Supplementary Information

Spirally deformable soft actuators and their designable helical actuations based on

highly oriented carbon nanotube film

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1. Detailed simulation results of finite element analysis (FEA) for the helical actuators

Fig. S1 The FEA simulation results with temperature distributions for the six spirally deformable actuators with typical helical angles, corresponding to the actuators in Fig. 4 and Fig. 5 of the manuscript.

2. General FEA process for the helical actuators

In this work, the FEA simulations were employed to simulate the movements and temperature distributions of the spirally deformable actuators with different CNT alignments. The commercial FEA software used in this work is ABAQUS. The electrode patterns and corresponding dimensions used in the FEA software are the same as the actual actuators. The relevant parameters of the CNT film and the PDMS layer are as follows:

	Thickness (µm)	Density (kg m ⁻³)	Specific Heat (J kg ⁻¹ K ⁻¹)	Poisson Ratio	Emissivity
CNT film	20	800	500	0.2	0.95
PDMS	150	1,030	1100	0.4	0.7

		Electrical Conductivity (S m ⁻¹)	Thermal conductivity (W m ⁻¹ K ⁻¹)	Young Modulus (MPa)	Thermal Expansion Coefficient (K ⁻¹)
CNT film		21,000	50	3000	3×10 ⁻⁶
		2,400	5	200	3×10 ⁻⁵
PDMS		10-8	0.18	1	3.1×10 ⁻⁴

The CNT film is highly anisotropic, so the electrical and mechanical properties are quite different along the CNT aligned direction and its vertical direction. The symbol " \parallel " in the table above represents the direction parallel to the CNT alignment, whereas " \perp " represents the direction perpendicular to the CNT alignment.

First, we used a "coupled thermal-electric" ABAQUS analysis project to simulate the temperature rise distribution of the U-shaped actuator which was heated by a certain current load. In this analysis project, we only got the temperature distributions of the actuators with different CNT alignments ($\theta' =$

60°, 45°, 30°, corresponds to Fig. 4 of the manuscript; and $\theta' = -60^\circ$, -45° , -30° , corresponds to Fig. 5 of the manuscript). All the results are saved as "**.odb" files. The coefficient of convection was set as $25 \text{ W/(m}^2 \cdot \text{K})$, the natural convection. The ambient temperature was 25 °C. When we set the properties of the materials in the ABAQUS, we also need to assign the characteristic direction of the CNT electrode, which is consistent with the CNT aligned direction of the actual actuator.

Second, we used a "Static, General" analysis project to imitate the thermal induced actuation process. The predefined field (initial temperature) in the initial step was set to 25 °C, which means at this temperature the actuator was flat. In Step-1 which is just after the initial step (25 °C), the corresponding "**.odb" file obtained in the above "coupled thermal-electric" project was transferred to the predefined field to supply the temperature information of the actuator heated by a certain power, and the software can use this temperature information to simulate the deformation process of the actuator. After these two projects, we finally got the simulated deformation form and temperature distribution for each actuator, as shown in Fig. S1.

3. The effect of aspect ratio on the deformation forms of the actuator.

How the aspect ratio affects the deformation form (helical angle and bending angle) is an important issue. First, as for the helical angle, the aspect ratio of the actuator has little to do with the helical angle, because the helical angle mainly depends on the CNT aligned direction in the electrode. In order to prove that, we conducted a series of finite element analyses (FEA) on the actuators with different aspect ratio. The following figure (Fig. S2) illustrates the deformations of three actuators with the same length of 25 mm, and with different widths (2.2 mm, 4.2 mm and 6.2 mm respectively). The CNT aligned

direction of the actuators is set -60° with their length direction. All the actuators undergo a temperature change from 25°C to 140°C, and we can see that the aspect ratios (different actuator widths) have no effect on the helical angle.



Fig. S2 The FEA simulations of the three actuators with same length of 25 mm and with different widths. (a) Width: 2.2 mm; (b) Width: 4.2 mm; (c) Width: 6.2 mm.

Second, as for the bending angle, the aspect ratio (especially the length) could influence the bending angle. When the width of the actuator is determined, the bending angle will increase with the length of the actuator (as long as aspect ratio<15), which is illustrated in Fig. 6c of the manuscript. However,

when the length of the actuator is determined, the width of the actuator has little or no effect on the bending angle, which is confirmed by the above FEA simulation.

In summary, it is not the aspect ratio but the CNT aligned direction that determines the helical angle of the actuator. And the bending angle is proportional to the length of the actuator, when the actuator is driven by a certain power density or undergoes a certain temperature change. And the width of the actuator has little effect on bending angle.