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Sheath-run artificial muscles

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Abstract
Although guest-filled carbon nanotube yarns provide record performance as torsional and tensile artificial muscles, they are expensive, and only part of them 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.

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Sheath-Run Artificial Muscles

While guest-filled, twisted and coiled carbon nanotube yarns provide record performance as torsional and tensile artificial muscles, they are expensive and only part of the yarn structure effectively contributes to actuation. We report an alternative topology that provides higher performance, wherein the guest that drives actuation is a sheath on a twisted or coiled core that can be an ordinary silk or electrospun polyacrylonitrile yarn. This topology change to a sheath-run artificial muscle increases the maximum contractile work capacity by factors between 1.73 and 2.20 for muscle that are driven electrothermally, electrochemically, or by vapor-absorption. A sheath-run electrochemical muscle provides an average contractile output mechanical power density of 2.24 W/g, which is 45 times higher than the typical power density of natural muscle and 1.75 times that for an electrolyte-filled carbon nanotube yarn. Demonstrated applications of sheath-run coiled muscles are for comfort-adjusting textiles, which respond to sweat by increasing porosity, and for yarns that actuate in response to the presence of an antigen to potentially release an antibiotic. Theoretical analysis quantitatively explains the performance advantage of sheath-run muscles.

Remarkable performance has been obtained for tensile and torsional carbon nanotube hybrid yarn muscles (1), whose actuation is driven by the volume change of a guest that is within a twisted or coiled carbon nanotube yarn host. During thermally-powered contraction, coiled hybrid muscles can provide 29 (1.36 J/g at 84 MPa) times the work and generate 85 (27.9 W/g) times the power as the same weight natural muscle (1). By changing the topological relationship of guest and host, major performance increases will be obtained, as well as the ability to replace expensive carbon nanotube yarns with cheap, commercially available yarns.

Carbon nanotube hybrid yarn artificial muscles (HYAMs) are made by inserting twist, or both twist and coiling, into a guest-filled carbon nanotube (CNT) yarn. Muscles that are twisted (but not coiled), which are called twisted muscles, are principally useful for torsional actuation. Extremely high inserted twist results in coiled muscles that can deliver tensile strokes exceeding those of nature’s skeletal muscles (1).

Polymer fiber and yarn muscles are also known (2,3,4) that operate similarly to CNT HYAMs - expansion of muscle volume drives muscle untwist, and this untwist produces both torsional and tensile actuation. These thermally or electrothermally driven polymer muscles
are cheap, since they can be inexpensively made by inserting extreme twist into high strength fibers or yarns used for fishing line and sewing thread. Other materials have also been exploited as fiber-like muscles, such as graphene oxide based yarn, twisted shape memory yarn, liquid crystal polymer fiber, carbon fiber/polydimethylsiloxane coiled yarn and twisted spider-silk dragline. However, CNT HYAMs have the advantage that changes in yarn guest can convert them from being thermally or electrothermally driven (1) to being driven by absorption (5,6) or electrochemically (7,8), or even as an actuating sensor that detects the presence of a biological agent and opens or closes a valve in response.

The present challenge is to develop a fundamentally new host-guest topology that eliminates the liabilities of CNT HYAMs. First, the ability of guest expansion to drive yarn untwist depends on the bias angle of the nanotubes with respect to the yarn direction, and this bias angle progressively decreases to zero on moving from yarn surface to yarn center. Hence, while the input energy causing actuation is provided to the entire volume of a HYAM during full-stroke actuation, the fraction of this energy delivered to guest near the yarn core is not effectively utilized, which should decrease energy conversion efficiencies for both tensile and torsional actuation. Second, muscle response times, and therefore output mechanical power, are limited by chemical or thermal transport times to access yarn volume, and these times should decrease if the volume-changing guest is a sheath on the yarn surface. Third, while homochiral coiled yarns (with identical twist and coiling directions) contract in length when yarn volume expands, heterochiral yarns (with opposite twist and coiling directions) expand, and this expansion is useful (but not required) for such applications as comfort adjusting textiles that should increase porosity when heated or exposed to perspiration. However, yarn twist and yarn coiling in heterochiral yarns will irreversibly cancel unless this cancellation is avoided by somehow setting the coiled structure.

We here describe a major change in muscle topology that addresses each of these problems. Rather than locating the volume-changing yarn guest within the yarn’s interior, as for a HYAM, this guest is a sheath on the surface of the host yarn. Since the dimensional and modulus changes of this sheath during muscle excitation drives torsional and tensile actuation, we call the resulting actuators “sheath-run artificial muscles” (SRAMs).
**Fabrication of sheath-run artificial muscles**

The carbon multiwalled nanotube (MWNT) yarns, which were initially used as the muscle core, were drawn as a sheet from a MWNT forest. In an exemplary experiment, a 12-cm-long and 2-cm-wide stack of seven sheet layers was then twisted to make a yarn. Using 74 turns/cm of twist (normalized to the sheet length) and an applied stress during twist insertion of 8 MPa (when normalized to the cross-sectional area of the twisted yarn), a ~41-µm-diameter yarn resulted that had a bias angle of 42°. Although twist insertion into a CNT sheet can result in yarns having either an Archimedean, dual Archimedean, or Fermat structure, depending upon the asymmetry or symmetry of stresses applied during twist insertion (9), yarns having an Archimedean structure were used since they optimized torsional actuation (10, fig. S1). Archimedean yarns resulted from inserting twist along one sheet edge, so that the other sheet edge ends up in yarn center, just like the ends of an Archimedean spiral (Fig. 1A).

Sheath/core yarn muscles were fabricated from twisted yarns (Fig. 1A). A targeted thickness of sheath polymer was deposited around a yarn core by drawing a vertically suspended core yarn through a large droplet of polymer solution multiple times. After completely drying in air to remove solvent, which was typically an ethanol/water solution designed to avoid polymer deposition within the twist-densified core yarn, the sheath/core ratio (i.e., the ratio of the sheath thickness to the yarn diameter) was measured using scanning electron microscope (SEM) images. The nomenclature used to designate a sheath X on a yarn core Y of a SRAM or a X guest inside a yarn core of a HYAM is X@Y.

For all experiments, both tensile and torsional, the thickness of the added sheath was close to the minimum thickness that prohibited such large untwist during release of tethering that the sheath cracked. Figure S7A shows the yarn-bias-angle dependence of the minimum sheath/core ratio needed to prevent sheath cracking for a PEO-SO₃@CNT non-coiled yarn, and fig. S7B shows that approximately this sheath/core ratio maximizes torsional stroke. For instance, for a PEO-SO₃@CNT yarn with a CNT bias angle of ~42°, a sheath/core ratio above 0.14 is needed to prohibit such large yarn untwist that sheath cracking occurs, and a sheath/core ratio lower than 0.22 is needed to allow near optimal torsional actuation (fig.
S7B). For all comparative studies, the weight ratio of guest to CNTs (or other core yarns) was essentially the same for the SRAM and HYAM. This was accomplished by using the above droplet method to add sufficient polymer solution to a low-twist-yarn that the targeted muscle composition was realized, partially drying the polymer solution to provide a gel-like state, and then adding sufficient additional twist that the degree of twist in the HYAM was identical to that in the SRAM.

As shown in Fig. 1C and fig. S2 for the fracture surface of a twisted sheath/core yarn muscle, this deposition process can result in a sharp interface between the sheath and the core and no noticeable guest within the core yarn. To determine if CNT yarns can be replaced by inexpensive yarns, we evaluated commercially available silk as the muscle core, as well as easily-manufacturable electrospun polyacrylonitrile (PAN) nanofibers.

Using the same load as applied during twist insertion for the CNT yarns, insertion of further twist provided a completely “self-coiled” homochiral yarn (Fig. 1B). In order to increase yarn stroke by increasing the spring index (the ratio of the average coil radius to the yarn diameter) twisted yarns or self-coiled yarns (Fig. 1D and 1E, respectively) were coiled by wrapping around a mandrel. After this coiling process, the sheath of the coiled yarn was thermally set to improve performance (10), which for PEO-SO$_3$@CNT was by heating at 115 °C for 4 hours. When referring to the diameter of either a twisted or a coiled yarn, the muscle diameter is defined as the diameter of the sheath/core fiber before coiling. Using the spring index, the coil diameter can be calculated from the yarn diameter (10).

**Torsional actuation of twisted muscles powered by sorption**

Figure 2A illustrates the untwist during vapor sorption and the uptwist during vapor desorption that occurs during the actuation cycle of a SRAM that is torsionally and translationally tethered at only one end. Unless otherwise indicated, for all described actuation cycles of SRAM, HYAM, and pristine yarn, a near equilibrium vapor pressure of an absorbable vapor was provided in a flow of room-temperature dry air and then removed by dynamic vacuum pumping, using the capillary tube system illustrated in Fig. 2B (left). For comparative studies, the same lightweight paddle at yarn end (weighing 60 mg, and having a moment of inertia of 0.28 kg·mm$^2$) was used to characterize paddle rotation angle and speed. Also, all
muscles for SRAM and HYAM comparisons were made from twisted yarn having nearly the same bias angle and the ratio of core yarn weight to guest weight was kept nearly constant for the SRAM and HYAM.

Figure 2B (right) compares the time dependence of paddle rotation and speed for a PEO-SO$_3$@CNT SRAM, a PEO-SO$_3$@CNT HYAM, and a pristine CNT muscle that are undergoing one complete cycle of ethanol-vapor-powered actuation. These results for fully reversible actuation show that the peak stroke and peak rotation speeds for the SRAM (143°/mm and 507 rpm) were about twice that for the HYAM (76°/mm and 254 rpm), and large compared to these parameters for the pristine yarn (4.7°/mm and 36 rpm). Slow, near-equilibrium measurements of torsional stroke versus percent-weight ethanol in the muscles (Fig. 2C) show that the ratio of PEO-SO$_3$@CNT SRAM to PEO-SO$_3$@CNT HYAM torsional strokes reaches a maximum 3.7 for 5.1 wt% ethanol, and thereafter gradually decreases to 1.6 for 18.8 wt% ethanol. There is little hysteresis in torsional strokes for the SRAM and HYAM for increasing and decreasing wt% ethanol, which means that both muscles could be used to reliably open and close valves in response to absorbed vapor. However, these results show that the torsional response of the SRAM is much more sensitive than the HYAM to the wt% absorbed ethanol, especially when this wt% is low.

Figure 2D show that the SRAMs and HYAMs reversibly actuate over 3000 cycles of ethanol absorption and desorption without substantial change in torsional actuation, despite the absence of tethering to an external torsional return spring. The explanation is that the guest in the sheath of the SRAM and the guest within the infiltrated yarn of the HYAM acts as a torsional return spring, so highly reversible actuation can be obtained for SRAMs having only one-end torsional tethering. In contrast, the torsional stroke of the pristine yarn rapidly decreases from the initial 27.3°/mm for the first cycle to about 4.73°/mm on the 27$^{th}$ cycle, thereafter stabilizing at close to this value for the next ~3000 cycles. This eventual stabilization of torsional actuation for the pristine yarn results when the degree of yarn twist becomes sufficiently low (from the initial 74 turns/cm to 21.5 turns/cm) that inter-nanotube interactions become strong enough to act as an internal torsional return spring for subsequent small torsional strokes.
We developed a theoretical model (10) that predicts the dependence of torsional actuation on the sheath/core ratio by using experimentally measured properties of the sheath and core. This model uses the torque balance between the sheath and yarn, both before and after actuation, to predict torsional stroke. The absence of slippage at the sheath/yarn interface is assumed, which is supported by the reversibility of actuation, and the scale invariance of the torsional actuation is utilized, which is derived from the single-helix model (2). This theory reveals that while a low sheath/core ratio limits the capability of the sheath to maintain the initially inserted twist before actuation, a very high sheath/core ratio provides less release of such an initially inserted twist after the SRAM is actuated. Consequently, there is an optimum sheath/core ratio that maximizes torsional stroke.

For actuation driven by ethanol absorption, Figure 2E shows the agreement between the observed and theoretically predicted dependence of torsional stroke on the sheath/core ratio for a PEO-SO$_3$@CNT SRAM made from a CNT yarn having a bias angle of 42° (10). The observed maximum torsional stroke (147 °/mm) occurs for a sheath/core ratio of 0.14, which agrees with the theoretically predicted maximum torsional stroke of 151 °/mm for a sheath/core ratio of 0.15. The experimental results of fig. S6 provide the dependence of torsional stroke on yarn bias angle for a PEO-SO$_3$@CNT SRAM having a sheath/core ratio of about 0.14. These results show that torsional stroke is close to maximum for yarn bias angles between about 39° and 42°, but sharply declines for bias angles below 37°.

In order to evaluate the effect of sheath volume change during ethanol absorption on torsional stroke, the PEO-SO$_3$ sheath of a PEO-SO$_3$@CNT SRAM was replaced by polyvinyl alcohol (PVA) and nylon 6 sheaths for equilibrium torsional measurements in ethanol-saturated dry air. Reflecting the much higher equilibrium percent volume expansion of the PEO-SO$_3$ (15.5%), compared with that for PVA (1.3%) or nylon 6 (0.5%) (fig. S5A), the torsional stroke of the PEO-SO$_3$ SRAM (143°/mm) was much larger than for the PVA SRAM (22°/mm) or the nylon 6 SRAM (11°/mm) (fig. 5B). However, this 13-fold range in torsional stroke reduced to a 2.4-fold range when torsional stroke was normalized to the percent polymer volume change on vapor absorption. The dependence of torsional stroke on vapor type is shown in fig. S8 for a PEO-SO$_3$@CNT SRAM.
We importantly demonstrated high comparative performance for ethanol-sorption-powered torsional SRAM muscles in which the expensive CNT yarn core of the above muscles was replaced by a core of electrospun PAN nanofiber yarn (Fig. S2) or a silk yarn (Fig. 2F). Unlike other comparative examples, the cores of the PEO-SO$_3$@CNT, PEO-SO$_3$@PAN, and PEO-SO$_3$@silk had differing core diameters and bias angles (37 µm and 42°, 76 µm and 30°, and 56 µm and 18°, respectively), since fibers in the silk core and in the PAN core broke when twisted to higher bias angles. For equilibrium torsional actuation of twisted yarns and fibers, the product of torsional stoke and yarn diameter should be invariant when the yarn’s bias angle is kept constant (1). Despite the lower bias angles and larger core yarn diameters that should decrease stroke compared with that for the PEO-SO$_3$@CNT SRAM, the torsional strokes for the PEO-SO$_3$@PAN (123°/mm) and PEO-SO$_3$@silk (70°/mm) SRAMs are comparable to that for the PEO-SO$_3$@CNT (141°/mm) SRAM.

The ratio of peak torsional speed of the SRAM to that of the corresponding HYAM is nearly the same for PEO-SO$_3$@CNT (1.75), PEO-SO$_3$@silk (1.74), and PEO-SO$_3$@PAN (1.79) muscles that are powered by ethanol-vapor, and close to that for water-vapor-powered nylon6@CNT muscles (1.86) (figs. S9). While there is greater variation in the ratio of peak stroke for the SRAM to that of the HYAM (1.86, 1.67, 1.36, 1.63, respectively, for the above), all measurements show the SRAM has an important performance advantage over the corresponding HYAM as a torsional actuator.

**Tensile actuation of coiled muscles powered thermally or by sorption**

By adding sufficient additional twist to a twisted HYAM yarn and a SRAM yarn used for torsional actuation, fully-coiled homochiral HYAMs and SRAMs were obtained that provide large-stroke tensile actuation. By comparing the performance of coiled yarn muscles made from twisted yarns having nearly the same host weight and guest weight per yarn length, we will demonstrate the increases in stroke, stroke rate, and contractile mechanical energy that results from transitioning from the HYAM structure to the SRAM structure.

As schematically illustrated in Fig. 3A, measurements of sorption-powered tensile actuation used the same capillary delivery system as for the torsional actuation measurements. However, for the tensile actuation measurements the torsional rotor was replaced by a weight.
that was prohibited from rotating. Allowing the weight to rotate decreases tensile contraction for a twisted PEO-SO$_3$@CNT SRAM (fig. S10), since yarn untwist increases muscle length. The comparison of electrothermal actuation for the SRAM and the HYAM was for the same input electrical power.

Fig. 3A and fig. S11 shows that an ethanol-vapor-driven coiled PEO-SO$_3$@CNT SRAM can deliver a maximum tensile stroke of 15.7%, as compared with the 10.2% stroke that is provided by an ethanol-vapor-driven coiled PEO-SO$_3$@CNT HYAM for the same 32 MPa tensile load. Muscle stroke and contractile work strongly depend upon the applied stress, as shown in Fig. 3B and table S2 for stresses below loads that cause muscle failure. Importantly, note from these results that the maximum contractile work capacity is 2.03 J/g for the SRAM, which is 1.77 times that for the HYAM. Another useful metric is the maximum average contractile power, which is defined as the maximum ratio of contractile work to actuation time (table S2). The maximum average contractile power output per cycle was 4.44 W/g for the SRAM and 1.51 W/g for the HYAM. While the tensile stroke and contractile work capacity of both the PEO-SO$_3$@CNT SRAM and the PEO-SO$_3$@CNT HYAM increased when vapor delivery was replaced by immersion of the muscle in liquid ethanol (from 15.7% and 2.03 J/g to 19% and 2.71 J/g and from 10.2% and 1.15 J/g to 13.3% and 1.62 J/g, respectively) (fig S8), the SRAM/HYAM work ratio for vapor-powered (1.77) and liquid-immersion-powered (1.67) muscles was little changed. The maximum average contractile power density during liquid immersion was much higher for the PEO-SO$_3$@CNT SRAM (5.8 W/g) than for the corresponding HYAM (3.1 W/g). The load-optimized maximum contractile work capacity (Fig. 3C, D and table S2) for an electrothermally-powered PU@CNT SRAM is 1.33 J/g and the corresponding stroke is 12.5%, which are 2.15 times and 1.83 times the work capacity and stroke, respectively, for the corresponding HYAM (where PU is an elastomeric polyester-based polyurethane, Elastollan®1195A10 from BASF Corporation).

The advantage in contractile work capacity of coiled SRAMs, versus coiled HYAMs, is similar for sorption-driven actuation and electrothermal actuation. More specifically, the ratio of load-optimized contractile work capacity of the SRAM to that of the HYAM was similar for ethanol-vapor-driven PEO-SO$_3$@CNT muscles (1.77), ethanol-immersion-driven
PEO-SO₃@CNT muscles (1.67), and electrothermally-driven PEO-SO₃@CNT muscles (1.73). The ratio of the load-optimized maximum average contractile power density of the SRAM to that for the HYAM was larger for ethanol-vapor-driven PEO-SO₃@CNT muscles (2.94) than for electrothermally-driven PEO-SO₃@CNT muscles (1.69) and electrothermally-driven PU@CNT muscles (2.19). This is understandable since the guest sheath of a SRAM has faster access to externally-applied vapor than does the guest within a HYAM (or to an externally provided temperature change), while the thermal diffusion distance for electrothermally heated muscles is shortest for the HYAMs.

We next theoretically predict the stress dependence of tensile stroke and contractile work capacity for coiled SRAMs and HYAMs for isobaric actuation, and compare theoretical and experimental results (10). This analysis first derives the torsional strokes of the twisted, non-coiled muscles, and then uses the relationship between the torsional stroke due only to volume expansion (ΔT) and the free tensile stroke (ΔL₀) that results from the helical spring equation: ΔL₀=PhΔT/N, where l is the fiber length and N is the number of coils in the fiber length. This ΔL₀ would correspond to the predicted tensile stroke if the spring stiffnesses for the muscle were the same in the actuated and non-actuated states. The illustration in fig. S24 and the associated text (10) describe the calculation of the observed stroke from ΔL₀ using the change in force constants of the coil in going from non-actuated to actuated states.

In the regime where coil-coil contact does not occur during actuation, the theory predicts that the contractile stroke would be independent of isobaric load and the contractile work capacity would increase linearly with increasing load. However, since the force constants for the actuated and non-actuated states differ, there is an optimal load that maximizes the work capacity for both SRAMs and HYAMs. The existence of such an optimal load reflects two competing factors that critically affect work capacity: the contractile stroke (ΔL₀) provided by the muscle fiber untwist as a result of volume expansion, and the elongation of the muscle due to the loss of stiffness during actuation.

Using the above theoretical approach and no fit parameters, the results of fig. S25 and Fig.3B show the remarkable agreement between experimentally measured and theoretically calculated stress dependence of stroke and contractile work capacity for isobaric actuation of
ethanol-powered SRAM and HYAM PEO-SO₃@CNT yarns. Importantly, the predicted ratio of the maximum contractile work capacity of the SRAM to that of the HYAM is 1.52, which is close to the experimentally measured ratio of 1.77.

**Electrochemical tensile actuation of coiled muscles**

Electrochemically powered artificial muscles have important advantages that are not found for thermally powered muscles: (1) their efficiency is not limited by the Carnot efficiency and (2) they have a natural latching state, meaning that stroke can be maintained for application-important times without the input of electrical energy. In this section, the fabrication of electrochemical CNT@nylon6 SRAMs will be described, together with their performance advantages compared to CNT yarn muscles. The electrochemical CNT yarn muscles is effectively a HYAM, wherein the yarn guest is a volume-changing electrolyte.

The CNT@nylon6 SRAM, which contains a nylon 6 yarn core that supports a CNT sheath, where made using the novel process shown in Fig. 4A. Like for a process used to make coiled CNT yarns for energy harvesting, a stack of CNT sheets was formed into a cylinder. For energy harvesters, twist was inserted into this cylinder to make the utilized CNT yarn. This process was modified in order to make the electrochemical SRAMs, by inserting a nylon yarn into the center of the cylinder. During the initial stage of twist insertion, the twist is inserted only into the CNT cylinder. However, once the CNT cylinder collapses to form a sheath on the nylon 6 yarn, torque transfer from this sheath to the yarn automatically occurs, which enables the nylon yarn to become fully coiled. Since the same CNT cylinder was used for fabricating the SRAM sheath and the CNT yarn, the CNT weight per sheath length and per yarn length were essentially identical for comparative studies.

The electrolyte-filled CNT sheath of the CNT@nylon6 SRAM and the electrolyte-filled volume of HYAM provide electrochemical actuation because of the volume changes produced by electrochemical double-layer charge injection. Consequently, the solvated sizes of the mobile ions of the electrolyte are important. For the presently used electrolyte of 0.2 M tetrabutyl ammonium hexafluorophosphate (TBA·PF₆) in propylene carbonate, the calculated van der Waals volume (V) of the TBA⁺ cation (~293 Å³) is much larger than for the PF₆⁻ anion (69 Å³). Hence, ignoring changes in relative ion size due to solvation, muscle contraction is
expected to be much larger at negative potentials than at positive potentials (if injected electrons and holes on the CNTs are compensated by removal and addition of cations and anions, respectively, from the electrochemical double layer).

In agreement with this prediction, the observed tensile contractions during a low rate potential scan (Fig. 5) are largest at negative electrode potentials for both the SRAM and the HYAM, although these contractions are far larger for the SRAM at both negative and positive potentials. The existence of peaks in muscle contraction might result from the fact that holes and electrons can be compensated by either the addition of oppositely charged ions or the removal of the same sign ions (depending upon potential scan rate and potential).

Since the electrical energy needed to produce actuation increases with increasing amount of electrochemically accessible CNTs in the muscle, the contractile work per weight of CNT is an important performance metric. Figure 5C shows that this work capacity is higher for the CNT@nylon6 SRAM (2.26 J/g) than for the CNT yarn muscle containing the same weight of CNT per yarn length (1.36 J/g) when the yarn potential is switched from 0V to -3.5 V (vs. Pt reference). Most importantly, the contractile energy conversion efficiency, which is the ratio of the contractile work to the net electrical energy consumed during a cycle (10), increases from 4.97% for the CNT yarn muscle to 7.18% for the SRAM. This contractile energy conversion efficiency for the SRAM (7.18%) is higher than previously reported for electrochemical yarn muscles. For further comparison, the highest energy conversion efficiency that have been reported for any other type of electrochemical muscle is 5.4% (8).

Since positive work results during muscle contraction and negative work occurs during muscle expansion, realizing a positive net mechanical work output during a cycle requires that the stress applied during muscle contraction is larger than during muscle expansion. Using the work loop method (12, 13) and experimental results in Fig. S17 to S21, lower bounds on the full-cycle energy conversion efficiency of the electrochemically-powered coiled CNT@nylon6 SRAM and coiled CNT muscle were obtained. This full-cycle energy conversion efficiency was higher for the electrochemically-powered coiled CNT@nylon6 SRAM (2.04%) than for coiled CNT muscle (1.37%). This full-cycle energy conversion efficiency for the CNT HYAM
is close to that previously reported (13) for an electrochemically driven CNT HYAM (1.5%) that uses the same electrolyte, but is a two-ply muscle rather than a single-ply muscle.

The enhancement in actuation rate of using SRAMs is shown in Fig. 5D, where the tensile stroke is plotted versus cycle frequency (Fig. 5D) for actuation of a SRAM and a HYAM (using square wave potential pulses with 50% duty cycle from 0.5 to -3.5 V). These results show that the maximum tensile stroke of the hybrid yarn (CNT yarn and infiltrated electrolyte as guest) of 12.7% at 0.01 Hz full cycle frequency can be realized at a fully cycle frequency of 0.08 Hz for the SRAM. Using this rate advantage of the SRAM and an optimized square wave potential during actuation (0.5 V to -3.5 V with 50% duty cycle), a muscle stroke of 5% was obtained at 1 Hz, which expands the application possibilities for electrochemical artificial muscle. If an application requires a higher tensile contraction, a higher spring index can be used, including the large spring indices obtainable by mandrel that enable arbitrarily high strokes (but with related reductions in load lifting capability). If the application requires a higher frequency response, the diameter of the SRAM can be decreased so that reduced ion diffusion times increase actuation rate.

**Actuation mechanism**

Since torsional actuation of twisted muscles is the origin of tensile actuation of coiled muscles, understanding the performance advantage of SRAM torsional actuation is key for understanding why coiled SRAMs consistently outperform coiled HYAMs in tensile actuation. To start, note that the twisted torsional muscles provide an initially torque-balanced structure in which the sheath guest of the SRAM or the yarn-infiltrated guest of the HYAMs restricts yarn untwist. Both guest volume changes and guest modulus changes can upset this balance. For instance, as illustrated in Figure 2A, vapor absorption causes volume increase (fig. S5A) and modulus decrease (fig. S4A) of the polymer sheath, which decreases the torsional stiffness of the sheath and causes the highly-twisted fiber core to untwist. The polymer sheath also acts as a torsional return spring for storing the twist release from the fiber core.

Since the yarn modulus is much larger in the axial direction than in the lateral directions, the main effect of the sheath is assumed to be in changing the yarn diameter. Based on this key hypothesis, a theoretical estimate on the torsional actuation is obtained by analyzing the
difference between the initial and new balanced states during the torsional actuation of SRAM. Detailed analysis is provided in the SOM. The theoretical analysis predicts that torsional actuation of SRAM is critically dependent on the sheath/core ratio, in addition to material parameters such as the moduli of the sheath and yarn in their initial and absorbed states. While this geometric dependence is expected, the quantitative analysis provides important guidance on the design of SRAMs for a wide range of applications.

**Applications**

The possible applications of SRAMs are diverse. The torsional actuation of a CNT HYAM has been used for the detection of glucose and dopamine. The increased sensitivity and response rate of SRAMs might eventually enable their use for detecting antigens in blood using sheaths of known antigen/antibody substituted polymers, which expand because of water absorption when native antigen breaks cross-links between polymer chains. Either torsional actuation or tensile actuation could be used as analyte-powered sensors that intelligently respond in the body to open and close valves that release drugs. However, it will likely be a long time (if ever) before contact between CNTs and blood is allowed in the human body. The presently demonstrated ability to eliminate CNTs from high performance artificial muscles might accelerate medical acceptance. In less challenging possible applications, the SRAMs could be used in mechanically valves that control the flow or mixing of liquids depending upon their chemical composition or temperature (or a combination of these effects, using mechanical logic circuits).

The use of SRAMs for harvesting chemical or thermal energy as mechanical energy, which is then converted to electrical energy, is also of interest for both small scale applications (like powering remotely communicating sensors for the internet of things) and large-scale applications (like harvesting the chemical energy difference between effluents in a chemical plant).

The wide range of core materials that can be used (Fig. 2D) shows the great potential towards smart textile industry (Fig. 4). We fabricated a new structure we called hot-pressed coiled that produces a 2D structure that changes its pores. Firstly, a mandrel coiled PEO-SO$_3$/Polycarbonate fiber (spring index of 11.5) which showed a 48.6% actuation once
triggered with moisture was fabricated (Fig. 4A). Then, flat-coil SRAM was fabricated through hot-pressing the mandrel coiled PEO-SO₃/Polycarbonate fiber (Fig. 4B). This new hot-pressed coiled SRAM structure could be used to fabricate smart textile that change porosity in response to humidity for clothing that increases wearer comfort (Fig. 4C). A knitted SRAM smart textile (Fig. 4D) was also prepared using highly twisted hydrogel coated polyester fibers. Change of textile porosity was demonstrated during isobaric tensile actuation while sample was clamped vertically, and ethanol vapor triggered the knitted SRAM smart textile.

We also demonstrated that mandrel supercoiled muscles can greatly amplify these strokes. We showed a maximum tensile stroke of about 70% for moisture-driven cone-mandrel SRAM (fig. S12A) and this structure enabled faster actuation than both the cylinder-mandrel and the supercoil SRAM structures (fig. S12B and S12C, respectively). Even though moisture driven muscles are typically slow, this is the highest stroke per time of contraction that has been reported for a CNT HYAM.

Figures and Figures Captions:

Fig. 1. Muscle fabrication and structures for torsional and tensile actuation. (A) Schematic lateral and cross-sectional views of a twisted CNT yarn and a SRAM, which was made by coating the twisted CNT yarn with a polymer sheath. SEM micrographs of PEO-SO₃@CNT muscles showing (B) a SRAM made by self-coiling a sheath-coated twisted
yarn, (C) the surface of a twisted SRAM that was broken by bending in liquid N₂, showing that the sheath polymer does not infiltrate the CNT core, (D) a mandrel-coiled twisted SRAM, and (E) a SRAM that was self-coiled and then mandrel-coiled. Although mandrel coiling is useful for providing both heterochiral muscles and SRAMs having giant strokes, unless otherwise indicated, the described results are for twisted SRAMs or self-coiled SRAMs. The scale bars for (B)–(E) are 35, 15, 200, and 200 μm respectively.
Fig. 2 Torsional actuation of twisted PEO-SO₃ SRAMs and HYAMs driven by ethanol-saturated dry air. (A) Illustration of a vapor-driven PEO-SO₃ SRAM before vapor exposure (left) and during ethanol vapor sorption (middle) and desorption (right), which cause yarn untwist and up-twist, respectively. (B) Illustration of the configuration used for vapor delivery to a one-end-tethered SRAM (left) and the time dependence of torsional stroke and rotation speed for one reversible sorption/desorption cycle for a PEO-SO₃@CNT SRAM, a PEO-SO₃@CNT HYAM, and a pristine yarn (right). The same 39-µm-diameter CNT yarn, containing 36 turns/cm of twist, was used for the pristine muscle and for the fabrication of the SRAM and HYAMs. Also, the SRAM and HYAM weighed nearly the same (0.24 mg/cm) and contained the same weight ratio of PEO-SO₃ to CNT (~0.53). The SRAM yarn diameter was 45 µm and the HYAM yarn diameter was 50 µm. (C) The dependence of equilibrium torsional stroke on weight change during sorption/desorption of ethanol (black squares) for the yarns of (B). (D) Torsional stroke as a function of cycle number for the muscles of (B). (E) Comparison of the experimentally observed (black squares) and the theoretically predicted (blue line) dependence of torsional stroke on sheath/core ratio for a twisted PEO-SO₃@CNT SRAM. (F) Torsional stroke and rotation speed for one reversible sorption/desorption cycle of ethanol-vapor-powered actuation of a PEO-SO₃@silk SRAM, a PEO-SO₃@silk HYAM, and a silk yarn. The same 56.0-µm-diameter silk yarn, containing 5.7 turns/cm of twist, was used for the pristine muscle and for the fabrication of a 88.7-µm-diameter SRAM and a 90.7-µm-diameter HYAM. The SRAM and HYAM weighed 0.48 mg/cm and contain the same weight ratio of PEO-SO₃ to silk (0.27).
Fig. 3. Tensile actuation of coiled sorption-powered and electrothermally-powered tensile SRAMs, HYAMs, and pristine yarn. (A) Tensile stroke vs time for a PEO-SO\textsubscript{3}@CNT SRAM, a PEO-SO\textsubscript{3}@CNT HYAM, and a pristine CNT yarn when isobarically actuated by ethanol absorption using the configuration shown on the left and a stress of 33 MPa. Sorption used a near-equilibrium ethanol concentration in dry air (0.234 mg/liter) and desorption was by dynamic pumping. Before coiling, the diameters of the PEO-SO\textsubscript{3}@CNT SRAM, the PEO-SO\textsubscript{3}@CNT HYAM, and the pristine yarn were 43, 47, and 38 µm, respectively. (B) Tensile stroke and contractile work capacity vs applied isobaric stress for the PEO-SO\textsubscript{3}@CNT SRAM, the PEO-SO\textsubscript{3}@CNT HYAM, and the pristine CNT yarn of (A) when sorption-actuated like in (A). (C) Tensile stroke vs time for a PU@CNT SRAM, a PU@CNT HYAM, and a pristine CNT yarn when electrothermally actuated using the same 0.25 W/cm power, which provided temperatures of 85, 93, 97°C, respectively, and the same isobaric load (~42 MPa). The device configuration is shown on the left. Before coiling, the diameters of the PU@CNT SRAM, the PU@CNT HYAM, and the pristine yarn were 65, 71, and 51 µm, respectively. (D) Tensile stroke and contractile work capacity vs applied isobaric stress for the PU@CNT SRAM, the PU@CNT HYAM, and the pristine CNT yarn of (C) when electrothermally actuated as in (C).

Fig. 4. Demonstration of giant stroke tensile actuation for a novel planar SRAM, which is powered by water vapor absorption using the apparatus of Fig. 3. (A) Photographs showing the fabrication of a planar SRAM: (top) A homochiral, mandrel-coiled, twisted SRAM using a 150-µm-diameter PEO-SO\textsubscript{3}/polycarbonate yarn having a sheath/core ratio of 0.33, an inserted twist of 22 turns/cm and a spring index of 11.5. Hot pressing this mandrel-coiled SRAM between planar plates at 120 °C for 6 hours produced the below SRAM, which is pictured in actuated and non-actuated states when the applied stress is 4.3 MPa. (B) For illustration of the possible use of planar heterochiral SRAM in a textile, photographs of three SRAM muscles interconnected by an inert 150-µm-diameter fiber are shown for the moisture-free state (top) and the expanded moisture-absorbed state. (C) Photographs showing (for an isobar load of 4.7 MPa) the 48.6% contraction obtained by exposing the homochiral planar SRAM to moist air (RH = 70%), after it had been equilibrated in an ambient RH of 34%. For clarity, the background of these muscles has been made black.
Fig. 5. Fabrication and electrochemical actuation of CNT@nylon6 yarn and a CNT yarn in 0.2 M TBA·PF$_6$. (A) Illustration of the cone spinning process for fabricating CNT yarns and the modification of this process to make SRAM yarns. The photographs show cross-sectional and lateral SEM micrographs of the pristine yarn (top) and the SRAM and core nylon yarn (bottom) whose actuation was characterized. (B) Electrochemical tensile stroke during a cyclic voltammetry scan, where the voltage are relative to a Pt reference electrode, the high capacitance counter electrode is a CNT covered Pt mesh, the isobaric stress is 22 MPa, and potential scan rate was 20 mV s$^{-1}$. Inset: Actuator stroke for these muscles versus interelectrode voltage scan rate for an isobaric stress of 22 MPa. (C) The stress dependence of tensile stroke and energy conversion efficiency for the muscles of (A) to (C) and a voltage scan rate of 20 mV s$^{-1}$. (D, E) The time dependence (D) and frequency dependence (E) of tensile stroke for a coiled SRAM and a coiled HYAM for square-wave potentials between 0.5 and -3.5 V with 50% duty cycle. The spring indices of the
87-µm-diameter CNT@nylon6 yarn and the 72-µm-diameter CNT yarn were 1.12 and 0.98, respectively.

References and Notes


10. Supplementary materials are available on Science Online.


Supporting Information

1. Materials and Fabrication

1.1 Core materials and core yarn fabrication

The sources and structural characteristics of the yarns used for hybrid yarn artificial muscles (HYAMs) and for the core of sheath-run artificial muscles (SRAMs), as well as the diameters of the torsional muscles, are provided in Table S1. These results are for core yarns of carbon multiwalled nanotubes (MWNTs), electrospun polyaclylonitrile (PAN) nanofibers, nylon 6, and natural silk.

The MWNT forests used to produce MWNT yarns were ~350 µm high, and comprised MWNT having approximately six walls and having an outer diameter of ~9 nm. In order to evaluate the effect of yarn structure on muscle properties, we varied the spinning method to obtain yarns having Archimedean, Fermat, and dual Archimedean structures (1), which are illustrated in Fig. S1. By designing the spinning method to insert twist at the center, one edge, and both edges of the spinning triangle, yarns having Fermat, Archimedean, and dual Archimedean structures were obtained. Fermat yarns were directly twist-spun from MWNT forests. By twisting a rigidly end-supported rectangular stack of forest-drawn MWNT sheets, we obtained Archimedean yarns if the applied stress was symmetric and dual Archimedean yarns if the applied stress was asymmetric.

Oriented sheets of electrospun PAN nanofibers with an average nanofiber diameter of ~245 nm were produced by electrospinning using a method previously described (2). In this method, PAN with an average molecular weight of Mw = 150,000 g/mol (Aldrich Company) was dissolved in N,N-dimethylformamide by stirring at room temperature overnight to yield an 8 wt.% solution. The solution feed rate to the electrospinning needle was 35 µL min⁻¹, the distance between the needle tip and the two metal wire collectors was about 13 cm, the applied electrostatic field was 0.9 kV/cm, and the metal wire collectors were 12 cm long and separated by 8 cm. Hence, the produced PAN sheet, comprising aligned PAN nanofibers, was 8 cm by 12 cm. Archimedean PAN yarns (Fig. S2 left) were obtained by twist spinning PAN sheets. Unless otherwise stated, the PAN and MWNT core yarns had an Archimedean structure.

To provide a well-defined interface between sheath and core, commercially available twisted yarns were inserted with additional twist before the coating process to make SRAMs. On the other hand, since guest penetration into the core yarn was needed to make HYAMs, additional twist into the commercially available yarns was made after guest infiltration.

1.2 Fabrication of SRAMs and HYAMs

The nomenclature used to designate a sheath X on a yarn core Y of a SRAM or a X guest inside a yarn core of a HYAM is X@Y. The following polymers were used as sheaths for the non-electrochemical SRAMs and as guests for non-electrochemical HYAMs: polyvinyl alcohol (PVA), nylon 6, an elastomeric polyester-based polyurethane (Elastollan®1195A10 from BASF Corporation, which is abbreviated PU), and a mixture of polyethylene oxide (PEO) and a co-polymer of tetrafluoroethylene and sulfonyl fluoride vinyl ether (F₂C=CF-O-CF₂-SO₂F), which we call PEO-SO₃. PVA (7 vol.% concentration in water) and nylon 6 (6 vol.% concentration in formic acid) were purchased from Aldrich Company. The PEO-SO₃ was prepared by mixing PEO (Aldrich Company) and co-polymer of tetrafluoroethylene and sulfonyl fluoride vinyl ether (Aquivion® PFSA from the Solvay Group) at a weight ratio of 3:7.

Uniform sheaths were deposited at room temperature in air on core yarns by manually drawing droplets of the viscous sheath polymer along the length of the vertically suspended core
yarn, while the yarn was torsionally tethered to prevent yarn untwist. Depending upon the thickness of the sheath that was desired, this droplet-coating method was repeated multiple times. After completely drying in air to remove solvent, the sheath/core ratio (i.e., the ratio of the sheath thickness to the yarn diameter) was measured using scanning electron microscope (SEM) images.

The above droplet deposition process was modified in order to infiltrate a guest polymer into a core yarn to make a HYAM, rather than to coat the core yarn to make a SRAM. This modification was of two types. First, the solvent used for polymer infiltration was selected to enable wetting of the core yarn. For instance, while PEO-SO$_3$ SRAM fabrication used an ethanol-to-water weight ratio of 5:5 for the solvent, an ethanol-to-water ratio of 8:2 was deployed for making a PEO-SO$_3$ HYAM. Additionally, the infiltration process for fabricating the HYAMs occurred while the yarn was in a less highly twisted state (i.e., lower density state) than used for depositing the sheath of a SRAM. After this deposition, while the guest was in a gel-like state, additional twist was inserted so that the degree of twist in the HYAM was identical to that in the SRAM.

According to the type of core yarn and the desired actuator properties, the coiled muscles used for tensile actuation were made either by self-coiling a twisted SRAM or a twisted HYAM (meaning inserting twist until complete yarn coiling occurred) or by wrapping a twisted or self-coiled SRAM or twisted or self-coiled HYAM around a mandrel. After these coiling processes, the coiled SRAMs or coiled HYAMs were thermally annealed to set the guest. The annealing conditions for the guests were 115°C for 4 hours for PEO-SO$_3$, 60°C for 2 hours for PU, and 180°C for 2 hours for nylon 6. The fabrication of the electrochemical muscles is described in the main text. The stresses applied during the twist insertion and self-coiling are identical, and are normalized with respect to the cross-sectional area of the twisted yarn muscle before annealing. This tensile stress during actuation was normalized in the same way.

The SEM micrograph of Figs. 1C and S3A for PEO-SO$_3$@CNT SRAM, and of Fig. S2A (right) for PEO-SO$_3$@PAN SRAM, show that the guest in the sheath does not infiltrate into the muscle core. Complete infiltration of the guest into the core yarn is shown in Fig. S3B for a PEO-SO$_3$@CNT HYAM. The SEM images of Fig. S3 were obtained by sectioning the PEO-SO$_3$@CNT SRAM and HYAM using Ga ions (5 nA beam current) in a Focused Ion Beam (FIB, Nova 2000) operated at 30 kV, followed by ion-polishing (consecutively decreasing ion-currents from 3.0 to 0.3 nA). The inset of Figure S3A shows that the core CNT yarn can be pulled out of the PEO-SO$_3$@CNT SRAM, thereby leaving only the PEO-SO$_3$ sheath.

### 1.3 Characterization of key guest materials

Using an Instron 5848 Microtester, the time dependence of the Young’s modulus during ethanol sorption and desorption of a 75-µm-thick, 5 x 30 mm sheet of PEO-SO$_3$ was measured (Fig. S4A). Since thermal actuation of PEO-SO$_3$@CNT will be characterized, we used thermogravimetric analyses (TA Instruments Q600 Thermogravimetric Analyzer) to determine the thermal stability of PEO-SO$_3$ (Fig S4B). These results show an initial weight loss of 8% up to ~280°C on heating (5°C/min in air), which results from desorption of water. Above this temperature, the polymer thermally degrades. Since the maximum temperature used for thermal actuation is 180°C, thermal degradation does not significantly occur during actuation.

Since they are important for understanding actuation, we characterized the effects of solvent absorption/desorption on guest volume and guest modulus. By recording the dimensions of a polymer sheet as a function of time during exposure to 0.234 mg/L of ethanol vapor in dry atmospheric pressure air (using time-lapse photography), the equilibrium percent volume change of PEO-SO$_3$, PVA and nylon 6 guests were measured (Fig. S5A). This 0.234 mg/L of ethanol approximately corresponds to a saturated atmosphere of ethanol in dry room temperature air.
1.4 Fabrication and characterization of SRAM textiles that increases porosity when exposed to perspiration

Moisture-responsive coiled SRAM yarn muscles were used to make a woven textile that respond to the presence of perspiration. For making these SRAMs, a 300-μm-diameter cotton yarn core was fully coiled by twist insertion and then overcoated with the SRAM sheath. This sheath was a double-network interpenetrating hydrogel comprising alginate-methacrylate mixed with polyvinyl alcohol (PVA), where the used chemicals were purchased from Sigma-Aldrich. Alginate-methacrylate was synthesized using a previously described method (3). To covalently attach the methacrylate groups to the alginate backbone, alginic acid was dissolved in distilled water (3% w/v) and subsequently methacrylic anhydride (15% v/v) was added dropwise to the solution while the pH was kept constant at 8 for 6 hours (using 5N NaOH solution). Afterwards the resulting mixture was dialyzed against distilled water for 5 days, the polymer was freeze-dried. The degree of methacyrlation of alginic-methacrylate was 48%, based on hydrogen NMR spectroscopy of the polymer powder in deuterated water. The aqueous coating solution used for sheath formation was made of a 75/25 (v/v) solution of 3% (w/v) alginate-methacrylate and 15% (w/v) PVA, respectively. Subsequently, 0.1% (w/v) of photoinitiator (Ingacure 2559 from Sigma-Aldrich) was added to the mixture for enabling UV crosslinking of the alginate-methacrylate. The coiled cotton core yarn was coated with the sheath polymer by immersing this yarn into this solution. Ultraviolet cross-linking of the hydrogel sheath was accomplished by exposing the sheath-coated coiled yarn to ultraviolet radiation (6.9 mW/cm for 360-480 nm radiation) for 15 minutes at 10°C, followed by storage of the sheath-coated yarn at this temperature for an hour to enable recrystallization of the PVA polymer chains. The perspiration-responsive textile, made by hand weaving, had a plain-weave structure, with the SRAM muscles in the warp direction, and a red 800-μm-diameter two-ply cotton yarn in the weft direction.

The effect of perspiration on textile actuation was mimicked by spraying water on the SRAM textile. Experimental results have shown that there was no significant difference in SRAM actuation using water and artificial adult perspiration (Artificial Eccrine Perspiration - Stabilized from Pichering Laboratories). Figure S23 and Movie S6 show the reversible actuation that occurs when the SRAM textile is exposed to water. Even though the SRAM muscles contract during absorption of the water, this absorption caused the through-yarn projected pore area to dramatically increase, from 8% of the total textile area before water exposure to 25% after water exposure. This opening of pores results, since contraction of the strong, small-diameter SRAM muscles, reduces the diameter of the much larger diameter ordinary cotton yarns.

Water-responsive twisted SRAM yarn muscles were also used to fabricate knitted textiles that respond to the presence of perspiration by opening pores. For making these SRAMs, a 2-mm-diameter aramid yarn core was twisted under 4 MPa stress (so that the total twist in the yarn was 16 turns/cm), and then the yarn was over coated with a SRAM sheath (PEO-SO3). This aramid yarn was a poly p-phenylene terephthalamide yarn (ST10M) from Jiangsu Shino New Materials Technology Co., which contains thirty-seven 53-µm-diameter filaments in the yarns cross-section. While maintaining tension on the yarn to prevent coiling, the SRAM was used to make a weft knitted textile using a commercial sock-knitting machine (WH-6H-C from Huaxing company). Upon release of this tension, yarn twist was partially converted to yarn writhe (i.e. coiling), thereby resulting in a weft knitted textile comprising homochiral SRAMs. As illustrated in Figure S23, this coiling during stress release transformed the knitted sock into one in which opposite textile sides have quite different structure, and wherein the heads of the textile loops bridge the separation between textile sides. To understand the structure of the coiled SRAM fibers in the released textile,
the structure of the textile was permanently set by coating the yarns in the textile with epoxy resin, then allowing the epoxy to crosslink. Removal of a set yarn from the textile showed that the yarn in the textile had a coil index of 27.5.

Figure S23 and Movie SXX show the reversible actuation that occurs when the knitted SRAM textile was sprayed with water. Absorption of water by the textile caused the through textile hole area to increase from 46.7% of the total textile area before water exposure to 55.4% after water exposure. This increase in porosity resulted from yarn untwist to produce increased writhe. This increase in writhe caused the textile to shear like a collapsing wine rack, thereby increasing porosity and decreasing the length in the warp direction by XX%, while maintaining the length in the weft direction.

2. Method and Apparatus for Characterizing Actuation

Torsional and tensile electrothermal and absorption-driven actuation were characterized using a commercial laser proximity sensor system (Keyence Corporation of America), which captured either the rotation of a paddle or the displacement of a weight attached at the end of the muscle. These electrothermal measurements were for muscles that used a carbon nanotube core, which served as the heating element. The measurements using the proximity sensor were complemented by results obtained by frame-by-frame analysis of movie pictures taken using a CASIO 12.5HS camera. Thermal tensile actuation was characterized using a TA Instruments TMA Q400 thermomechanical analyzer, and heating and cooling rates of 5°C/minute. Vapor-driven actuation used the equilibrium vapor pressure of a liquid in dry air.

Electrochemical measurements of tensile actuation used a high capacitance counter electrode (a CNT covered Pt mesh). For three-electrode measurements, the reference electrode was Pt wire. A contactless inductive proximity sensor (Omega LD701 5/10) with data acquisition (Omega module OM-USB-1408Fs) was used to record the displacement of a weight attached at the end of a muscle.

Unless otherwise indicated, all stresses are nominal values determined by normalizing applied force to the cross-sectional area of the non-loaded, non-actuated muscle and the torsional stroke was normalized to the loaded length of the actuator. Unless otherwise described for data plots of actuation versus time, actuation was reversed after essentially the entire muscle stroke was realized (as indicated by previous measurements of muscle stroke versus time, where actuation was observed until muscle stroke reached essentially constant value).

3. Characterization of Torsional Actuation

3.1 Effect of scroll topology and inserted twist on torsional actuation of a PEO-SO₃@CNT SRAM

The results of Fig. S1 show that the scroll type of the SRAM core importantly affects torsional actuation. More specifically, an Archimedean core structure provides higher torsional stroke and a higher peak rotation speed than either a Fermat or a dual-Archimedean core SRAM, and the stroke and peak rotation speed of the dual-Archimedean SRAM are lowest. This especially low performance of the dual-Archimedean SRAM is understandable, since this SRAM has two centers of torsional rotation rather than the one center found for the other core structures.

Figure S6 shows the torsional stroke of an ethanol-absorption-driven PEO-SO₃@CNT SRAM rapidly increases with the increasing CNT yarn bias angle until the stroke reaches a plateau at ~40° and then decreases for bias angles above ~43°, above which yarn coiling is initiated. The ratio of torsional stroke to inserted twist reaches a peak at ~27° and decreases at higher angles.
3.2 Effect of guest material on the torsional actuation of an ethanol-vapor-driven SRAM

Figure S5B shows that the torsional stroke of the PEO-SO3@CNT SRAM (143.1°/mm) is giant compared with that for PVA@CNT SRAM (21.7°/mm) and nylon6@CNT SRAM (10.2°/mm). This relatively giant stroke of the PEO-SO3@CNT SRAM can not be explained by only considering the much larger percent volume change of the PEO-SO3 (16.7%) than for the PVA (1.3%) and nylon 6 (0.5%), as shown in Fig. S5A. This is indicated by the fact that the ratio of the diameter-scaled torsional stroke (the product of torsional stroke and the muscle diameter) to the percent volume change of the sheath is not constant, and varies from 0.38 for PEO-SO3@CNT SRAM to 0.84 for PVA@CNT SRAM and 0.9 for nylon6@CNT SRAM. The explanation is, in part, that the modulus of the sheath decreases with increasing solvent sorption, enhancing torsional stroke, and this modulus decrease increases with increasing percent volume change of the sheath. The sheath/core ratio of these SRAMs are relatively constant, varying from 0.12 and 0.14 for nylon 6 and PEO-SO3, respectively, to 0.17 for PVA.

3.3 Effect of sheath/core ratio on the fabricability and torsional actuation of a PEO-SO3@CNT SRAM

Figure S7A shows the combinations of sheath/core ratio and yarn bias angle that result in sheath cracking (red region) during SRAM fabrication and combinations that result in crack-free sheaths (white region). The data points denote the maximum bias angle that can be inserted for a given sheath/core ratio without causing cracks in the sheath of the SRAM. Figure S7B shows the dependence of torsional stroke and maximum rotation speed on sheath/core ratio for a non-coiled 39-µm-diameter PEO-SO3@CNT SRAM containing 74 turns/cm of twist. These results indicate that a sheath/core ratio of ~0.14 enables a bias angle of 42° (corresponding to 74 turns/cm of inserted twist) without crack formation. The decrease in both maximum rotation speed and torsional stroke for sheath/core ratios above ~0.19 results from mechanical hinderance of torsional rotation by a sheath thickness that is too high. The dash line is the theoretically predicted dependence of torsional stroke on sheath/core ratio (Section 6), which agrees with the experimental results (Fig. 2E).

There should be a minimum sheath/core ratio (~0.14) to provide a torque that prohibits such large untwist that the sheath cracks, and this depends upon the torsional modulus and inserted twist of the core and the failure stress of the sheath. On the other hand, sheath/core ratio above ~0.2 decreases torsional stroke. These experimental findings agree with theoretical predictions (Fig. 2E), as explained in Section 6.

3.4 Effect of different vapors on the torsional actuation of a PEO-SO3@CNT SRAM

Of the investigated solvents, ethanol provided the largest torsional stroke for a PEO-SO3@CNT SRAM. However, the results in Fig. S8A show that usefully large torsional strokes can also be obtained for this SRAM when using various other vapors in saturated room temperature air. All of these results were obtained using the same SRAM as deployed for Fig. 2B,C,F.

3.5 Comparison of the torsional actuation of a nylon6@CNT SRAM, a nylon6@CNT HYAM, and a neat CNT yarn

The results of the present study demonstrate that similar performance advantages of SRAM structure over the hybrid yarn structure can be obtained when ethanol-driven PEO-SO3@CNT muscles are replaced by water-driven nylon6@CNT muscles. Figure S9 shows the time-dependence of torsional rotation angle and torsional rotation speed for water-vapor-driven torsional actuation of a 43.7-µm-diameter nylon6@CNT SRAM, a 47-µm-diameter nylon6@CNT HYAM and a 39-µm-diameter pristine CNT yarn. The performance advantage of using a SRAM compared with using a HYAM is nearly the same as for the PEO-SO3@CNT muscles of Fig. 2B.
More specifically, the ratio of torsional stroke and maximum torsional speed for the nylon6@CNT HYAMs are 1.62 and 1.86, respectively, compared with 1.86 and 1.75, respectively, for the PEO-SO₃@CNT muscles.

4. Characterization of tensile actuation

4.1 The effect of torsional tethering on the ethanol-vapor-driven tensile actuation of a twisted PEO-SO₃@CNT SRAM

Figure S10 shows the effect of torsional tethering on the time-dependence of tensile stroke during actuation of an ethanol-vapor-driven twisted PEO-SO₃@CNT SRAM. Actuation was by exposure of the muscles to 0.234 mg/L of ethanol in dry air, and actuation was reversed by desorbing the ethanol using dynamic pumping. These results indicate that the tensile stroke of a torsionally-tethered SRAM (2.07%) is much larger than for a SRAM that is free to rotate (1.25%). Nevertheless, this tensile stroke for the torsionally-tethered twisted SRAM is much smaller than for an identical SRAM that is coiled and torsionally tethered (Fig. 3A), which has maximum tensile stroke of 15.7%. The larger tensile stroke for the torsionally tethered SRAM than for the non-torsionally-tethered SRAM is understandable, since torsional untwist during actuation causes the yarn to elongate.

4.2 Effect of different solvents on liquid-immersion-driven tensile actuation of a PEO-SO₃@CNT SRAM

Of the investigated solvents for tensile actuation, acetone provided the largest tensile stroke for a liquid immersion driven PEO-SO₃@CNT SRAM (28.6%). However, the results in Fig. S8B, show that usefully large liquid-immersion-driven tensile strokes can also be obtained for this SRAM (which was used for Fig. 3A,B) when using various other solvents. When immersed in liquid ethanol, PEO-SO₃@CNT SRAM provided a maximum tensile stroke and maximum contractile work capacity of 38% and 2.6 J/g, which are XX times and 1.6 times, respectively, the values for a PEO-SO₃@CNT HYAM (Fig. S8C). This work capacity of the PEO-SO₃@CNT SRAM (2.6 J/g) is the highest reported in literature for sorption-driven muscles.

4.3 Comparison of ethanol-vapor-driven tensile actuation of a coiled PEO-SO₃@CNT SRAM, a coiled PEO-SO₃@CNT HYAM, and a coiled neat CNT yarn

Figure S11 shows the time-dependence of tensile stroke for an ethanol-vapor-driven coiled PEO-SO₃@CNT SRAM, a coiled PEO-SO₃@CNT HYAM, and a coiled pristine yarn that were torsionally tethered. Before coiling, these actuating structures had nearly the same diameters (43, 47, and 38 µm, respectively), and the applied stress during coiling and actuation were 8 MPa and 33 MPa, respectively, for all structures. All muscles contained the same weight of CNTs per untwisted length and both PEO-SO₃@CNT muscles have similar linear densities (0.033 mg/cm before coiling). The spring indices of the SRAM, the HYAM and the pristine yarn were 2.3, 2.7, and 1.8, respectively. The observed tensile stroke for the coiled pristine yarn is minuscule (2.1%), compared to that of the coiled PEO-SO₃@CNT SRAM (15.1%) and the coiled PEO-SO₃@CNT HYAM (9.6%). The ratio of tensile stroke for the coiled PEO-SO₃@CNT SRAM to that of the coiled PEO-SO₃@CNT HYAM is high (1.57), despite the lower spring index of the SRAM (2.3) compared to that of the HYAM (2.7). The higher average stroke rate of the SRAM (3.2 %/s) compared to that of the HYAM (1.5 %/s) provides an additional performance benefit of the SRAM compared to the HYAM.

For the same coiled PEO-SO₃@CNT SRAM as used above, Fig. S12 shows the dependence of equilibrium isometric tensile stress generation on the concentration of ethanol in dry air. These results indicate that the generated tensile stress is reversibly related to the concentration of ethanol.
vapor, reaching a maximum of 68 MPa at 0.234 mg/L (which corresponds to a saturated atmosphere of ethanol in dry room temperature air). When this SRAM was operated isobarically at 33 MPa using a saturated atmosphere of ethanol in dry air, a tensile contraction of 15.7% was obtained at an average contraction rate of 10.6 % s⁻¹ (Fig. 3A).

While self-coiled structures provide the highest work capacity, because of their low spring indices and correspondingly high tensile moduli, the contraction during tensile actuation can be amplified by using mandrel-coiled muscles having very large spring indices. This amplification is evident in the plots of Fig. S13 of contractile strain versus time for water-vapor-driven actuation of cylindrical and cone-shaped PEO-SO₃@CNT SRAMs that have coiled and supercoiled structures.

4.4 Comparison of thermal and electrothermal tensile actuation of SRAM and hybrid coiled muscles based on PEO-SO₃@CNT and nylon6@CNT yarns

Figure S14 compares the stress dependence of tensile stroke and contractile work capacity for PEO-SO₃@CNT SRAM and a PEO-SO₃@CNT HYAM for a temperature increase from 25°C to 200°C. The contractile work capacity for both the SRAM and the HYAM increases monotonically with applied load, as does the ratio of SRAM to HYAM contractile work capacity (which is 1.5 for a tensile stress of 45 MPa).

The results of Fig. S15 are for the electrothermal actuation of a PEO-SO₃@CNT SRAM, a PEO-SO₃@CNT HYAM, and a pristine CNT yarn by providing a power input per yarn length of 0.18 W/cm. A 0.08 Hz square-wave potential with 50% duty cycle was applied, which provided peak temperatures of 167, 175, 190°C, respectively, for the three types of yarns. The tensile contraction and load-optimized contractile work capacity of the SRAM (8.04% and 0.65 J/g) was significantly larger than for the HYAM (5.53% and 0.39 J/g), and the tensile contraction and load-optimized contractile work capacity of the pristine CNT yarn (1.95% and 0.10 J/g) were quite small. The ratios of tensile stroke and maximum contractile work capacity (up to loads that cause muscle damage) of the SRAM to that of the HYAM were 1.45 and 1.67, respectively.

The results of Fig. S16 show that, for all temperature changes, the thermal actuation stroke of a nylon6@CNT SRAM is significantly larger than a nylon6@CNT HYAM, and that the thermal stroke of a pristine CNT yarn is too small to be useful. For these comparative results, the same amount of CNT sheet was used to prepare the three yarns and the weight of nylon 6 per muscle length was the same for the SRAM and for the HYAM, which resulted in similar linear density of 0.033 mg/cm for both muscles. For these results, measured using a temperature scan rate of 5°C/minute, there is no hysteresis between data taken on increasing temperature and decreasing temperature for the nylon6@CNT SRAM and for the pristine CNT muscle. The small hysteresis observed for the nylon6@CNT HYAM likely reflects the slower thermal equilibration of the HYAM, compared to that for the SRAM and pristine yarn.

4.5 Comparison of electrochemical tensile actuation of a coiled CNT@nylon6 SRAM and a coiled CNT yarn in 0.2 M TBA·PF₆ (tetrabutyl ammonium hexafluorophosphate in propylene carbonate)

The cyclic voltammetry curves of Fig. S17A, B were used to obtain the overall electrical energy input during tensile actuation of a coiled CNT@nylon6 SRAM and a coiled CNT yarn. Since the electrolyte in the coiled CNT yarn is a guest whose volume change drives actuation, this coiled CNT yarn is a HYAM. As shown here, the electrical energy input during muscle charging and the electrical energy output during muscle discharge for the SRAM (81.5 and -52.5 J/g) and for the CNT yarn (73.7 and -47.3 J/g) are similar. Hence, the net electrical energy cost for an actuation cycle of the SRAM (29 J/g) is slightly higher than that for the CNT yarn (26.4 J/g), which
is consistent with the fact that Fig. 5C contractile mechanical energy output (2.26 J/g) for the SRAM is higher than for the CNT yarn (1.36 J/g). Consequently, the contractile energy efficiency of the SRAM is 7.18% versus 4.97% for the CNT yarn. This contractile efficiency for the hybrid CNT muscle is nearly identical to the previous record for an electrochemically-driven CNT yarn (5.4%) (7), and is much higher than previously reported for an electrochemical muscle that does not employ CNTs. For comparison of these results, there are a few reports in the literature for contractile efficiencies (and none for full-cycle efficiencies) for electrochemical muscles. However, contractile efficiencies of 5.1 x 10^-3%, and <1% have been reported for electrochemically actuated polypyrrole/polyethylene glycol bilayer actuator (4), and an electrochemically actuated CNT/polymer gel tensile muscle (5).

A work loop method (6, 7) was used to calculate a lower bound on the full-cycle energy conversion efficiency of the electrochemically-powered coiled CNT@nylon6 SRAM and coiled CNT muscle. Since over a full mechanical cycle, positive work results during muscle contraction and negative work occurs during muscle expansion, realizing a positive net mechanical work output during the work loop requires that the stress applied during muscle contraction is larger than during muscle expansion. Maximizing this full-cycle energy conversion efficiency involves balancing competing effects. For instance, while the present electrochemical muscles provide the highest full-cycle mechanical energy output at the lowest scan rate, scan rates below 20 mV/s suffered so much self-discharge that low Coulombic efficiencies resulted, which degraded energy conversion efficiencies. Using the experimentally determined dependence of muscle stroke on the applied force (Fig. S18), we determined the isobaric loads applied during muscle contraction and muscle expansion that maximize the work done during the work loop. These work loops are shown in Figs. S19A and S20A for the CNT@nylon6 SRAM and CNT muscle, respectively. Electrochemical measurements enabled calculation of the electrical input energy and the electrical energy recovered during the actuation cycle at 20 mV/s scan rate, which are 105 and 44 J/g for the CNT@nylon6 SRAM (Fig. S19B) and 110 and 44 J/g for the hybrid CNT muscle (Fig. S20B). During the actuation cycles of Figs. S19A and S20A, the CNT@nylon6 SRAM and CNT muscle produced 1.25 and 0.95 J/g of mechanical work, respectively. Using these gravimetric full-cycle work capacities and the above net gravimetric electrical energy input, full-cycle energy conversion efficiencies of 2.04% and 1.37% were obtained for the SRAM and CNT muscle, respectively (Fig. S21). These time-consuming measurements of full-cycle energy conversion efficiencies were repeated using newly fabricated yarns, providing a contractile efficiency (6.64%) and a full-cycle efficiency (1.94%), which are in good agreement with the above initial measurement results (7.18% and 2.04%, respectively). Like here demonstrated for both thermal and electrochemical contractile efficiencies, these results show that the electrochemical SRAM provides a higher full-cycle energy conversion efficiency than does the electrochemical CNT muscle. These full-cycle energy conversion efficiencies for the CNT muscle is close to that previously reported (7) for an electrochemically driven hybrid CNT yarn muscle (1.5%).

4.6 Maximum average contractile power
Maximum average contractile power is defined as the maximum ratio of contractile work to actuation time. Results for maximum average contractile power for tensile actuation of various type of SRAMs are shown in Figure S22. These results are for the tensile load that maximizes contractile work.

5. Prediction of the effect of sheath/core ratio on the absorption-driven torsional actuation of a
SRAM

Corresponding to the method presently used to make a twisted SRAMs, this theoretical analysis considers a SRAM core yarn having an initial twist of $T_0$ (turns per muscle length), which is clamped and coated with the sheath material until the sheath dries to set the initial state of the SRAM. If the yarn was unclamped before the sheath was applied, then the yarn will decrease twist by $\Delta T_0$ due to elastic removal of most of the inserted twist. The fully dried SRAM is unclamped at one end allowing rotation that removes a twist of $\Delta T_{off}$ from the core yarn, and introduces twist into the SRAM’s sheath. In the equilibrium non-actuated state, the torque on the CNT core is balanced by the opposing torque in the elastically twisted sheath, so that:

$$0 = k_{\text{core}}^\text{on}(\Delta T_0 + \Delta T_{\text{free,off}} - \Delta T_{\text{off}}) - k_{\text{sheath}}^\text{off}\Delta T_{\text{off}},$$

(5.1)

where $k_{\text{core}}$ and $k_{\text{sheath}}$ are the torsional stiffnesses of the core and sheath, respectively, and $\Delta T_{\text{free,off}}$ is the change in core twist due solely to the compression applied by the drying sheath. Rearranging Eqn. 5.1 gives:

$$\Delta T_{\text{off}} = (\Delta T_0 + \Delta T_{\text{free,off}})(1 + k_{\text{sheath}}^\text{off}/k_{\text{core}}^\text{on}).$$

(5.2)

Using the above approach, we can now calculate the twist change ($\Delta T_{\text{on}}$) in going from initial yarn twist ($T_0$) to the twist that results from actuation of the SRAM. Now the torque balance equation is written as:

$$0 = k_{\text{core}}^\text{on}(\Delta T_0 + \Delta T_{\text{free,on}} - \Delta T_{\text{on}}) - k_{\text{sheath}}^\text{on}\Delta T_{\text{on}}$$

(5.3)

Hence,

$$\Delta T_{\text{on}} = (\Delta T_0 + \Delta T_{\text{free,on}})(1 + k_{\text{sheath}}^\text{on}/k_{\text{core}}^\text{on}).$$

(5.4)

The torsional stroke of the SRAM (in turns per muscle length) is:

$$\Delta T_{\text{SRAM}} = \Delta T_{\text{on}} - \Delta T_{\text{off}}.$$  

(5.5)

Since the modulus of the yarn in the axial direction is much lower than that in radial directions, we can approximate that the length of the SRAM does not change during torsional actuation. This approximation is justified the results of Fig. S10, which show that a torsional non-tethered twisted PEO-SO3@CNT SRAM undergoes little change in length during ethanol-vapor-driven torsional actuation, and that this tensile stroke is a 1.25% contraction. On the other hand, the SRAM’s diameter increases by 9% during this actuation.

Hence, we approximate that the change in twist of the yarn core occurs without energetically costly changes in nanotube length (which is the string length in the helix model) or SRAM length. Correspondingly, the helix equation (1) gives that the fractional change in twist equals the fractional change of the diameter of the core yarn. Consequently, we obtained that:

$$\Delta T_{\text{free,on}} = (T_0 + \Delta T_0)(d_{\text{core}}^\text{uncoated}/d_{\text{core}}^\text{on} - 1)$$

(5.6)

$$\Delta T_{\text{free,off}} = (T_0 + \Delta T_0)(d_{\text{core}}^\text{uncoated}/d_{\text{core}}^\text{off} - 1),$$

(5.7)

where $d_{\text{core}}$ parameters are the yarn core diameters in the uncoated ($d_{\text{core}}^\text{uncoated}$), actuation-off ($d_{\text{core}}^\text{off}$), and actuation-on ($d_{\text{core}}^\text{on}$) states.

The core diameter of the SRAM after drying in the non-actuated state ($d_{\text{core}}^\text{off}$) is estimated by first considering the compression ratio $\lambda = d_{\text{core}}^\text{on}/d_{\text{core}}^\text{off}$. This compression ratio, which is a function of the sheath/core ratio (SCR), can be evaluated from elasticity theory by drawing analogy to the case where a core cylinder is inserted into a hollow tube that has an initial inner radius that is smaller
than the initial radius of the core cylinder. Using literature results \( \delta \), \( \lambda \) is obtained from:

\[
\lambda = 1 - (1 - \phi)(\lambda E_c/( (1 + 2SCR/\lambda)^2 + 1) + \nu)/(1 - \nu_s + \lambda),
\]

where \( \phi \) is the ratio of the sheath thickness in the non-actuated state to the sheath thickness in the actuated state. \( E_s \) and \( E_c \) are the elastic moduli of the sheath and the core, respectively, in the radial direction, and \( \nu_s \) and \( \nu_c \) are the corresponding Poisson’s ratios that couple a stretch in the circumferential direction to the deformation in the radial direction.

We will now apply these theoretical results for the specific case of predicting the dependence of torsional actuation on sheath/core ratio for ethanol-driven actuation of a PEO-SO₃@CNT SRAM. Since experimental measurements show that the Young’s modulus for the sheath of the ethanol-saturated SRAM is very low (50.4 MPa), the stresses on the SRAM core are similar for the actuated state and the initial uncoated state (which are like those for the undried-sheath state). Hence, we approximate that \( d_{\text{Uncoated}}^\text{core} = d_{\text{initial}}^\text{core} \).

Solving Eqns. 5.5-5.8, using measured values of the Young’s moduli of sheath and core (294 MPa and 354 MPa, respectively, in the non-actuated state), the Poisson’s ratios of sheath and core (0.40 and 0.18, respectively), and the relationship for an isotropic material between these parameters and the shear modulus, shear moduli of 105 MPa and 150 MPa are calculated for the non-actuated state of the PEO-SO₃ sheath and the CNT core yarn, respectively. The relationship between shear modulus \( (G) \) and torsional stiffness \( (k) \) for a rod of length \( (L) \) and cross-sectional polar 2nd moment of area \( (J) \), which is \( k = GJ/L \), is here exploited. Calculation of the torsional stiffness for the HYAM proceeds analogously, except that the rule-of-mixtures is used to calculate the effective shear modulus of the HYAM. Consequently, for the non-actuated state, we obtained the torsional stiffness of the sheath of the SRAM (2.35×10⁻² N·m/rad), the core of the SRAM (2.09×10⁻² N·m/rad), and the yarn host of the HYAM (3.30×10⁻² N·m/rad), the guest of the HYAM (3.30×10⁻² N·m/rad). Similarly, using measured values of the Young’s moduli of sheath and core (50.4 MPa and 302.8 MPa, respectively, in the actuated state), the Poisson’s ratios of sheath and core (0.4 and 0.18, respectively) and the relationship for an isotropic material between these parameters and the shear modulus, shear moduli of 18 MPa and 128 MPa are calculated for the actuated state of the PEO-SO₃ sheath and the CNT core yarn, respectively. Likewise, for the actuated state, we obtained the torsional stiffness of the sheath of the SRAM (5.80×10⁻² N·m/rad), the core of the SRAM (2.09×10⁻² N·m/rad), and the yarn host of the HYAM (3.30×10⁻² N·m/rad), the guest of the HYAM (3.30×10⁻² N·m/rad).

Using these experimental measurements of sheath and core properties, and the approximation that \( d_{\text{Uncoated}} = d_{\text{initial}} \) (and using no fit parameters), exceptional agreement was obtained between the predicted and observed dependence of torsional actuation on the sheath/core ratio (Fig. 2E). It can be seen that the theory predicts zero torsional stroke when the sheath/core ratio is either zero or infinite. In addition, there is an optimum value of the sheath/core ratio for maximizing torsional stroke.

6. Predicted tensile stroke and isobaric contractile work capacity of coiled SRAMs and HYAMs

While the calculations in this section pertain to any means of driving actuation, we here focus discussion on the specific case where actuation results from vapor or liquid absorption and desorption. Hence, mechanical parameters have subscripts “dry” or “wet” depending upon whether they pertain to the non-actuated or actuated states. The dependence of muscle stroke \( (\Delta L) \) and contractile work capacity \( (W) \) on isobaric load \( (F) \) were calculated for both SRAMs and HYAMs from the force-extension curves shown in Fig. S24, which approximate that the helical springs in non-actuated and actuated states have strain-independent spring stiffnesses, \( K_{\text{dry}} \) and \( K_{\text{wet}} \), Commented [EG2]: This equation should be re-edited.
respectively. The isobaric tensile stroke becomes
\[ \Delta L = \delta_1 + \Delta L_0 - \delta_2 = \Delta L_0 - F(1/K_{\text{wet}} - 1/K_{\text{dry}}), \] (6.1)

where \( \delta_1 = F/K_{\text{dry}} \) and \( \delta_2 = F/K_{\text{wet}} \). \( \Delta L_0 \) is defined as the free tensile stroke, which is the stroke that would arise for a vanishing load if inter-coil contact did not occur. This free stroke would also be the observed stroke if the spring stiffnesses for the muscle were the same in the actuated and non-actuated states. When actuation takes place under isotonic loading, the area enclosed by the rectangle ABCD provides the contractile work capacity, which is:
\[ W = F\Delta L = F(\delta_1 + \Delta L_0 - \delta_2) = F\Delta L_0 - F^2(1/K_{\text{wet}} - 1/K_{\text{dry}}). \] (6.2)

Equations 6.1 and 6.2 show that isobaric tensile stroke and contractile work capacity are determined by both the free stroke \( \Delta L_0 \) and the stiffness change \( 1/K_{\text{wet}} - 1/K_{\text{dry}} \). However, the two terms provide opposite effects: contractile stroke \( \Delta L_0 \) is produced when the muscle volume expands during activation to produce muscle untwist, while the stiffness change (from non-activated to activated state) leads to an extension of the spring when \( K_{\text{wet}} < K_{\text{dry}} \), which is usually the case.

In order to complete the calculations, we next derive the free stroke \( \Delta L_0 \). Based on the twist-driven coil actuation mechanism, the free stroke is \( \Delta L_0 = l_2 \Delta T/N \), where \( l \) is the fiber length, \( \Delta T \) is the fiber untwist (torsional actuation) due to guest volume expansion and modulus decrease, and \( N \) is the number of coils in the fiber length.

Using a theoretical framework that is similar to the one used for non-coiled muscles, the torsional actuation stroke \( \Delta T \) can be estimated as follows: In the case of a coiled SRAM, the torque on the CNT core is balanced by the external torque \( TQ \) needed to provide torsional tethering. In the non-actuated state,
\[ TQ = k_{\text{off}} \alpha (\Delta T_0 + \Delta T_{\text{free,off}}), \] (6.3)
where \( k_{\text{off}} \alpha \) is the torsional stiffness of the core in the non-actuated state, and \( \Delta T_{\text{free,off}} \) is the change in core twist due solely to the compression applied by the drying sheath. The torque balance equation in the actuated state is written as:
\[ TQ = k_{\text{on}} \alpha (\Delta T_0 + \Delta T_{\text{free,on}} - \Delta T) - k_{\text{on,sheath}} \Delta T, \] (6.4)

Combining Eqs. 6.3 and 6.4 gives
\[ k_{\text{wet}} \alpha (\Delta T_0 + \Delta T_{\text{free,off}}) = k_{\text{act}} \alpha (\Delta T_0 + \Delta T_{\text{free,on}} - \Delta T) - k_{\text{act,sheath}} \Delta T, \] (6.5)
which provides the following for the torsional stroke of the SRAM (in turns per muscle length):
\[ \Delta T = [k_{\text{wet}} \alpha (\Delta T_0 + \Delta T_{\text{free,off}}) + k_{\text{act}} \alpha (\Delta T_0 + \Delta T_{\text{free,on}})]/(k_{\text{act}} + k_{\text{act,sheath}}). \] (6.6)

Using an essentially identical approach, the torsional stroke for the HYAM is:
\[ \Delta T = (k_{\text{wet}} \alpha (\Delta T_0 + \Delta T_{\text{free,off}}) + k_{\text{act}} \alpha (\Delta T_0 + \Delta T_{\text{free,on}}))/(k_{\text{act}} + k_{\text{act,sheath}}). \] (6.7)
where \( k_{\text{wet}} \alpha \) and \( k_{\text{act}} \alpha \) are the torsional stiffnesses of the host in the non-actuated and actuated state, respectively.

These calculations of torsional stroke for a coiled PEO-SO3@CNT SRAM and coiled PEO-SO3@CNT HYAM use the same torsional stiffnesses as used for predicting the torsional stroke of a twisted SRAM and a twisted HYAM in Section 5. The resulting \( \Delta T \) is 0.17 turns/mm for the coiled SRAM and 0.11 turns/mm for the coiled HYAM.
We next predict the free tensile stroke ($\Delta L_0$) for an ethanol-vapor-driven coiled PEO-SO$_3$@CNT SRAM and for an ethanol-vapor-driven coiled PEO-SO$_3$@CNT HYAM using the relationship that $\Delta L_0 = \frac{F \Delta T}{N}$. For the investigated muscles, the number of coils ($N$) in the SRAM and in the HYAM were $N = 106$ and $N = 116$, respectively and the fiber length within the coiled SRAM and HYAM were $l = 5.6$ cm and $l = 5.8$ cm, respectively. Using the $\Delta T$ values predicted using Eqns. 6.6 and 6.7 (0.17 and 0.11 turns/mm for the SRAM and HYAM, respectively), $\Delta L_0$ becomes 5.1 mm and 3.1 mm for the SRAM and HYAM, respectively.

Using Eqns. 6.1 and 6.2, respectively, and the above values of $\Delta L_0$, provides the predicted dependence of tensile stroke and contractile work capacity on the applied isobaric stress. As shown in Fig. S25, the theoretically and experimentally obtained tensile stroke and work capacities are in exceptional agreement for stress levels that are sufficiently high that coil-coil contact does not limit actuation. While the results in the above figures are for tensile stroke normalized to the non-loaded length of the coiled muscles, Fig. 3B shows the agreement obtained when tensile stroke is normalized to loaded muscle length.
Fig. S1. The effect of yarn topology on the maximum torsional stroke and torsional rotation speed of a PEO-SO₃@CNT SRAM while driven by a saturated atmosphere of ethanol in dry air. These results are for three different scroll types: an Archimedean yarn (45-µm-diameter, 74 turns/cm), a Fermat yarn (44-µm-diameter, 69 turns/cm), and a dual-Archimedean yarn (49-µm-diameter, 71 turns/cm). The applied load during twist insertion was ~8 MPa.

Fig. S2. SEM micrographs of: (A) A 76-µm-diameter pristine yarn made from electrospun PAN nanofibers having an average diameter of 245 nm (left) and the fracture surface of an 87-µm-diameter PEO-SO₃@PAN SRAM (sheath/core ratio of 0.073) that was made from the yarn in the left (right). (B) The time dependence of torsional stroke and rotation speed for one reversible sorption/desorption cycle for an 87-µm-diameter PEO-SO₃@PAN SRAM (black), a 92-µm-diameter PEO-SO₃@PAN HYAM (blue), and a 76-µm-diameter pristine yarn (red). The same 76-µm-diameter PAN yarn, containing 23 turns/cm of twist, was used for the neat muscle and for the fabrication of the SRAM and HYAMs. Also, the SRAM and HYAM had about the same weight (4.8 mg/cm).
Fig. S3. SEM images of the cross-sections of (A) a PEO-SO$_3$@CNT SRAM (Inset: SEM micrograph of PEO-SO$_3$@CNT SRAM yarn after tensile rupture, which removed parts of the core yarn. The scale bar of inset image is 20 μm) and (B) a PEO-SO$_3$@CNT hybrid yarn muscle, which were obtained by yarn sectioning using ion milling. The core CNT yarn in the SRAM has a diameter of 37 μm and the PEO-SO$_3$ polymer sheath is 5 μm thick.

Fig. S4. (A) Young’s modulus as a function of time during the sorption and desorption of ethanol by a free-standing, 137-μm-thick PEO-SO$_3$ film. This modulus (black line and data points) was obtained by the pictured cycling of 7% strain at 0.8 Hz (blue line). Ethanol sorption (using a saturated atmosphere of ethanol in dry air) was continued until the modulus became essentially constant (at about 10 s, where the weight of the film had increased by 16.3 %). Thereafter, desorption of ethanol occurred in dry air using the same gas flow as for ethanol sorption. (B) Thermal gravimetric analyses of the PEO-SO$_3$ film and of the PEO-SO$_3$@CNT hybrid yarn at a heating rate of 5°C/min in air.
**Fig. S5.** (A) The equilibrium volume change of PEO-SO$_3$, PVA, and nylon 6 films when exposed to a saturated atmosphere of ethanol in dry air. (B) Comparison of equilibrium torsional strokes for an applied stress of 0.033 MPa (and the product of torsional stoke and core yarn diameter, normalized to the percent polymer volume change) for a 37.5-µm-diameter, non-coiled CNT yarn (twist density = 74 turns/cm) that is coated with either PEO-SO$_3$, PVA, or nylon 6, using the same conditions as in (A). The sheath/core ratios for the resulting PEO-SO$_3$@CNT, PVA@CNT, and nylon6@CNT SRAMs were 0.14, 0.17, and 0.12, respectively. These torsional strokes resulted from exposure of the SRAMs to a saturated atmosphere of ethanol in dry air.

**Fig. S6.** The dependence of torsional stroke on the measured bias angle ($\alpha$) of a CNT core yarn on a linear scale (before sheath deposition) for a twisted PEO-SO$_3$@CNT SRAM. These torsional strokes are equilibrium values for a SRAM exposed to a saturated atmosphere of ethanol in dry air. The lower-top axis is the inserted twist per yarn length ($T$), which is calculated using the bias angle of the core yarn and the yarn diameter ($D$, which is provided by the upper-top axis) at this degree of inserted twist using $T = \tan(\alpha)/\pi D$. Yarn coiling does not initiate until the bias angle exceeds 43.4°, at which point the diameter of the CNT yarn is 36.5 µm. After over coating with PEO-SO$_3$, this diameter yarn provides a SRAM having a sheath/core ratio of 0.14.
Fig. S7. Effects of sheath/core ratio on SRAM fabricability and performance. (A) Combinations of sheath/core ratio and yarn bias angle (prior to sheath coating) that result in sheath cracking (red region) during SRAM fabrication and combinations that result in crack-free sheaths (white region). (B) The dependence of torsional stroke and maximum rotation speed on sheath/core ratio for a non-coiled 39-µm-diameter PEO-SO$_3$@CNT SRAM containing 74 turns/cm of twist, which has a bias angle of 42° before sheath coating. Inset: SEM micrographs at two magnifications showing the sheath cracking that occurs for sheath/core ratios that are in the red-colored region of the data plot. The scale bars are 150 µm (left) and 30 µm (right).
Fig. S8. (A) Vapor-driven torsional actuation (using vapor-saturated dry air) of a twisted PEO-SO₃@CNT SRAM under 0.1 MPa tensile stress. (B) Liquid-immersion-driven tensile actuation of a coiled PEO-SO₃@CNT SRAM. All SRAMs were made from the same 37.5-µm-diameter CNT yarn containing 74 turns/cm of inserted twist. The spring index of the coiled SRAM was 2.3. The applied tensile stress for these isobaric tensile actuation measurements was 33 MPa. (C) Tensile stroke (black symbols) and contractile work capacity (blue symbols) versus applied isobaric stress for a PEO-SO₃@CNT SRAM (squares), a PEO-SO₃@CNT HYAM (triangles), and a pristine CNT yarn (circles), which were all actuated by immersion in liquid ethanol. The maximum contractile work capacity of the SRAM was 1.6 times that of the HYAM.

Commented [MJdA3]: Missing add diameter, twist density and spring index.
Fig. S9. The time-dependence of torsional stroke and torsional rotation speed for water-vapor-driven actuation of a 43.7-µm-diameter nylon6@CNT SRAM, a 47-µm-diameter nylon6@CNT HYAM, and a 39-µm-diameter pristine CNT yarn, which was obtained by switching between 45% RH air and 90% RH air. All yarns contain the same weight of CNTs and both nylon6@CNT muscles have similar linear densities (0.029 mg/cm). The applied tensile stress for these experiments was 0.035 MPa. The twist densities of the SRAM, the HYAM, and the pristine yarn are 36 turns/cm.

Fig. S10. Comparison of the time-dependence of tensile stroke for a torsionally tethered and a torsionally non-tethered twisted PEO-SO₃@CNT SRAM, when muscle contraction resulted from exposure of the muscles to a saturated atmosphere of ethanol in dry air and actuation was reversed by desorbing the ethanol using dynamic pumping. These measurements (for a tensile load of 60 mg) are for the same SRAM as used in Fig. 2B, C, and F.
Fig. S11. Tensile stroke versus time for a coiled PEO-SO$_3$@CNT SRAM (top), a coiled PEO-SO$_3$@CNT hybrid yarn muscle (middle), and a coiled pristine yarn (bottom). Actuation cycles were obtained by exposing the muscles to dry air containing 0.17 mg/L of ethanol and then desorbing the ethanol using dynamic pumping. Before coiling, the diameters of the SRAM, the HYAM, and the pristine yarn were 43, 47, and 38 µm, respectively. The applied stress during coiling and during actuation were 8 MPa and 33 MPa, respectively. All muscles contained the same weight of CNTs per untwisted length and both PEO-SO$_3$@CNT muscles have similar linear densities (0.033 mg/cm before coiling). The spring indexes of the SRAM, the HYAM, and the pristine yarn are 2.3, 2.7, and 1.8, respectively.

Fig. S12. Equilibrium tensile stress generation as a function of ethanol concentration in dry air for isometric tensile actuation of a coiled PEO-SO$_3$@CNT SRAM. The outer diameter of the self-coiled SRAM was 44 µm, its inserted twist was 110 turns/cm and its sheath/core ratio was 0.14.
Fig. S13. Water-vapor-driven tensile actuation (during transition from 55% to 90% RH air) versus time for PEO-SO₃@CNT SRAMs having a sheath/core ratio of 0.14, which were made from a precursor 44-µm-diameter twisted SRAM. Tensile stroke versus time is shown for (A) a twisted SRAM that was coiled on a 0.35-cm-diameter, 1.2-cm-long cone-shaped mandrel, thereby forming a non-uniformly coiled SRAM. (B) A twisted SRAM that was coiled on a 0.2-cm-diameter, 1-cm-long cylindrical mandrel, thereby forming a uniformly coiled SRAM. (C) A self-coiled SRAM that was coiled on a 0.2-cm-diameter, 0.5-cm-long cylindrical mandrel, thereby forming a supercoiled SRAM. The applied tensile stress for these experiments were 0.5 MPa, 0.5 MPa, and 0.7 MPa, respectively. Insets: The structures of these muscles are illustrated in (A-C).

Fig. S14. Comparison of the dependence of tensile stroke and contractile work capacity on applied isobaric stress for a PEO-SO₃@CNT SRAM (square symbols) and a PEO-SO₃@CNT HYAM (triangle symbols) for a temperature increase from 25°C to 200°C. Before coiling, the diameters of the SRAM and the HYAM yarn were 44 and 47 µm, respectively, and the spring index after coiling were 3.2 and 3.5 for the SRAM and HYAM, respectively. The sheath/core ratio of the SRAM was 0.12.
Fig. S15. Comparison of electrothermally-powered tensile actuation for a coiled SRAM, a coiled HYAM, and a coiled pristine yarn when the input power per yarn length was 0.18 W/cm and a 0.08 Hz square-wave potential with 50% duty cycle was applied, which provided peak temperatures of 167, 175, 190°C, respectively. (A) Tensile stroke versus time for a PEO-SO₃@CNT SRAM, a PEO-SO₃@CNT HYAM, and a pristine CNT yarn when electrothermally actuated using the isobaric load that maximized muscle stroke (18, 15, and 19 MPa, respectively). Before coiling, the diameters of the SRAM, the HYAM, and the pristine yarn were 44, 47, and 36 µm, respectively. (B) Tensile stroke and contractile work capacity versus applied isobaric stress for the PEO-SO₃@CNT SRAM, the PEO-SO₃@CNT HYAM, and the pristine CNT yarn of (A), when using the same electrothermal heating conditions as in (A).

Fig. S16. Tensile stroke versus temperature (for increasing and decreasing temperature) for a nylon6@CNT SRAM (black line), a nylon6@CNT HYAM (blue line), and a pristine CNT yarn (red line). The same 39-µm-diameter CNT yarn was used for these experiments and the weight of nylon 6 per muscle length was the same for the SRAM and for the HYAM. The sheath/core ratio of the SRAM was 0.12. The spring index of the CNT yarn within the coiled yarn was 3.2, 3.5, and 1.8 for the SRAM, HYAM, and neat yarn muscles, respectively. Counting the sheath on the SRAM, the coiled SRAM has a spring index of 1.1. The applied tensile load for these experiments was 16.9 MPa. Note that tensile stroke measurements for increasing temperature and decreasing temperature superimposed for the SRAM and pristine CNT yarn, but not for the HYAM, for the thermal mechanical analyzer measurements of 5°C/minute.
**Fig. S17.** Cyclic voltammetry scans at 20 mV/s during the tensile actuation of (A) a CNT@nylon6 yarn and (B) a CNT yarn in 0.2 M TBA·PF₆ (tetrabutyl ammonium hexafluorophosphate in propylene carbonate), which were used to derive the contractile energy conversion efficiencies of electrochemical tensile actuation. The applied tensile stress for these measurements was 22 MPa.

**Fig. S18.** Initial measurements used for optimizing the full-cycle energy conversion efficiencies of the CNT@nylon6 SRAM and the hybrid CNT muscle. The dependence of actuation stroke on the applied force during cyclic voltammetry from 0 to -3.5 V at 20 mV/s scan rate for CNT@nylon6 SRAM (filled black squares) and for a CNT HYAM (open blue squares). The diameters of the CNT@nylon6 SRAM and the hybrid CNT muscle, before coiling, were 87 µm and 72 µm, respectively. The spring indices and lengths of the CNT@nylon6 SRAM and the hybrid CNT muscle were 1.12 and 3.10 cm and 0.98 and 3.60 cm, respectively.
Fig. S19. Electrochemical and work loop measurements enabling calculation of the full-cycle energy conversion efficiency for the electrochemical CNT@nylon6 SRAM (A) The dependence of gravimetric current on the applied inter-electrode voltage during actuation cycle for the electrochemical CNT@nylon6 SRAM, which is used to calculate the input electrical energy during a full electrochemical cycle. (B) The work loop cycle, which is derived from the Fig. S18 results on the electrochemical actuation of a CNT@nylon6 SRAM.

Fig. S20. Electrochemical and work loop measurements enabling calculation of the full-cycle energy conversion efficiency for the electrochemical hybrid CNT muscle (A) The dependence of gravimetric current on the applied inter-electrode voltage during actuation cycle for the electrochemical hybrid CNT muscle, which is used to calculate the input electrical energy during a full electrochemical cycle. (B) The work loop cycle, which is derived from the Fig. S18 results on the electrochemical actuation of a hybrid CNT muscle.
Fig. S21. Using the results of Figs. S18 to S20, the dependence of full-cycle energy conversion efficiency of the CNT@nylon6 SRAM and the CNT HYAM are plotted versus the isobaric force applied during the contractile part of the actuation cycle. For each isobaric force applied during contraction, the data in Fig. S18 enables calculation of the optimum isobaric force during muscle expansion.
Fig. S22. Tensile stroke and maximum average contractile power, which is defined as the maximum ratio of contractile work to actuation time. These results are for the tensile load that maximizes contractile work. (A) Results for the ethanol-vapor-driven PEO-SO$_3$@CNT SRAM of Fig. 3A and a tensile load of 48 MPa. (B) Results for the electro-thermally-driven PU@CNT SRAM of Fig. 3C and a tensile load of 57 MPa. (C) Results for the electro-chemically-driven CNT@nylon6 SRAM of Fig. 5E and a tensile load of 22 MPa. Actuation resulted from switching the inter-electrode voltage from 0.5V to -3.5V. (D) Results for the electro-thermally-driven PEO-SO$_3$@CNT SRAM of Fig. S15A and a tensile load of 48 MPa.

Fig. S23. Reversible actuation that occurs when the SRAM textile is exposed to the artificial perspiration.
Fig. S23. Fabricated SRAM woven textile actuator. (B) Tensile stroke and contractile work capacity versus applied isobaric stress for the woven SRAM actuator.

Fig. S24. Schematic illustration of the force-extension dependence for actuated and non-actuated coils, which is used for the calculation of contractile work capacity. This calculation uses the approximation that the stiffness of the muscle coil is strain independent for both non-actuated (dry) and actuated (wet) states. The symbols in this illustration are defined in this section.

Fig. S25. Comparison of theoretical and experimental results for ethanol-vapor-driven actuation of a PEO-SO3@CNT SRAM and a PEO-SO3@CNT HYAM. (A) Comparison of the stress dependence of theoretically calculated and experimentally measured tensile stroke for a PEO-SO3@CNT SRAM (dashed black line and black squares, respectively) and a PEO-SO3@CNT HYAM (dashed blue line and blue triangles, respectively). (B) Comparison of the stress dependence of theoretically calculated and experimentally measured work capacity for a PEO-SO3@CNT SRAM (dashed black line and black squares, respectively) and a PEO-SO3@CNT HYAM (dashed blue line and blue triangles, respectively).
HYAM (dashed blue line and blue triangles, respectively). This remarkable agreement between theoretical and experimental results was obtained without the use of any free parameter.
Table S1. Structural parameters and sources for host core yarns and the diameters and inserted twist of twisted hybrid and SRAM PEO-SO$_3$@yarn torsional actuators.

<table>
<thead>
<tr>
<th>Core yarn type and origin</th>
<th>Yarn diameter (µm)</th>
<th>Fiber diameter (µm)</th>
<th>Number of fibers in yarn cross-section</th>
<th>Diameter of twisted SRAM (µm)</th>
<th>Yarn bias angle (α) in degrees</th>
</tr>
</thead>
<tbody>
<tr>
<td>CNT yarn (precursor forest from Lintec Corp.)</td>
<td>36.5</td>
<td>0.009</td>
<td>4060</td>
<td>50</td>
<td>42</td>
</tr>
<tr>
<td>PAN nanofiber yarn (electrospun)</td>
<td>76</td>
<td>0.25</td>
<td>304</td>
<td>87</td>
<td>30</td>
</tr>
<tr>
<td>Silk yarn (Sulky of America)</td>
<td>56.0</td>
<td>1.5</td>
<td>33</td>
<td>88.7</td>
<td>18</td>
</tr>
</tbody>
</table>
Table S2. Contractile work densities (J/g) and peak average-power densities of a SRAM divided by that of a HYAM as a function of the applied stress. Bold numbers for these performance figures indicate that they are maximized at the listed applied stress.

<table>
<thead>
<tr>
<th>Type of coiled muscle (muscle drive method)</th>
<th>PEO-SO₃@CNT (ethanol-vapor-driven)</th>
<th>PEO-SO₃@CNT (electrothermally-driven)</th>
<th>PU@CNT (electrothermally-driven)</th>
<th>CNT@nylon (electrochemically-driven)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>@ 13.6 MPa</td>
<td>@ 17.0 MPa</td>
<td>@ 25.0 MPa</td>
<td></td>
</tr>
<tr>
<td>0.56/0.47 = 1.17</td>
<td>0.41/0.18 = 2.27</td>
<td>0.53/0.28 = 1.89</td>
<td>1.82/1.17 = 1.55</td>
<td></td>
</tr>
<tr>
<td>@ 17.5 MPa</td>
<td>@ 19.0 MPa</td>
<td>@ 24.0 MPa</td>
<td>@ 32.0 MPa</td>
<td></td>
</tr>
<tr>
<td>0.81/0.66 = 1.23</td>
<td>0.51/0.24 = 2.12</td>
<td>0.79/0.38 = 2.07</td>
<td>2.26/1.27 = 1.78</td>
<td></td>
</tr>
<tr>
<td>@ 24.5 MPa</td>
<td>@ 24.0 MPa</td>
<td>@ 33.0 MPa</td>
<td>@ 39.0 MPa</td>
<td></td>
</tr>
<tr>
<td>1.12/0.87 = 1.28</td>
<td>0.63/0.30 = 2.10</td>
<td>1.12/0.49 = 2.28</td>
<td>2.21/1.29 = 1.71</td>
<td></td>
</tr>
<tr>
<td>@ 32.0 MPa</td>
<td>@ 38.0 MPa</td>
<td>@ 42.0 MPa</td>
<td>@ 46.0 MPa</td>
<td></td>
</tr>
<tr>
<td>1.61/1.15 = 1.40</td>
<td>0.64/0.34 = 1.88</td>
<td>1.18/0.51 = 2.31</td>
<td>1.87/1.00 = 1.87</td>
<td></td>
</tr>
<tr>
<td>@ 48.0 MPa</td>
<td>@ 48.0 MPa</td>
<td>@ 49.0 MPa</td>
<td>@ 52.0 MPa</td>
<td></td>
</tr>
<tr>
<td>0.59/0.37 = 1.60</td>
<td>1.33/0.62 = 2.15</td>
<td>1.42/0.73 = 1.95</td>
<td></td>
<td></td>
</tr>
<tr>
<td>@ 55.0 MPa</td>
<td>@ 57.0 MPa</td>
<td>@ 55.0 MPa</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2.03/1.01 = 2.01</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>@ 66.0 MPa</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Contractile work density of a SRAM divided by that of a HYAM as a function of the applied stress</th>
</tr>
</thead>
<tbody>
<tr>
<td>@ 13.6 MPa</td>
</tr>
<tr>
<td>0.06/0.03 = 2.00</td>
</tr>
<tr>
<td>@ 13.6 MPa</td>
</tr>
<tr>
<td>0.14/0.06 = 2.28</td>
</tr>
<tr>
<td>@ 17.5 MPa</td>
</tr>
<tr>
<td>0.17/0.08 = 2.13</td>
</tr>
<tr>
<td>@ 24.5 MPa</td>
</tr>
<tr>
<td>0.21/0.10 = 2.12</td>
</tr>
<tr>
<td>@ 33.0 MPa</td>
</tr>
<tr>
<td>0.22/0.11 = 1.88</td>
</tr>
<tr>
<td>@ 48.0 MPa</td>
</tr>
<tr>
<td>0.20/0.13 = 1.60</td>
</tr>
<tr>
<td>@ 55.0 MPa</td>
</tr>
<tr>
<td>0.28/0.15 = 1.95</td>
</tr>
<tr>
<td>@ 55.0 MPa</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Peak average-power density of a SRAM divided by that of a HYAM as a function of the applied stress</th>
</tr>
</thead>
<tbody>
<tr>
<td>@ 13.6 MPa</td>
</tr>
<tr>
<td>0.27/0.13 = 2.10</td>
</tr>
<tr>
<td>@ 17.0 MPa</td>
</tr>
<tr>
<td>0.18/0.09 = 1.89</td>
</tr>
<tr>
<td>@ 13.6 MPa</td>
</tr>
<tr>
<td>0.41/0.18 = 2.27</td>
</tr>
<tr>
<td>@ 19.0 MPa</td>
</tr>
<tr>
<td>0.53/0.28 = 1.89</td>
</tr>
<tr>
<td>@ 24.0 MPa</td>
</tr>
<tr>
<td>0.79/0.38 = 2.07</td>
</tr>
<tr>
<td>@ 33.0 MPa</td>
</tr>
<tr>
<td>1.12/0.49 = 2.28</td>
</tr>
<tr>
<td>@ 38.0 MPa</td>
</tr>
<tr>
<td>1.18/0.51 = 2.31</td>
</tr>
<tr>
<td>@ 49.0 MPa</td>
</tr>
<tr>
<td>1.42/0.73 = 1.95</td>
</tr>
<tr>
<td>@ 55.0 MPa</td>
</tr>
<tr>
<td>3.52/1.51 = 2.33</td>
</tr>
<tr>
<td>@ 48.0 MPa</td>
</tr>
<tr>
<td>0.22/0.11 = 1.88</td>
</tr>
<tr>
<td>@ 48.0 MPa</td>
</tr>
<tr>
<td>0.24/0.122 = 1.96</td>
</tr>
<tr>
<td>@ 49.0 MPa</td>
</tr>
<tr>
<td>0.37/0.19 = 1.87</td>
</tr>
<tr>
<td>@ 52.0 MPa</td>
</tr>
<tr>
<td>1.74/0.93 = 1.90</td>
</tr>
<tr>
<td>@ 52.0 MPa</td>
</tr>
<tr>
<td>0.20/0.13 = 1.60</td>
</tr>
<tr>
<td>@ 55.0 MPa</td>
</tr>
<tr>
<td>0.26/0.122 = 2.19</td>
</tr>
<tr>
<td>@ 57.0 MPa</td>
</tr>
<tr>
<td>0.28/0.15 = 1.95</td>
</tr>
<tr>
<td>@ 55.0 MPa</td>
</tr>
</tbody>
</table>

Movie S1.

Movie S2.

References and Notes


2. T. Wang, S. Kumar, Electrospinning of polyacrylonitrile nanofibers. J. Appl. Polym. Sci. 102,


