## **Supplementary Information**

# Artificial Muscle with Reversible and Controllable Deformation

## Based on Stiffness-variable Carbon Nanotube Spring-like

#### Nanocomposite Yarn

Liangliang Xu<sup>1#</sup>, Qingyu Peng<sup>1,2#\*</sup>, Yue Zhu<sup>1</sup>, Xu Zhao<sup>1</sup>, Minglong Yang<sup>1</sup>, Shasha Wang<sup>1</sup>, Fuhua Xue<sup>1</sup>, Ye Yuan<sup>1</sup>, Zaishan Lin<sup>1</sup>, Fan Xu<sup>1</sup>, Xianxian Sun<sup>1,2</sup>, Jianjun Li<sup>1</sup>, Weilong Yin<sup>1,2</sup>, Yibin Li<sup>1,2\*</sup> and Xiaodong He<sup>1,2</sup>.

<sup>1</sup> National Key Laboratory of Science and Technology on Advanced Composites in Special Environments, Center for Composite Materials and Structures, Harbin Institute of Technology, Harbin 150080, P. R. China

<sup>2</sup> Shenzhen STRONG Advanced Materials Research Institute Co., Ltd., Shenzhen 518000, P. R. China

\*Corresponding authors. Email: pengqingyu@hit.edu.cn, liyibin@hit.edu.cn,

<sup>#</sup>These authors contributed equally to this work.

### **Supplementary Information:**

Figure S1. SEM image of the pre-stretched spiral CNT fiber.

Figure S2. SEM images of the surface (a) and cross-section (b) for the CNT nanocomposite yarn artificial muscle.

**Figure S3.** Morphological changes of pure CNT yarn when the voltage was applied (a) and output stress of the pure CNT yarn under different pulsed voltages vs. Time (b).

**Figure S4.** Stress nephogram of the CNT nanocomposite yarn artificial muscle simulated by finite element analysis.

**Figure S5.** The strain variation of the CNT nanocomposite yarn artificial muscle when the applied voltages increased first and then decreased.

**Figure S6.** The output stress variation of the CNT nanocomposite yarn artificial muscle when the applied voltages increased first and then decreased.

**Figure S7.** The Volt-Ampere(V-A) curve of the CNT nanocomposite yarn artificial muscle.

 Table S1 The comparison of other artificial muscles with the CNT nanocomposite

 artificial muscle we prepared.

Movie 1 The whole reversible driving process of the CNT nanocomposite yarn artificial muscle.

**Movie 2** Driving deformation of the CNT nanocomposite yarn artificial muscle observed by the optical microscope.

**Movie 3** Simulation of the driving deformation of the CNT nanocomposite yarn artificial muscle by finite element analysis.

**Movie 4** The clamped process of the clamping device based on the CNT nanocomposite yarn artificial muscle.

Movie 5 The reusability test of the clamping device.



Figure S1. The pre-stretched spiral CNT fiber.



**Figure S2.** SEM images of the surface (a) and cross-section (b) for the CNT nanocomposite yarn artificial muscle. We can see the CNTs with orientation on the nanocomposite artificial muscle surface (a), and cross-section of the spring-like nanocomposite artificial muscle was divided into two parts (b), the results indicated that epoxy resin was infiltrated into the spiral CNT yarn.



**Figure S3.** Morphological changes (a-c) and output stress under pulsed voltages vs. Time (d) for the pure CNT yarn. (a-c) There is no morphological changes for pure CNT yarn when the voltage was applied. (d) The pure CNT yarn also can output periodic stress under the pulsed voltage, but it was very small (< 0.3 MPa), which indicated that the driving deformation of the CNT nanocomposite yarn artificial muscle is realized by the resilience force of the spring.



**Figure S4.** Stress nephogram of the composite artificial muscle simulated by finite element analysis. (a) In the original state, large tensile stress in CNT yarn because the CNT yarn spring was stretched, and compressive stress in epoxy resin because epoxy resin spring was slightly compressed; (b) when the voltage was applied, the nanocomposite yarn artificial muscle was shortened. Due to the recovery of the CNT yarn spring, the stress of CNT yarn was almost lost, and the stress of epoxy resin was also nearly disappeared because its modulus decreased sharply due to the rise of temperature; (c) when the voltage was removed, the nanocomposite yarn artificial muscle was realized reverse deformation and recovered to its initial state, stress state of the CNT yarn and epoxy resin was restored.



**Figure S5.** The strain variation of the CNT nanocomposite yarn artificial muscle when the applied voltages increased first and then decreased. We can see that the strain of the nanocomposite yarn artificial muscle can be changed with the applied voltages, which proves that the artificial muscle can achieved reversible and controllable deformation.



**Figure S6.** The output stress variation of the CNT nanocomposite yarn artificial muscle when the applied voltages increased first and then decreased.



Figure S7. The Volt-Ampere(V-A) curve of the CNT nanocomposite yarn artificial muscle.

 Table S1 The comparison of other artificial muscles with the CNT nanocomposite

 artificial muscle we prepared

|                     | Strain (%)  | Response<br>time (s) | Cycle<br>number     | Inference |
|---------------------|-------------|----------------------|---------------------|-----------|
| MWCNT/wax           | 2.5 (200°C) | ~2.5                 | 1.4×10 <sup>6</sup> | [1]       |
| hybrid yarn         | 7.3 (2560℃) |                      |                     |           |
| MWCNT               | 1           | 5                    | -                   | [2]       |
| Pure CNT yarn       | 0.1         | ~1                   | -                   | [4]       |
| Nanotube-based      | ~5          | ~300                 | -                   | [29]      |
| fuel-cell muscles   |             |                      |                     |           |
| MWCNT               | <65         | -                    | 50                  | [30]      |
| Niobium NW/wax      | 0.24        | >100                 | -                   | [32]      |
| Twist spun          | 1.2         | 6                    | -                   | [33]      |
| and coiled CNT yarn |             |                      |                     |           |
| CNT nano-           | 12 (40°C)   | ~2                   | >1000               | This      |
| composite yarn      |             |                      |                     | article   |