

Supporting Information for

Multiple configuration transitions of soft actuators under single external stimulus

Yanlin Cao,^{a,b} Xianke Feng,^c Shuang Wang,^{*d} Qi Li,^a Xiyong Li,^a Hongyuan Li,^{a,b} Wei Hong,^c Huiling Duan^{a,b} and Pengyu Lv^{*a}

^a State Key Laboratory for Turbulence and Complex Systems, Department of Mechanics and Engineering Science, BIC-ESAT, College of Engineering, Peking University, Beijing 100871, China.

^b CAPT, HEDPS and IFSA Collaborative Innovation Center of MoE, Peking University, Beijing 100871, China

^c Department of Mechanics and Aerospace Engineering, Southern University of Science and Technology, Shenzhen, Guangdong 518055, China.

^d School of Physical and Mathematical Sciences, Nanjing Tech University, Nanjing, 211816, China.

*Corresponding authors: lvpy@pku.edu.cn, shuangwang@njtech.edu.cn

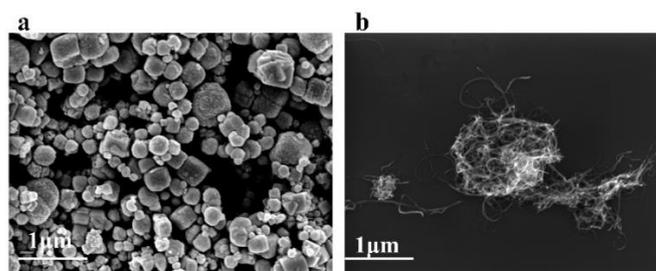


Fig. S1 SEM images of Fe₃O₄ nanoparticles (a) and CNT (b).

The ink is composed of CNT, Fe₃O₄, PDMS base agent and curing agent. The CNT herein served as a rheological tailoring agent for the ink while Fe₃O₄ provided magnetic navigation for the actuator. The microscopic morphologies of Fe₃O₄ and CNT are shown in Fig.S1.



Fig. S2 The PDMS film made between two PVC boards showed pure swelling without bending in ethyl acetate.

To figure out the reason for the PDMS film bending, the PDMS mixture was spin-coated onto the PVC sheet and then covered with another PVC sheet to ensure that both sides of the PDMS film are cured under the same conditions. As shown in Fig. S2, the prepared film between two identical PVC sheets showed pure swelling without bending in ethyl acetate.

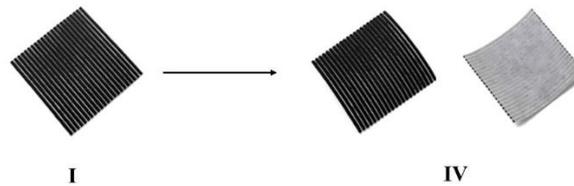


Fig. S3 Morphing process of the bilayer actuator with copy paper as the substrate film.

In order to explore the effect of substrate film modulus on configuration II, the strips were printed directly on copy paper to obtain the actuator. And this actuator in ethyl acetate morphed from the initial configuration I to configuration IV, without going through configuration II.

Finite element simulation

The basic thermodynamic algorithm is used to simulate the deformation process based on Flory-Huggins model.¹ Assume that the elastic deformation of the polymer lattice and the long-range transport of solvent molecules dominate at constant temperature and pressure, and thus our internal variables of interest are the deformation gradient F_{ij} and the solvent concentration C .

Adopting Lagrangian description, under no mechanical load, the polymer before putting into solution was taken as the reference configuration, and X was used to describe the positions of particles in the reference configuration. At time t , the polymer deforms, and the point marked X moves to $X(X, t)$, then the deformation gradient

can be expressed as:

$$F_{iJ} = \frac{\partial x_i(\mathbf{X}, t)}{\partial X_J} \quad (1)$$

Suppose the work done by the weight on the volume element is $B(X, t)dV(X)$, and the work done on the surface element is $T(X, t)dA(X)$. Define the nominal stress $s_{ij}(X, t)$ such that

$$\frac{\partial s_{ij}}{\partial X_j} + B_i = 0, \text{ in } \Omega \quad (2)$$

$$s_{ij}N_j = T_i, \text{ on } \partial\Omega \quad (3)$$

where N_j is the unit vector perpendicular to the interface between substances, marked with $+$ and $-$, and the direction pointing to the polymer is $+$.

Suppose that in the current state, the number of small molecules injected per volume element per unit time is $r_e^a(X, t)dV(X)$, and the number of small molecules injected on a plane element is $i_l^a(X, t)dA(X)$. At the same time, small molecules migrate through the polymer—the number of small molecules per unit time passing through a volume element is $C^a A(X, t)dV(X)$, and J_l^a is the diffusion flux. Assume that no chemical reaction takes place, it can be obtained from the conservation of the number of small molecules:

$$\frac{\partial C^a(\mathbf{X}, t)}{\partial t} = -\frac{\partial J_l^a(\mathbf{X}, t)}{\partial X_l} + r_e^a, \text{ in } \Omega \quad (4)$$

$$J_l^a(\mathbf{X}, t) = i_l^a \text{ on } \partial\Omega \quad (5)$$

The nominal flux is related to the chemical potential gradient of the corresponding species by a linear kinetic relation:¹

$$J_l^a(\mathbf{X}, t) = -\frac{1}{\det\mathbf{F}} \frac{C^a(\mathbf{X}, t)D^a}{kT} H_{Ji}H_{Li} \frac{\partial \mu^a(\mathbf{X}, t)}{\partial X_L} \quad (6)$$

where kT is the temperature in the unit of energy, H is the inverse of the deformation

gradient, and $\mu^a(\mathbf{X}, t)$ is the chemical potential.

The free energy of polymer comes from two molecular processes: stretching the network of the polymers and mixing the polymers and the small molecules. Following Flory and Rehner,^{2,3} we assume that the free energy of the polymer takes the form:

$$W(F, C) = W_s(F) + W_m(C) \quad (7)$$

with pre-swelling:

$$W_s(\mathbf{F}) = \frac{1}{2} NkT \left(\lambda_0^2 F_{ij} F_{ij} - 3 - 2 \ln(\lambda_0^3 \det \mathbf{F}) \right) \quad (8)$$

$$W_m(\mathbf{F}) = -kTC^a \log \left(1 + \frac{1}{\Omega C^a} \right) - \frac{kT}{\Omega} \frac{\chi}{1 + \Omega C^a} \quad (9)$$

$$\Omega C^a = \lambda_0^3 \det \mathbf{F} - 1, \lambda_0 = (1 + \Omega C_0)^{\frac{1}{3}} \quad (10)$$

where Ω is the molecule volume and χ is the dimensionless parameter.

Taking the partial derivatives of the free energy function and applying the constraint Eq. (10) by using a Lagrange multiplier Π , we arrive at the equations of state:

$$\mu^a = \frac{\partial W(\mathbf{X}, t)}{\partial C^a} + \Pi \Omega \quad (11)$$

Substituting the Eq. (11) into Eq. (6) and further into Eq. (4) and Eq. (5), together with the constraint (10) and proper initial and boundary conditions, the fields of displacement $\mathbf{u} = (\mathbf{X}, t)$, solvent concentrations $C(\alpha)(\mathbf{X}, t)$, and Lagrange multiplier $\Pi(\mathbf{X}, t)$ can be obtained. We write them into equivalent weak forms and solve them numerically in the commercial finite element package COMSOL MULTIPHYSICS 5.5, with parameters $C_0 = 0.1$, $\chi_1 = 0.64$ for the PDMS film part, and $\chi_2 = 0.58$ for the stripe part.

References

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