Supplementary Information

Supplementary Figures



Supplementary Figure 1. Schematic illustration of the overall scheme used for the fabrication of a hybrid carbon nanotube yarn artificial muscle. In the sequential process indicated by the arrows in the figure: A MWNT sheet was tethered at one end and attached at the opposite end to a motor. Rotating one sheet end by ~228 turns/cm collapsed a 110 mm long sheet into a 70 mm long yarn having dual Archimedean scroll structure. The yarn diameter was ~20 μ m. A mixture of paraffin wax and polystyrene-poly(ethylene-butylene)-polystyrene (SEBS) copolymer was dissolved in toluene, and infiltrated into one half of the yarn length by immersion of one-half of the yarn length in the paraffin/SEBS solution for 24 hours at 60°C. The yarn was removed from the solution and the toluene was evaporated. Then, the unfilled *S* type yarn was untwisted and retwisted to *Z* type by a motor attached at one end, thereby providing a heterochiral *S*-*Z* yarn in which only the S segment is infiltrated with the paraffin/SEBS guest.



Supplementary Figure 2. Illustration of the configuration used for characterizing torsional thermomechanical actuation. A half-infiltrated *S-Z* carbon nanotube yarn was vertically mounted in a cylindrical chamber within a thermal mechanical analyzer, using end tethers that prohibited changes in yarn length or rotation of yarn ends. A high speed camera was used for recording the torsional rotation of a paddle that was attached at yarn center.



Supplementary Figure 3. The approximately linear dependence of yarn current on yarn voltage (per yarn length) for a 4-cm-long hybrid yarn artificial muscle.



Supplementary Figure 4. The theoretical analysis of damping ratio.

Supplementary Notes

Supplementary Note 1. Theoretical Analysis of Dampening

The fundamental parameters of a single degree of freedom vibratory system are ω_n , the undamped natural frequency (or λ_n , the overdamped natural frequency), and ζ , the damping ratio. If the system is lightly damped, these parameters can be determined from the ratio of successive maxima of free motion:

$$\delta = \ln \frac{u_0}{u_1} = \frac{1}{n} \ln \frac{u_0}{u_n}$$
$$\zeta = \frac{\delta}{\sqrt{4\pi^2 + \delta^2}} \quad \text{and} \quad \omega_n = \frac{\delta}{\tau_d \sqrt{1 - \zeta^2}}$$

Then

 τ_d : the damped period of the free motion.

The equation and differential equation which define the free motion of an over–damped single degree of freedom system¹⁹ are

$$m\ddot{u} + c\dot{u} + ku = 0 \tag{1}$$

$$\ddot{u} + 2\zeta \omega_n \dot{u} + \omega_n^2 u = 0 \tag{2}$$

$$\zeta >1 \text{ (if over-damping)}$$

$$u = u(0) \qquad \dot{u} = \dot{u}(0)$$

$$u(t) = e^{-\zeta \lambda_n t} \left[u(0) \cos \lambda_d t + \frac{\dot{u}(0) + \zeta \lambda_n u(0)}{\lambda_d} \sin \lambda_d t \right] \quad (3)$$

 $\lambda_d = \lambda_n \sqrt{\zeta^2 - 1}$: natural frequency of damped vibration From the Supplementary Fig. 4, we can calculate the damping ratio. The specific initial conditions u = u(0) = 0 and $\dot{u} = \dot{u}(0) = v_0$

$$u(t) = e^{-\zeta \lambda_{n} t} \left[\frac{v_{0}}{\lambda_{d}} \sin \lambda_{d} t \right]$$

$$\lambda_{1} = \frac{1}{\Delta t} \ln \frac{u_{A}}{u_{B}}, \quad \Delta t = t_{B} - t_{A},$$

$$\zeta = \frac{T \lambda_{1}}{2\sqrt{T \lambda_{1} - 1}}$$

$$\lambda_{n} = 2\zeta / T$$
(6)

From the equations (5) and (6), we can obtain the damping ratio of 80% paraffin yarn in the value of \sim 2 which indicates the over damping. The equilibrium time requires about 0.5 s. In case of 20% paraffin yarn, the damping ratio is ~ 0.26 as under-damping.

Supplementary Note 2. Theoretical Analysis for the Scalability of Torsional Actuation

Here we demonstrate that yarn torsional actuation is highly scalable, with generated torque proportional to the cube of yarn diameter and torsional work output scaling linearly with yarn volume. We consider yarns prepared to different diameters (*D*) and twisted to the same bias angle of the nanotubes relative to the yarn direction. This bias angle depends on yarn diameter and the inserted twist per length *T*, as given in the equation $\alpha = \tan^{-1}(\pi DT)$ for nanotubes on the yarn surface. The inserted twist needed to produce a given α correspondingly decreases in proportion to the inverse of yarn diameter. We assume that the torsional stroke per yarn length (ϕ_{max}/L) is directly proportional to inserted twist. From torsion mechanics the torque (τ_{max}) generated by blocking this rotation is:

$$\tau_{\max} = \frac{\varphi_{\max} JG}{L} \tag{1}$$

where G is the yarn shear modulus and J the polar moment of inertia ($\pi d^4/32$). Combining these equations predicts that maximum generated torque scales with the cube of yarn diameter and is independent of yarn length:

$$\tau_{\max} \approx D^3 G \tan \alpha \tag{2}$$

Such muscle scaling was verified experimentally, using 67, 140, and 220 μ m diameter yarns (Fig. 4b). Each yarn was twisted to a bias angle of approximately 27°, and infiltrated with 8:2 wax:SEBS in both the S and Z segment. Force was measured from a paddle attached at the midpoint of the yarn by using a microbalance. The torque, normalized to the yarn's diameter cubed, varied only slightly for each yarn (Fig. 4b). The relatively minor differences between results for different diameter yarns can be a result of slight variation in the amount of wax:SEBS infiltrated, minor differences in the yarn bias angle, and subtle difference in yarn structure caused by the need to fabricate increasing diameter yarns from increasing numbers of stacked sheets.

Torsional work output (*W*) will be the product of generated stroke (ϕ) and torque (τ) with stroke reduced linearly in proportional to the externally applied torque from the maximum free stroke, ϕ_{max} achieved in the absence of external torque:

$$\varphi = \varphi_{\max} - \frac{\tau L}{JG} \tag{3}$$

The maximum work output is shown by differentiation to be:

$$W_{\rm max} = \frac{\varphi_{\rm max}^2 JG}{4L} = \frac{1}{4} \varphi_{\rm max} \tau_{\rm max}$$
(4)

And using the above equations:

$$W_{\rm max} \approx GLD^2 \tan^2 \alpha$$
 (5)

Consequently, for constant α , the torsional work output per yarn volume is predicted to be independent of yarn diameter, which is consistent with the experimental results of Fig. 4c.

Supplementary Reference

¹⁹ Karnopp, B. H. & Fisher, F. E. On the vibrations of overdamped systems, *Journal of the Franklin Institute*, **327**, 601–609 (1990).