SUPPLEMENTARY INFORMATION

Hydro-Actuation of Hybrid Carbon Nanotube Yarn Muscles

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Calculation details

The relationship between the peak rotation speed (v_{max}) and the minimum rotation period (t_{min}) is

$$v_{max}[r.p.m.] = \frac{1}{t_{min}[s]} \times 60 \qquad (1).$$

For the 8,032-turns/m yarn, the paddle's minimum rotation period could be obtained by the feedback of the laser displacement sensor, which was 43 ms. So according to equation (1), the peak rotation speed was 1,395 r.p.m..

In the rotation experiment, moment of inertia (I) of the rectangular paddle with a weight (M) of 33.4 mg and length (L) of 8 mm is calculated as $I = \frac{ML^2}{12} = 1.8 \times 10^{-10}$ kg m^2. The paddle rotated around half circle in the initial 0.12 s, so the acceleration (a) could be estimated as 436 rad/s^2. The specific torque is $\tau = \frac{Ia}{m}$ = 0.52 N m/kg, where m (150 μg) is the weight of the hybrid yarn.

In the contraction experiment, the energy is totally converted into the gravitational potential energy of the hanging weight, omitting some energy loss from slight rotation (when the paddle was in light mass) and friction (from the gliding between the edge of the paddle and the glass slide). The paddle is lifted up to the maximum height (H) by the coiled yarn. So the specific energy density is $\frac{MgH}{m}$, where g is the acceleration of gravity. For example, when the paddle with a weight of 71.4 mg was lifted up with a maximum height of 19.53 mm (Supplementary Fig. 5) by the 9,972-turns/m coiled yarn (141 μg), the specific energy density was calculated as 96.9 J/kg. The energy loss from rotation is estimated as $\tau\theta$, where τ is the specific torque and θ is the rotation angle. Another portion of energy loss from friction is determined to be not more than $\frac{\mu Mg cos\alpha}{m sin\alpha}$, where μ is the coefficient of kinetic friction between the copper paddle and the glass slide, and α is the inclination angle of the glass slide relative to the horizontal direction. Given the substitution of τ = 0.52 N m/kg, θ = 45° (Supplementary Fig. 5), μ = 0.2[S1] (considering that the surfaces of the copper paddle and the glass slide are both very smooth), α = 78.5° (Fig. 3a), the energy consumption from the rotation and friction were 0.41 J/kg and 3.9 J/kg respectively, which are rather small compared to the transformation of gravitational potential energy and thus can reasonably be omitted.

Supplementary figures

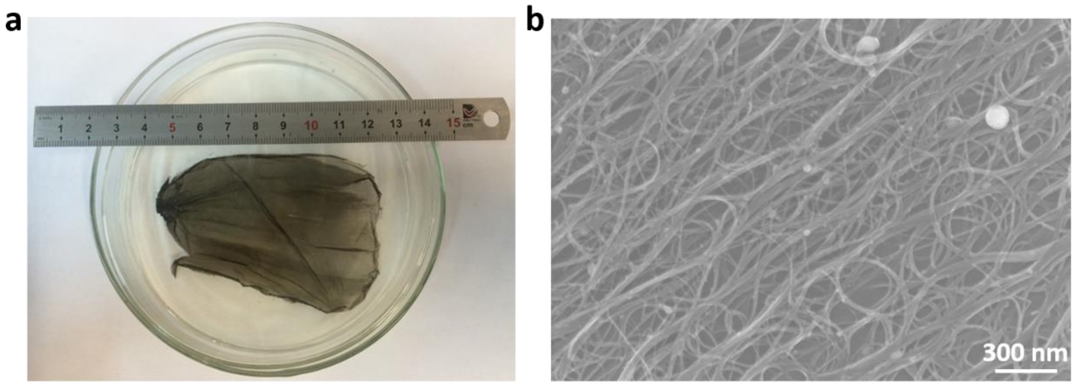

Fig. S1 Optical photograph (a) and SEM image (b) of the pristine freestanding SWCNT film. The reticulate film is composed of entangled SWCNT bundles. The strong junctions formed at high temperature contribute to the excellent mechanical, electrical and thermal properties.

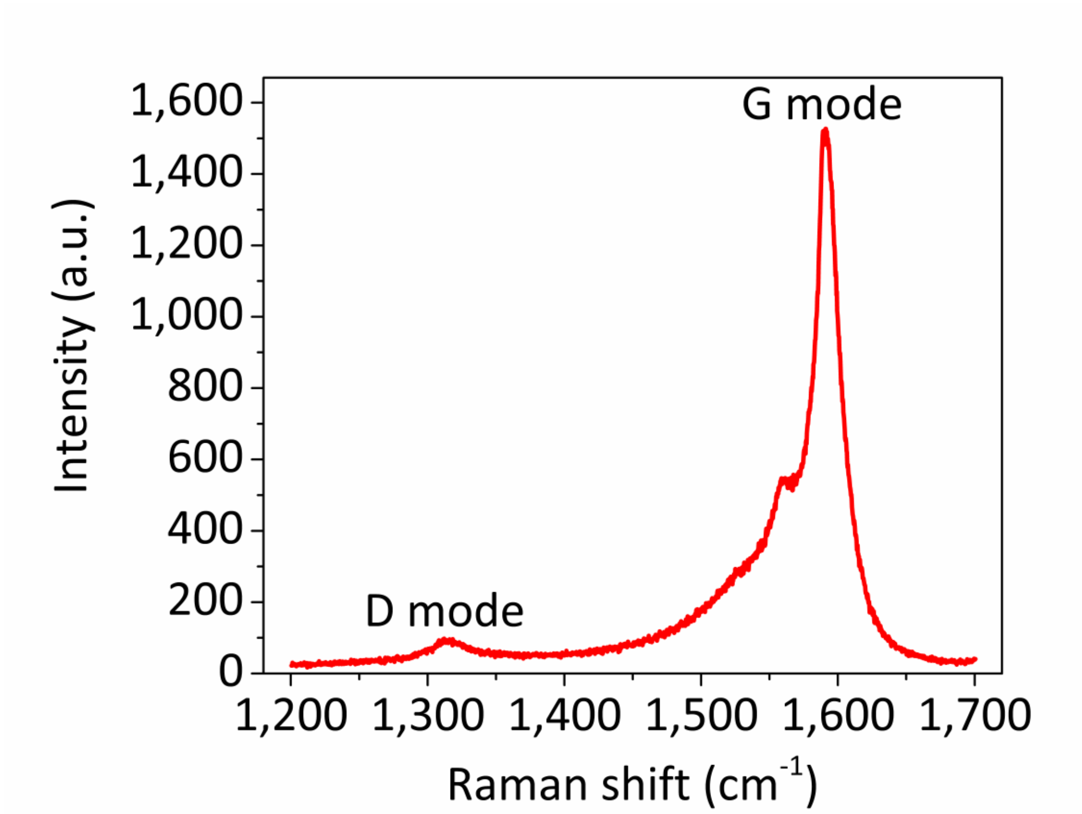

Fig. S2 Raman spectra of the pristine SWCNT film. The intensity ratio of D mode to G mode is ~ 0.064, indicating few defects and fine crystallization of the nanotubes.

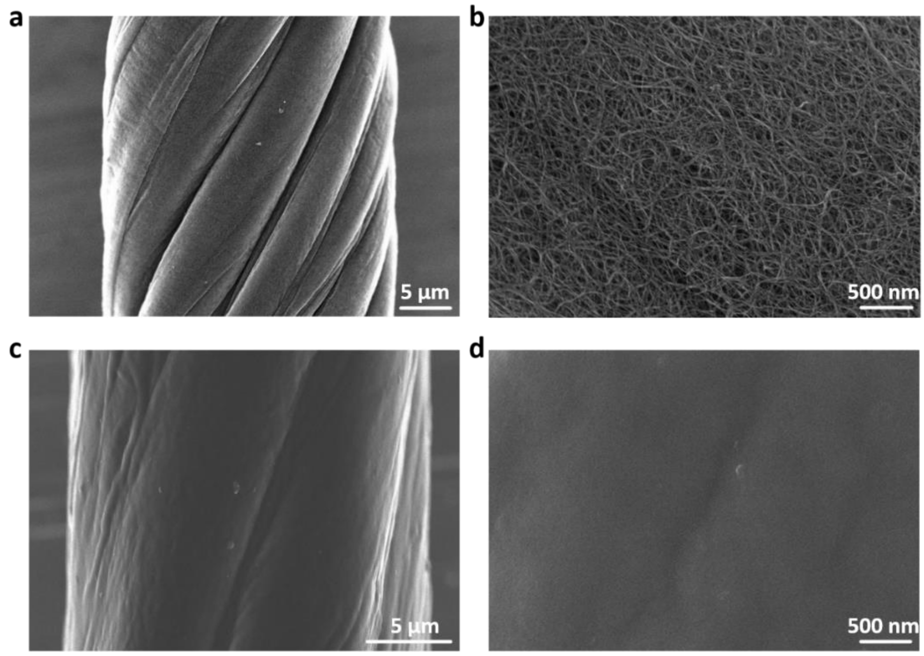

Fig. S3 SEM images of the pure yarn made of pristine CNTs (a, b) and the hybrid yarn with core-shell structure (c, d). This hybrid yarn in (c, d) was made by first twisting the pristine CNT ribbon into a yarn and then infiltrating it with PEDOT:PSS aqueous solution.

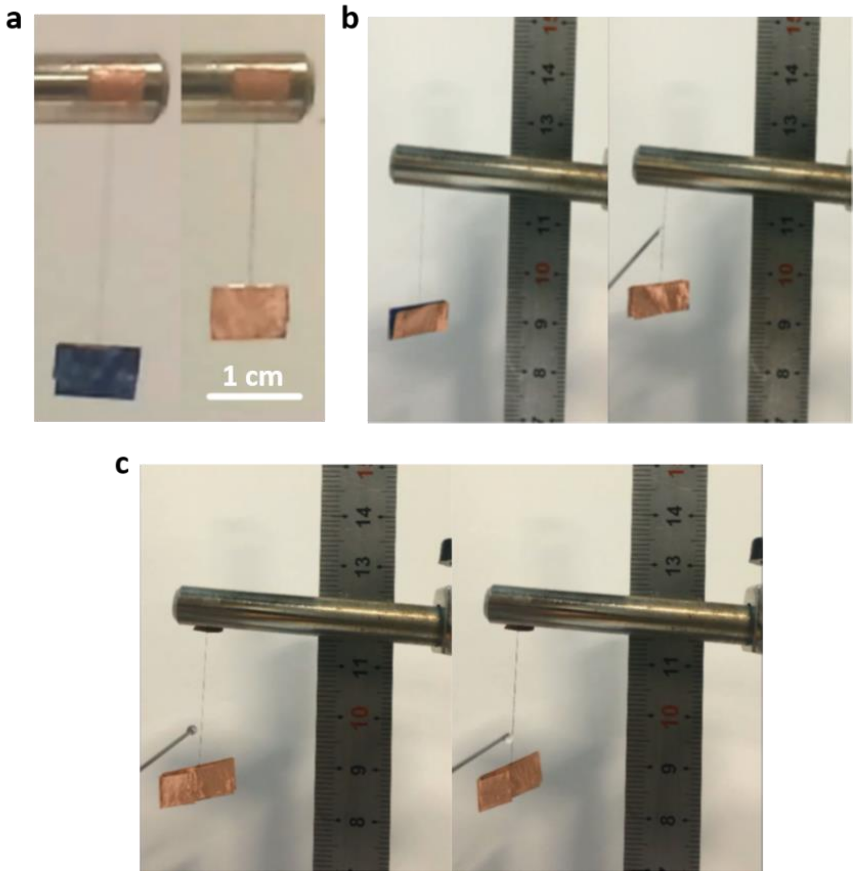

Fig. S4 Optical photograph for contraction. The coiled hybrid yarn is 25 mm in length and 9,736 turns/m in twist density. The paddle-to-yarn mass ratio is 350 (a), 1,300 (b), 3,300 (c), respectively.

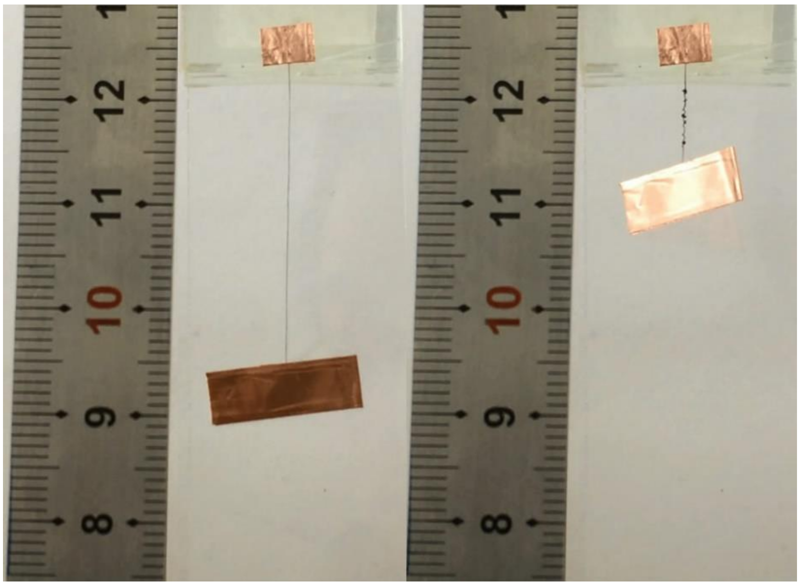

Fig. S5 Optical photograph for contraction. The coiled hybrid yarn is 29 mm in length and 9,972 turns/m in twist density. The corresoponding stress applied by the paddle is ~ 1 MPa. The diameter (~ 30 μm) of the lower part, where the yarn is close to the paddle, is adopted for the calcuation of the stress.

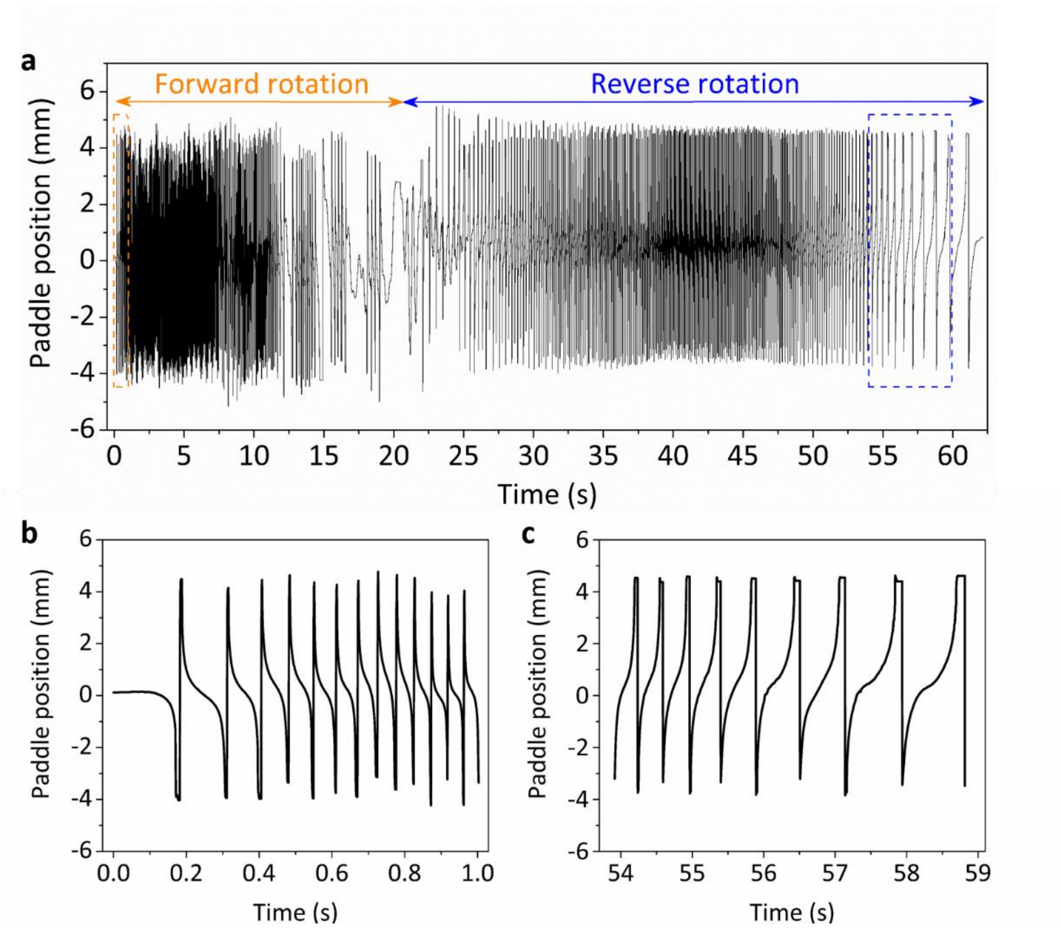

Fig. S6 (a) Feedback by the laser displacement sensor of the whole rotating process. The non-coiled hybrid yarn is 45 mm in length and 8,032 turns/m in twist density. The paddle's moment of inertia is 1.8×10^{-10} kg m^2. (b, c)

Representative wave pattern. Forward (b) and reverse (c) rotation can be readily distinguished.

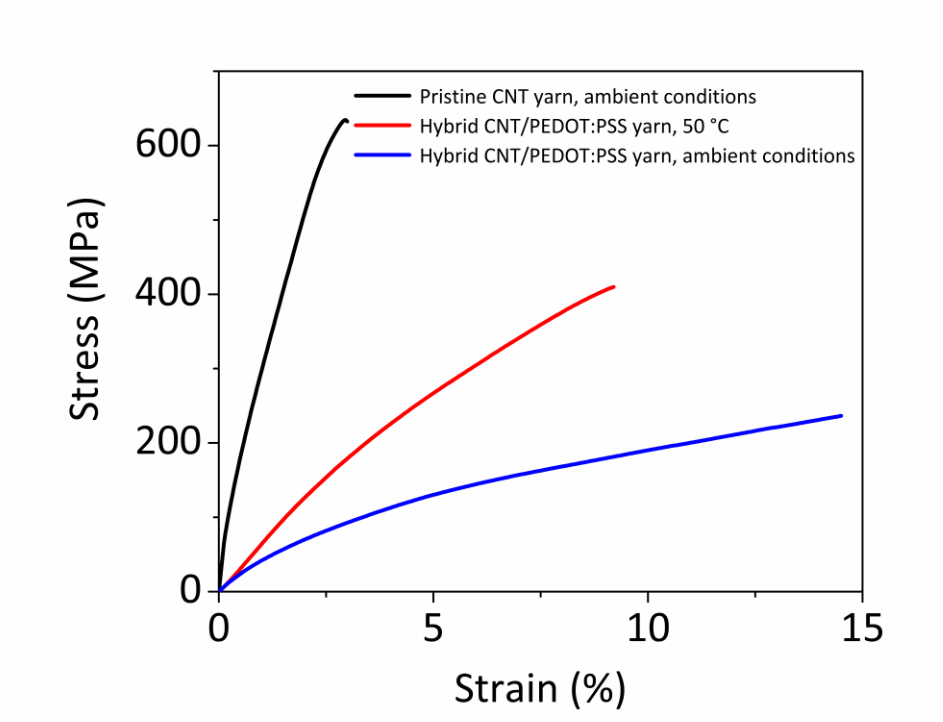

Fig. S7 Typical stress-strain curves of pristine and hybrid yarn. The tests were conducted under ambient conditions of ~ 18 % RH and 24 ± 0.3 °C except for the isothermal test at 50 °C. The decreased rigidity of the hybrid yarns is due to the occupation of mobile liquids in the actuating sites.

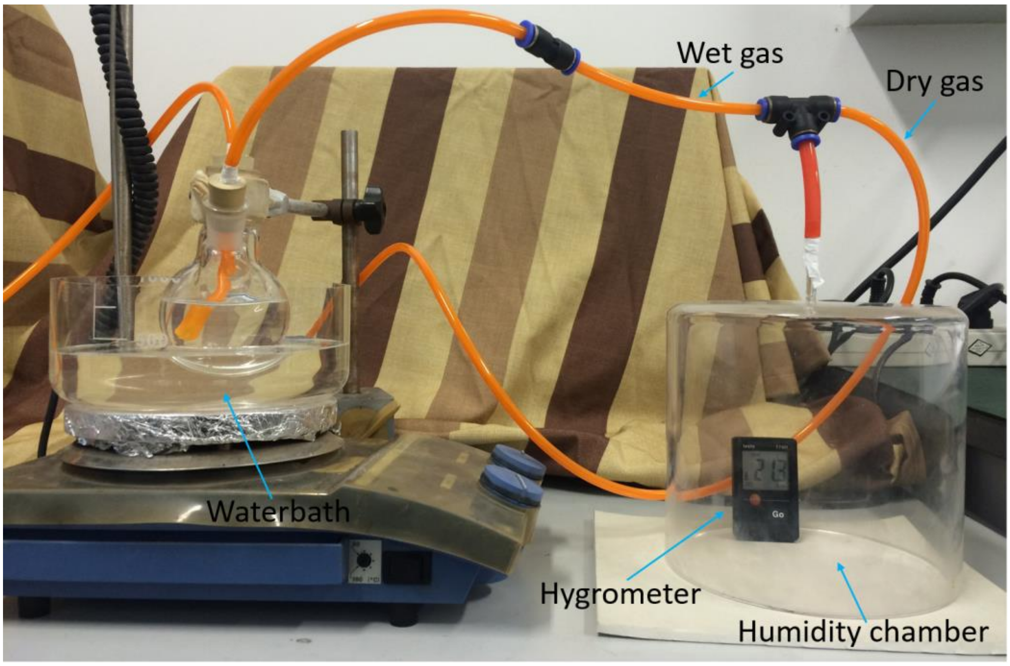

Fig. S8 Photography of a homemade humidity control system. Theoretically, the relative humidity inside the quartz chamber monitored by a hygrometer (Testo 174H) can be regulated by tuning the flow ratio of dry and wet gas (N_2) that are transported into it, and waterbath can be used to accelerate the humidity change. However, small gas flow is needed in our experiment and the corresponding rate of humidity variation was rather slow. In practice, we adopted tiny gas flow of 20 sccm to reduce perturbation and mimic a humidity changing environment as shown in Fig. S9.

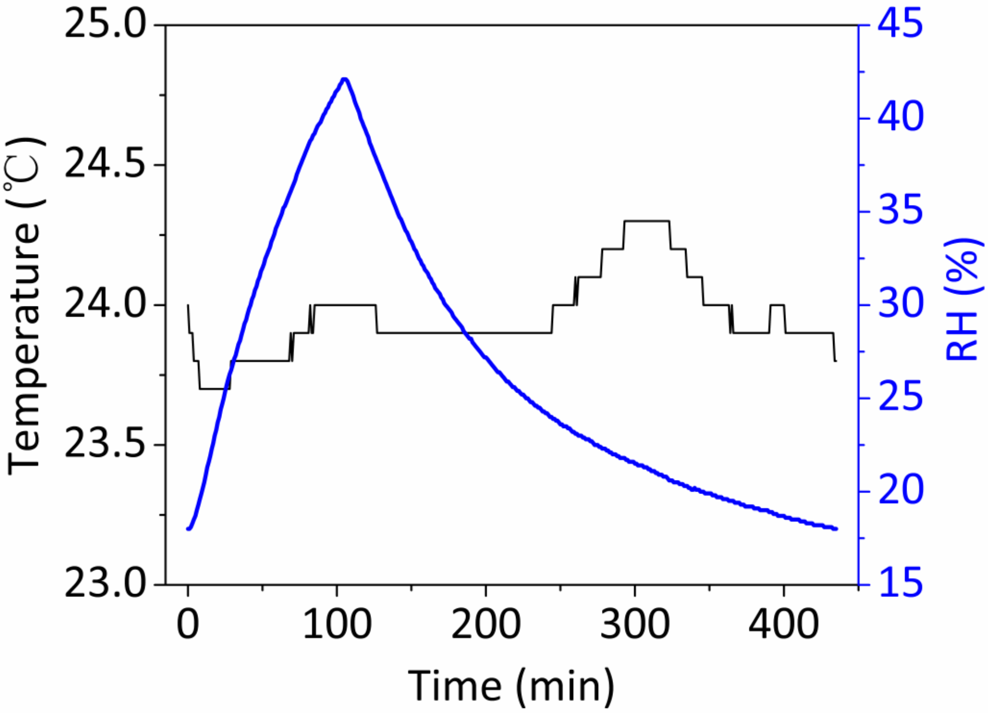

Fig. S9 Relative humidity and temperature variation in the humidity chamber. A wet gas flow of 20 sccm was adopted in the humidity increasing process and changed to dry gas with the same flow rate in the humidity decreasing process.

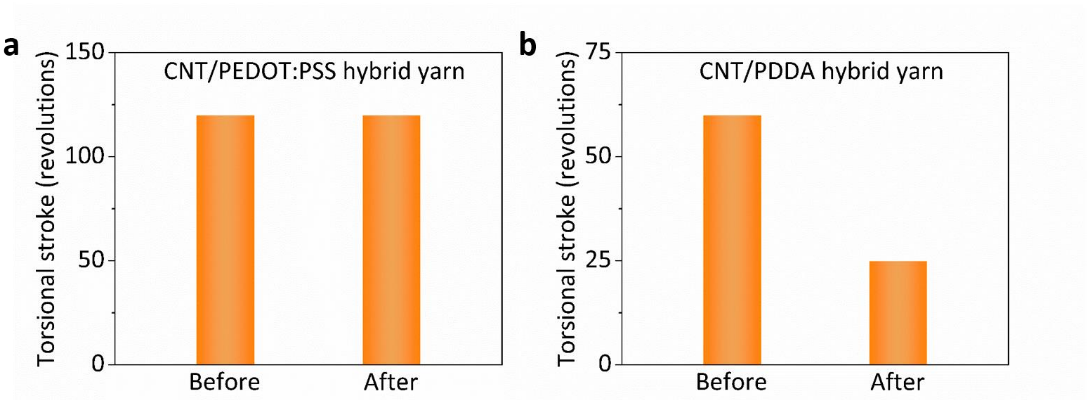

Fig. S10 Torsional stroke of CNT/PEDOT:PSS (a) and CNT/PDDA (b) hybrid yarns before and after soaked in water for 24 hours.

Table. S1 Comparisons between the as-prepared hybrid CNT/PEDOT:PSS yarn actuator and the previously reported CNT yarn-based actuators[S2-S6].

Muscles / Characters	Single-ply MWCNT yarn muscle[S2]	Single -ply hybrid CNT/paraffin wax yarn muscle[S3]	Two-ply solid-state MWCNT yarn muscle[S4]	Multi-ply MWCNT yarn muscle[S5]	Multi-ply plasma-treated MWCNT yarn muscle[S6]	This work
Stimulus	Electricity	Heating	Electricity	Ethanol solvent or vapor	Water solvent or vapor	Water solvent or vapor
Mechanism	Electrochemical intercalation of ions	heat-induced volume expansion of wax	Electrochemical intercalation of ions	Solvent infiltration	Solvent infiltration	Solvent infiltration
Torsional stroke (revolutions/m)	694▲	48	147	2,050	170.3	4,240
Peak velocity (r.p.m.)	590	11,500★	2,330	127	—	1,395
Normalized peak velocity (r.p.m. per meter)	9,833	166,667	—	6,361	—	31,000
Initial specific torque (N m/kg)	1.85	—	0.067	0.63	0.4	0.52
Maximum contractive strain (%)	—	10	1.3	15	—	68
Maximum contractive stress (MPa)	—	>80	>17.8	1 - 1.1	22.4	>39

▲ The two ends of the yarn were tethered in the measurement.

★ This value was obtained by a two-end-tethered, 6.9-cm-long hybrid yarn with a 15-Hz, 40-V/cm, square-wave voltage when the mass ratio of paddle to yarn was 16.5.

Captions for Supplementary Videos

Video S1. The CNT/PEDOT:PSS hybrid yarn before it dries is controllably twisted by a motor with a copper weight fixed at the free end. The beaker is against the copper weight to prevent it from rotating.

Video S2. The pure CNT yarn (left) and the hybrid yarn with infiltration after twist-insertion (right) show little actuation at the excitation of water.

Video S3. Two hybrid yarns with twist density of 9,092 turns/m (left) and 9,736 turns/m (right) generate fast rotation and large contraction when a water droplet is moved up and down along the lengthwise direction.

Video S4. A coiled hybrid yarn with twist density of 9,972 turns/m lifts up a copper paddle that is 11,039 times as heavy to a maximum contractive strain of ~ 9.4 % stimulated by water.

Video S5. A coiled hybrid yarn with twist density of 9,049 turns/m lifts up a copper paddle that is 1,524 times as heavy to a maximum contractive strain of ~ 30 % stimulated by water. The paddle also shows some rotation in the actuation process in spite of the impediment of the glass slide.

Video S6. A non-coiled hybrid yarn with twist density of 8,032 turns/m drives the copper paddle ($I = 1.8 \times 10^{-10}$ kg m^2) and exhibits rapid rotation activated by water. The laser displacement sensor records the actuating and recovery process of the paddle target.

Video S7. A hybrid yarn is actuated by finger-evaporated sweat or body temperature and shows rotations in opposite directions.

Video S8. A copper paddle (moment of inertia is 3.7×10^{-10} kg m^2) that connects two hybrid yarns with opposite chirality shows fast rotation once the electrical current of 10 mA is on, demonstrating actuation stimulated by Joule heat.

Video S9. A smart fabric lifting a copper ball driven by humidity or water. The mass ratio of the ball and the fabric is 977.

Supplementary References

[S1] *http://www.engineershandbook.com/Tables/frictioncoefficients.htm*

[S2] *J. Foroughi, G. M. Spinks, G. G. Wallace, J. Oh, M. E. Kozlov, S. Fang, T. Mirfakhrai, J. D. W. Madden, M. K. Shin, S. J. Kim and R. H. Baughman, Science, 2011, 334, 494-497.*

[S3] *M. D. Lima, N. Li, M. J. de Andrade, S. Fang, J. Oh, G. M. Spinks, M. E. Kozlov, C. S. Haines, D. Suh, J. Foroughi, S. J. Kim, Y. Chen, T. Ware, M. K. Shin, L. D. Machado, A. F. Fonseca, J. D. W. Madden, W. E. Voit, D. S. Galvao and R. H. Baughman, Science, 2012, 338, 928-932.*

[S4] *J. A. Lee, Y. T. Kim, G. M. Spinks, D. Suh, X. Lepro, M. D. Lima, R. H. Baughman and S. J. Kim, Nano Lett., 2014, 14, 2664-2669.*

[S5] *P. Chen, Y. Xu, S. He, X. Sun, S. Pan, J. Deng, D. Chen and H. Peng, Nat. Nanotechnol., 2015, DOI: 10.1038/nnano.2015.198.*

[S6] *S. He, P. Chen, L. Qiu, B. Wang, X. Sun, Y. Xu and H. Peng, Angew. Chem. Int. Ed., 2015, 54, 14880-14884.*