diverse applications, from fast, powerful muscles for humanoid robots and exoskeletons to intelligent comfortadjusting clothing and drug-delivery systems.

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0.56, respectively. (D) The frequency dependence of work capacity for a

between 0 and -3 V. For 1 Hz cycle frequency, the tensile stroke, workper-cycle, and average contractile power density for the SRAM at the highest

loads were 4.7%, 0.99 J/g, and 1.98 W/g, compared with 0.90%, 0.11 J/g,

and 0.22 W/g for the HYAM. (E) The scan rate dependence of work capacity

and energy conversion efficiency for the SRAM and HYAM, using an applied

indices of the 87-µm-diameter CNT@nylon6 SRAM and the 79-µm-diameter

stress of ~30 MPa for the SRAM and HYAM. For (D) and (E), the spring

CNT HYAM were 0.97 and 0.67, respectively.

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needed to evaluate the conclusions in the paper are present in the paper or the supplementary materials.

SUPPLEMENTARY MATERIALS

science.sciencemag.org/content/365/6449/150/suppl/DC1 Materials and Methods Supplementary Text Figs. S1 to S26 Tables S1 and S2 References (32–37) Movies S1 to S3

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Getting the most out of muscles

Materials that convert electrical, chemical, or thermal energy into a shape change can be used to form artificial muscles. Such materials include bimetallic strips or host-guest materials or coiled fibers or yarns (see the Perspective by Tawfick and Tang). Kanik *et al.* developed a polymer bimorph structure from an elastomer and a semicrystalline polymer where the difference in thermal expansion enabled thermally actuated artificial muscles. Iterative cold stretching of clad fibers could be used to tailor the dimensions and mechanical response, making it simple to produce hundreds of meters of coiled fibers. Mu *et al.* describe carbon nanotube yarns in which the volume-changing material is placed as a sheath outside the twisted or coiled fiber. This configuration can double the work capacity of tensile muscles. Yuan *et al.* produced polymer fiber torsional actuators with the ability to store energy that could be recovered on heating. Twisting mechanical deformation was applied to the fibers above the glass transition temperature and then stored via rapid quenching.

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