## Supplementary Material

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## **1** The customized machine for fabricating TCAs with free strokes

We design a customized machine to fabricate TCAs with free strokes as shown in Fig. s1a. The machine has two step motors A and B (coiling motors) facing each other in a horizontal line and a traveler controlled by another step motor C (guiding motor) that can travel between the two motors. The two coiling motors are used to hold a mandrel core (a cylindrical rod or wire) and apply a tension to keep the mandrel taut. The traveler is used to guide a copper wire wrapped on the mandrel core to form a helical groove. The main function of the machine is to fabricate a helical mandrel and coil a twisted fiber on the mandrel in the guiding groove. The machine can fabricate a TCA that has a maximum length of 350 mm in 8 mins except for the annealing and training time.

## 2 Fabrication of the helical mandrel

To fabricate the helical mandrel, we first tie the two ends of a rigid carbon fiber rod to the two coiling motors' shafts (Fig. s1a). A copper wire travels with the traveler at a constant speed from one end to the other. When the two motors are rotating in the same direction, the copper wire will be wrapped around the carbon rod in a helical shape to form the guiding groove. After

that, we fix the two ends of the copper wire on the carbon rod and a firm groove will be formed on the mandrel. The speed ratio between the guiding motor and the coiling motors can be tuned to fabricate a helical mandrel with different coil bias angles.

## **3** Fabrication of TCAs with free strokes

Then, we fabricate TCAs with free strokes by coiling a twisted fiber on the helical mandrel along the groove. In Fig. s1b, We twist 1 thread (Shieldex Trading, 235/36 dtex 4 ply HC+B) with an initial length of 350 mm by hanging a weight of 240 g, and the process ends after inserting N = 287 rotations before auto-coiling starts. A weight heavier than 240 g may easily break the threads and a lighter weight will not allow for enough twisting of the threads. Fig. s1c shows that the both motors rotate the helical mandrel to coil the twisted threads on it along the groove in the same direction as twisting. After that, the two ends of the twisted threads are clipped on the mandrel to prevent it from uncoiling. The actuator is then annealed in an oven (Quincy Lab 10GCE, accuracy  $0.5^{\circ}$ C) for 2.5 hours at a temperature of  $180^{\circ}$ C or  $200^{\circ}$ C. Finally, we obtain a stabilized TCA that does not untwist when the copper wire is unwrapped from the carbon fiber rod (Fig. s1 d-e).

After the TCA is removed from the mandrel core, a training process in Fig. s1f is performed to endow the TCA a reversible stroke. In the process, a weight of 2 g is hanged at the end of the TCA to keep it straight, and we apply electricity to make it fully contracted for 10 times. Finally, the TCA will have a reversible stroke (free stroke) and recover to a certain length even without any external load after cooling. The length of the TCA after training (natural length in main texts) will be shorter than the length when it is with the mandrel (called made length in main texts). We can easily fabricate TCAs with different parameters. For example, we can fabricate TCAs with different inner diameters and coiling angles by changing the diameter of the carbon fiber rod and traveling speed of the traveler. We totally fabricate 5 types of TCAs

using the same twisting fiber and their parameters are summarized in TAB. s1.

With the machine, it takes total 8 mins to fabricate the TCA: 3 mins to fabricate one helical mandrel, 3 mins to fabricate a twisting thread and another 2 minutes to coil the twisted threads on the mandrel. To improve efficiency, a long TCA could be fabricated and cut into several pieces for different usages.

## **4** Fabrication parameters of TCAs used in this work

The five TCAs (type 0, type 1, and type 2) have the following same parameters

- $d_0$ : Diameter of the twisted thread (after twisting), 0.34 mm
- $d_m$ : The diameter of the mandrel core (equals to the inner diameter of the TCA), 0.405 mm
- D: Outer Diameter of the TCA, 1.08 mm
- C: Spring index  $C = D/d_0 = 2.17$
- $l_0$ : Initial twisted thread length, 350 mm

The five TCAs (type 0, type 1, and type 2) have the following parameters with different values listed in TAB. s1). Note that the pitch angle during coiling for conventional TCA without free strokes (Type 0) cannot be changed.

 $\alpha_m$ : pitch angle during coiling process, °(Note that this angle is equal to the pitch angle of the helical groove)

 $\alpha_n$ : pitch angle corresponding to the natural Length, °

- $l_m$ : Made length, mm
- $l_n$ : Natural Length, mm
- $S_f$ : Free stroke,  $S_f = (l_n l_{min})/l_n$
- $T_a$ : Annealing temperature, °C

NO.	$\alpha_m$	$\alpha_n$	$l_m$	$l_n$	$S_f$	$T_a$
Туре 0-180	11.53	10.8	70	61	5.7%	180
Type 1-180	15.57	14.56	94	88	20.4%	180
Type 2-180	22.54	18.14	133.5	109	35.7%	180
Type 1-200	15.57	15.57	96	96	27.2%	200
Type 2-200	22.54	22.54	134	134	48%	200

TAb. s 1. Parameters of different types of TCAs fabricated in the paper

#### **5** Experiments for static response with respect to temperature

First, the TCA is placed in the oven that is used to fabricate the TCA (Fig. s2). Its top is fixed to the oven and its bottom is connected to a carbon fiber rod that comes out from the vent hole of the oven. We place a marker at the top of the carbon fiber rod and use a laser sensor (OPT2006, Wenglor sensoric GmbH) to monitor the contraction of the TCA. The weight of the carbon fiber rod with the marker is only 0.2g. In an experiment, a specific weight (one of 0 g, 10 g, 20 g, 30 g, 40 g, and 50 g) is hanged at the bottom of the TCA.

In the experiment, the temperature of the oven gradually increases to 160° from the room temperature in around 14 mins and the temperature is recorded with a thermistor (EPCOS Inc., B57540G0503F000, time constant 3s). Due to the comparable sizes of the TCA and the thermistor and the low increasing rate of the temperature, we can treat the temperature of the TCA is approximately the temperature measured by the thermistor. Before an experiment, we place the corresponding weight and conduct a heating cycle using electricity and wait 3 mins to start an experiment. This will allow the TCA to quickly creep to a length close to the steady state length corresponding to the weight, which is shown in Fig. s3. The maximum standard deviates for the 6 measurements (0g, 10g, ..., 50g) for the Type 0-180 (Fig. s4a) are respectably

0.6623, 0.3615, 1.6505, 1.6360, 0.8487, and 0.3535 mm and for the Type 1-180 (Fig. s4b) are respectably 1.8909, 1.4810, 1.6113, 1.6045, 1.6045, and 2.1412 mm

## 6 Parameter identification of TCA's displacement-temperature relationship

Based on our observations, the tailored TCAs' displacement cannot be described using a linear model, where both c and k are constant. Therefore, we approximate  $\lambda$  using a polynomial  $\lambda = a\Delta T + b$  in terms of temperature. The value of a and b can be found using a least-square fit,  $a = 0.0012 \text{ mm/K}^2$ , b = 0.140 mm/K. We can see this model can match the experiments very well before the coils becoming too close.

Since the length changes with respect to time due to creep (Fig. S3), k is dependent on the TCA's time as well as length. To avoid modeling creep, we direct measure  $l'_n$  and use it as a modeling parameter. The simulated results are plotted in Fig.4 (a) and (b) as dash-dotted lines, which also match well with experimental results.

## 7 Temporary length and aging test

#### **Temporary Length**

Our TCAs with free strokes exhibit a temporary length right after fabrication or subject to an external load due to the creep of polymer materials. After the annealing process and removing from the mandrel, it will slowly (around one day) creep to its natural length from a *temporary natural length* (slightly shorter than the made length). Such a creeping process can be sped up by several heating cycles. Therefore, we apply 10 heating cycles before we embed a TCA into a soft body when fabricating soft robots to make sure the TCA has a natural length instead of a temporary natural length.

When subject to an external load, the TCA also exhibits a temporary length. We conduct an experiment to demonstrate the temporary length. In the experiment, a Type 1-180 TCA is hanged and a motion tracking system (OptiTrack, V120:Trio) is used to track the length change of the TCA by sticking two markers at its two ends (see inset in Fig. s5a). Fig. s5a shows the TCA's length with respect to time. At the beginning, the TCA is at its natural length  $l_1 = l_n$ with no load at the end. Then a weight m = 20 g is hanged at its end, and the length becomes  $l_2$  due to the instant elastic deformation. After a heating cycle is applied, the length starts to creep to  $l_3$  after the TCA cools down for about 100 s. Also, when we remove the weight, the TCA's length (becomes  $l_4$ ) will not able to immediately recover to the natural length  $l_1$ . The creeping effect will temporally keep the TCA's length, which is a temporary length. Finally, the length recovers to  $l_1$  after we apply another heating cycle. We find  $l_1 < l_2 < l_4 < l_3$ , and the difference between  $l_2$  and  $l_3$  is not only due to instant elastic deformation but due to creep, which is accelerated by the heating process. In another experiment, we first hold a weight of 20 g with hands when the TCA is at its natural length  $l_1$ . Then, we suddenly release the weight, the length will first instantly increase to a length  $l_2$ , and finally creep to a steady-state length after tracking the length of the TCA for 20 hours as in Fig. s5b. When we remove the weight, it takes also the same amount of time to recover to the natural length.

#### Aging Test

The setup that is used for measuring the temporary length is used again in this test. We hang a weight of 20g (2 MPa) at the end of the TCA (Type 1-180) and actuate it at 0.25 Hz for 10,000 cycles. Fig. s6 provides the stroke as a function of cycle (each point averages 100 cycles). The results show that the long-time creeping of the stroke is around 2% after 10,000 cycles.

#### 8 General fabrication procedures for TCA-actuated soft modules

Instead of directly embedding the TCA in the curing process, we fabricate them by assem-

bling the TCA and the soft bodies together afterward. The detailed fabrication procedures for all the TCA-actuated soft modules are as following: 1) Fabricate soft bodies with channels. 2) Assemble TCA in the soft bodies. 3) Connect electrical leads to the TCA. 4) Reshape the soft bodies. 5) Assemble the soft bodies together.

Fabricate soft bodies with channels. We use the same elastomer (Ecoflex 30, Smooth-On Inc.) for all of our soft robots in this study. Its pre-polymer mixture is prepared in three steps: (i) mixing the two components, A and B, in a 1:1 ratio, (ii) manually stirring them for  $\sim$ 5 min, and (iii) degassing the mixture under vacuum for  $\sim$ 10 min. Then the mixture is poured into a mold with carbon fiber rods (diameter 0.9 mm) that are used to create channels as shown in Fig. s7.

Assemble TCA in channels. Based on the length of channels in a soft body, we first determine the required length for a TCA. After that, in each fabrication, we cut a TCA into the required length to make sure the fabrication to be uniform. Then the TCA is sewed into the soft body (for example, the body in the twisting manipulator is the tube) passing through the channels. To reduce the friction between the soft body and the TCA, oil (3-IN-ONE Multi-Purpose Oil) is used to lubricate the channels. Note that oil is only to hlep the assembling process and it is not necessary when TCA is working in the soft body. This assembly method allows more flexibility to arrange multiple TCAs, because we can fabricate a soft body with channels in simple geometry and arrange it to a complex shape when assembling (e.g., a helical shape).

Connect electrical leads. Two copper wires are connected to the ends of the TCA. After that, we fix the two ends of the TCA on the body with Sil-Poxy Silicone Adhesive (Smooth-On Inc.).

Reshape the body. The reason to reshape the body is to circumvent difficulties in direct fabricating a soft body that has channels in a complex shape. For example, we use pre-stretching to create a curved shape for the gripper and thus a curved U shape for the TCA, which is very

difficult to create using the conventional molding method. For the twisting manipulator, we use a wrapping method to arrange a TCA into a helical shape.

Assemble bodies together. If there are multiple bodies that are actuated separately by TCAs, we can assemble them together. For example, the soft robotic arm is an assembly of the three individual modules. Sometimes, assembling and reshaping happen at the same time, for example, wrapping a TCA on a cylinder in a helical shape.

### **9** The fabrication process of the 2D bending module: gripper

We first fabricate a soft body with channels as shown in Fig. 7. Then we use a force stand (MARK-10, ESM 303) to drag a layer of a silicone tape (LOCTITE, Go2) 120%, and stick another layer of the silicone tape that is not stretched on it. After that we sew a TCA (87 mm) into the channels of the soft body as a U shape, make electrical leads and fix the two ends of the TCA on the soft body. The body is bonded to the tape that is not stretched using Sil-Poxy Silicone Adhesive. After the adhesive cures (2 hours), we release the stretching and the whole assembly becomes a curved shape.

#### **10** The fabrication process of the twisting module

We first fabricate a soft cylindrical body (diameter 6 mm, length 18 mm) and a soft tube (inner diameter 0.9 mm and outer diameter 2.5 mm) with Ecoflex 30. Then we sew a TCA (140 mm) into the tube to make a TCA-tube assembly. After that, we make two leads and fix the two ends of the TCA on the tube. The soft cylindrical body has a channel at its center allows electrical wires running through it.

For the single-helix twisting module, we directly wrap the TCA-tube assembly on the cylindrical body. For the double-helix twisting module, we first fold the TCA-tube assembly and then wrap it on the cylindrical body. After that, we fix the two ends of the TCA-tube assembly on the body with Sil-Poxy Silicone Adhesive.

## **11** The fabrication process of the **3D** bending module

We first fabricate a soft body with 3 parallel channels along its perimeter (an additional channel at its center allows electrical wires running through it), sew 3 TCAs (45 mm for each) of the same length into the channels in three directions. Then we make two leads on each TCA and fix the ends of the TCAs on the body. After that, we fixed the manipulator on a 3D printed solid base.

# 12 Experiments for characterization of the 2D bending module: gripper

To obtain the relationship between the opening width and the input power, we first calculate the required current corresponding to a specific power by measure the resistance of the TCA (assume the resistance is constant). A DC regulated power supply (Tekpowe, TP 3005T) is used to supply the required current. We record a video of the opening process for 2 s when a current is applied. Tracker software (https://physlets.org/tracker/) is used to process the videos and extract the opening width.

To find the relationship between the gripping force and the opening width, we use a force stand (MARK-10, ESM303 with M5-2 Force gauge) to drag the two ends of the gripper using two strings. The force stand is equipped with an encoder, so that it allows us to export the opening width and the force simultaneously. The experiments are repeated 3 times.

## 13 Experiments for characterization of the twisting module

First, the module is bond to a rigid base, and a carbon fiber rod is stuck to the top of the manipulator, and the power supply (Tekpowe, TP 3005T) is used to supply the required current corresponding to a specific power. The rotation processes are recorded and the rotational angle is extracted using Tracker software.

#### 14 Experiments for characterization of the 3D bending module

We place two markers on the end of the module and the top of the rigid base, respectively, as shown in Fig. s8. Marker 1 is used to calculate the origin of the module's frame, which is the connection point of the module and the rigid base (Marked as O). Marker 2 is stuck at the end of the module. The radius of the marker is 3 mm and the weight is 0.24 g.

#### 15 Fabrication and Control of the soft robotic arm

The gripper, the twisting module and the 3D bending module are connected in serial using Sil-Poxy Silicone Adhesive. All the common ground wires are connected together as one to reduce the number of wires.

We use five motor drivers (MC33926 Motor Driver Carrier) to apply electricity to five TCAs in the manipulators (1 in the gripper, 1 in the twisting module and 3 in the 3D bending module). An Arduino board is used to control the motor drivers.

## **16** Supplementary Videos

- 1. s1 Overcome Friction.mp4
- 2. s2 Overcome Gravity.mp4

- 3. s3 Gripper Characterization.mp4
- 4. s4 Twisting Module.mp4
- 5. s5 3D bending Module.mp4
- 6. s6 Soft Robotic Arm.mp4



Fig. s 1. The fabrication process of TCAs with free strokes. (a) Make the helical mandrel on the machine. (b) Twist a thread. (c) Coil the twisted thread in the groove on the helical mandrel using the machine. (d) Anneal the TCA with the mandrel. (e) Remove the TCA from the mandrel. (f) Train the TCA to get a reversible free stroke.



Fig. s 2. The experimental setup to obtain the displacement-temperature relationship in the oven. Note that only a small part of the oven is shown in this figure.



Fig. s 3. The steady-state length of the two types of TCAs corresponding to different weights after the creeping process.



Fig. s 4. The experimental results of a TCA with free strokes. Length with respect to temperature. Solid lines indicate the average and shaded regions indicate the standard deviation. (a) and (b) are corresponding to the Fig. 4 b and c in the main text.



Fig. s 5. Temporary length (a) An experiment demonstrating the temporary length with a weight hanging at a TCA's end. (b) A creeping process that lasts for 20 hours.)



Fig. s 6. Tensile stroke versus actuation cycle for a Type 1-180 TCA that is driven electrothermally at 0.25 Hz under a 2-MPa load (each point averages 100 cycles). The inset provides creep (decrease of the stroke) as a function of cycle.



Fig. s 7. The fabrication process of the soft modules with an example.



Fig. s 8. Tracking the end position of the 3D bending module.