# ELECTRONIC SUPPLEMENTARY INFORMATION

# Hybrid artificial muscle: enhanced actuation and load-bearing performances via origami metamaterial endoskeleton

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**Fig. S1. Fabrication and performance characterization of SWCNT electrode.** (A) Fabrication processes of SWCNT electrodes. (B) Sheet conductivity as a function of the volume of the SWCNT dispersion used.



Fig. S2. Experimental setup for strain measurement.



Fig. S3. Experimental setup for and blocked force measurement.





Fig. S4. Geometry of a Kresling origami unit at initial state. (A) 3D view. (B) Top view.

A single Kresling structure is analyzed, the relationship between reaction force and height of the Kresling is determined based on the energy method and then the model is extended to multiple Kresling structures, such as KDOM.

#### S1.1 Geometry

The Kresling structures (Fig. S4) are generated by folding flat sheet patterns with parameters of  $L_p=4.90$  mm,  $L_v=9.74$  mm,  $\theta=53.78^{\circ}$  (Fig. 3a). The edges number of top and bottom polygons of Kresling is p=6. And in the initial free state, the length of mountain creases  $L_m$ , polygon radius  $R_0$ , initial height  $h_0$  and initial twist angle  $\gamma_0$ , can be obtained from the relation: <sup>[1]</sup>

$$L_{p} = 2R_{0}\sin\left(\frac{\pi}{p}\right)$$

$$L_{m} = \sqrt{h_{0}^{2} + 4R_{0}^{2}\sin^{2}\left(\frac{\gamma_{0}}{2}\right)} = \sqrt{L_{p}^{2} + L_{v}^{2} - 2L_{p}L_{v}\cos\theta} \#(1)$$

$$L_{v} = \sqrt{h_{0}^{2} + 4R_{0}^{2}\sin^{2}\left(\frac{\gamma_{0}}{2} + \frac{\pi}{p}\right)}$$

To express the state of Kresling during compression, a coordinate system is established with  $A_0$  as the origin and  $e_1, e_2, e_3$  as the base vectors. Assuming that during the compression the bottom surface is fixed and the polygon radius  $R = R_0$  does not change, the state of Kresling structure is determined by the twist angle  $\gamma$  and the height *h*.

The positions of  $A_i$  and  $B_i$  can be represented by

$$r_{A_{i}} = R_{0} \cos\left(\frac{2\pi}{p}i - \frac{2\pi}{p}\right)e_{1} + R_{0} \sin\left(\frac{2\pi}{p}i - \frac{2\pi}{p}\right)e_{2} \quad i = 1 \sim p\#(2)$$

$$r_{B_{i}} = R_{0} \cos\left(\frac{2\pi}{p}i - \frac{2\pi}{p} + \gamma\right)e_{1} + R_{0} \sin\left(\frac{2\pi}{p}i - \frac{2\pi}{p} + \gamma\right)e_{2} + he_{3} \quad i = 1 \sim p.\#(3)$$

The vectors of mountain creases  $r_{B_1A_1}$  and valley creases  $r_{B_2A_1}$ , can be written as

$$r_{B_1A_1} = r_{A_1} - r_{B_1} \# (4)$$
$$r_{B_2A_1} = r_{A_1} - r_{B_2} \# (5)$$

And the length of mountain creases and valley creases at arbitrary state can be written as

$$l_m = |r_{B_2A_1}| #(6)$$

$$l_v = |r_{B_2A_1}| \#(7)$$

 $n_1, n_2, n_3$  is the normal vector of plane  $A_1B_2A_2$ , plane  $A_2B_2B_3$ , plane  $A_2A_3B_3$  respectively, and can be written as

$$n_{1} = r_{B_{2}A_{1}} \times r_{B_{2}A_{2}} \#(8)$$

$$n_{2} = r_{A_{2}B_{2}} \times r_{A_{2}B_{3}} \#(9)$$

$$n_{3} = r_{B_{3}A_{2}} \times r_{B_{3}A_{3}} \#(10)$$

Dihedral angles at the mountain crease  $\theta_m$ , valley crease  $\theta_v$  and bottom and top crease  $\theta_p$  of Kresling origami can be expressed as

$$\theta_m = \angle (n_1, n_2) \# (11)$$
  
 $\theta_v = \angle (n_2, n_3) \# (12)$   
 $\theta_p = \angle (n_1, e_3) \# (13)$ 

# S1.2 Mechanical model

The elastic potential energy of the Kresling origami structure consists of two parts: one is the elastic potential energy stored by the panel bending. The second is the elastic potential energy

stored by the elastic hinges at the creases, including side valley creases, top and bottom creases.<sup>[2]</sup>

In order to simplify the calculation of panel deformation, we use the truss model to capture the energy of panel bending. The spring stiffnesses at the mountain folds and valley folds are respectively  $K_m$  and  $K_v$ . The values of them are determined by fitting the experimental results. The elastic potential energy stored in the panel deformation can be written as

$$E_b = \frac{p}{2}K_m(l_m - l_{m0})^2 + \frac{p}{2}K_v(l_v - l_{v0})^2 \#(14)$$

where  $l_{m0}$  and  $l_{\nu0}$  are the length of mountain and valley creases in the initial free state respectively. The creases are modeled as rotational springs. The spring stiffness  $K_c$  is

$$K_c = \frac{EI_c}{h_c} \#(15)$$

where  $h_c$  is length of the flexural pivot, and  $I_c = \frac{klb^3}{12}$  is the area moment of the cross section of the crease to the midline, where k is the crease ratio, l is the length of the creases, b is the thickness of the film, E is the Young's modulus of the material. At different creases,  $K_c$  is also different due to the difference of l and k. The spring stiffness of the valley creases is  $K_{cv} = \frac{EI_v}{b}$ ,

and the spring stiffness of the bottom and top creases is  $K_{cp} = \frac{EI_p}{b}$ . The elastic potential energy stored in the creases can be written as

$$E_{c} = \frac{1}{2}pK_{cv}(\theta_{v} - \theta_{v0})^{2} + \frac{1}{2} \times 2pK_{cp}(\theta_{p} - \theta_{p0})^{2} \# (16)$$

where  $\theta_{v0}$  and  $\theta_{p0}$  are the dihedral angles of valley creases and top and bottom crease in the initial free state, respectively.

The total elastic potential energy of KDOM with n = 2 Kresling is  $G_K = n(E_b + E_c) \#(17)$   $G_K$  depends on the height *h* and the rotation angle  $\gamma$ , where  $h = h_0 - \Delta L/n$ , and  $\Delta L$  is the amount of compression. Since the KDOM the rotation is free, the rotation angle  $\gamma$  can be obtained as following when the compression amount  $\Delta L$  is given:

$$\frac{\partial G_K}{\partial \gamma}\Big|_{\Delta L} = 0 \cdot \frac{\partial^2 G_K}{\partial \gamma^2}\Big|_{\Delta L} > 0\#(18)$$

The total reaction force F and contribution of panel bending  $F_b$  and creases  $F_c$  are:

$$F = \frac{\partial G_K}{\partial \Delta L} \Big|_{\Delta L, \gamma}$$
$$F_b = \frac{\partial E_b}{\partial \Delta L} \Big|_{\Delta L, \gamma}$$
$$F_c = \frac{\partial E_c}{\partial \Delta L} \Big|_{\Delta L, \gamma} \# (19)$$

Note S2. Analytical model for OHAM



Fig. S5. Four configurations of the OHAM.

The OHAM consists of an external DEA and an internal KDOM. KDOM contains n = 2Kresling origamis. The OHAM has four key configurations (Fig. S5). In the initial configuration, both Kreslings and DEA are in the free state. The height of KDOM is  $nh_0$ , and the initial height of DEA is  $L_0$ . In the reference configuration, the KDOM is compressed by  $\Delta L$ to  $L_0$ , with the same height of the DEA. Thus, we obtain the relation that  $nh_R = L_R = L_0$ . In the intermediate configuration, the DEA and KDOM are connected together to get OHAM, and then the OHAM is released and reached an equilibrium state. The KDOM and DEA maintain the same height, that is  $nh_I = L_I$ . Finally, in the current configuration OHAM is activated by external stimuli and undergoes deformation.

Here, it is assumed that the steady state of OHAM in the current configuration does not change regardless of whether the voltage is applied in the reference configuration or in the intermediate configuration. Then it can be considered that the voltage has been applied in the reference configuration, so that only the relationship between reference configuration and the relationship should be considered. The intermediate configuration is the special state of the actual configuration when the voltage is 0.

The free energy of the OHAM is composed of the elastic potential energy of the DEA, the energy stored in the electric field and the energy stored in the KDOM. Next, we will discuss them separately.

S2.1 Elastic potential energy of the DEA



Fig. S6. Schematic figure for the reference and current configurations of a rolled DEA.

As shown in Fig. S6, subjected the reaction force of KDOM and direct-current voltage, the rolled DEA undergoes extensions and inflation deformation and transformed from the reference configuration (R,  $\Theta$ , Z) to the current configuration (r, $\theta$ , z). Ignoring the non-uniform deformation caused by the top and bottom caps, and no torsion occurs, assuming that the DEA remains cylindrical, and the extension and inflation are uniform, the deformation law can be written as

$$r = r(R)$$
  

$$\theta = \Theta$$
  

$$z = z(Z) = \lambda_z Z \# (20)$$

where  $\lambda_z = \frac{L_C}{L_R}$  is the longitudinal stretch.

The deformation gradient reads

$$F = \begin{bmatrix} \frac{dr}{dR} & 0 & 0\\ 0 & \frac{r\partial\theta}{R\partial\Theta} & 0\\ 0 & 0 & \frac{dz}{dZ} \end{bmatrix} = \begin{bmatrix} \frac{dr}{dR} & 0 & 0\\ 0 & \frac{r}{R} & 0\\ 0 & 0 & \lambda_z \end{bmatrix} \# (21)$$

The left Cauchy-Green deformation tensor can be express by

$$B = FF^{T} = \begin{bmatrix} \left(\frac{dr}{dR}\right)^{2} & 0 & 0\\ 0 & \left(\frac{r}{R}\right)^{2} & 0\\ 0 & 0 & \left(\lambda_{z}\right)^{2} \end{bmatrix} \#(22)$$

Its three invariants  $I_1$ ,  $I_2$ ,  $I_3$  are

$$I_1 = \left(\frac{dr}{dR}\right)^2 + \frac{r^2}{R^2} + \lambda_z^2$$
$$I_2 = \frac{\left(\lambda_z^2 R^2 + r^2\right) \left(\frac{dr}{dR}\right)^2 + r^2 \lambda_z^2}{R^2}$$
$$I_3 = \left(\frac{r \, dr}{R \, R}\right)^2 \lambda_z^2 \# (23)$$

By the incompressibility condition, the following relation is satisfied:

$$I_3 = \left(\frac{r\,dr}{R\,R}\right)^2 \lambda_z^2 = 1\#(24)$$

Thus:

$$\frac{rdr}{RR} = \frac{1}{\lambda_z} \#(25)$$

Suppose the inner wall of the rolled DEA is deformed from  $R_1$  to  $r_1$ , and the outer wall is deformed from  $R_2$  to  $r_2$ , and we have

$$r^{2} = \frac{1}{\lambda_{z}} \left( R^{2} - R_{1}^{2} \right) + r_{1}^{2} = \frac{1}{\lambda_{z}} \left( R^{2} - R_{2}^{2} \right) + r_{2}^{2} \# (26)$$

The energy per unit volume of the elastomer is determined using the incompressible Gent model:<sup>[3]</sup>

$$w_{S} = -\frac{\mu J_{m}}{2} \ln \left( 1 - \frac{I_{1} - 3}{J_{m}} \right) \#(27)$$

where  $\mu$  is the shear modulus of the elastomer, and  $J_m$  describes the strain stiffening limit.

Both  $\mu$  and  $J_m$  are fitted from uniaxial tensile experimental result (Fig. S7).



**Fig. S7.** Uniaxial tensile test curves of DEA with electrodes and DEA without electrodes, and the result of Gent model.

In the current configuration, the total elastic energy in volume V can be expressed as

$$G_{S} = \int_{V} w_{s}(r) dV = 2\pi L_{C} \int_{r_{1}}^{r_{2}} r w_{S}(r) dr \# (28)$$

For the M-layer elastomer,  $r_m$  represents the inner diameter of the m<sub>th</sub> layer, and  $r_{m+1}$  represents the outer diameter of the m<sub>th</sub> layer. The total energy of the elastomer is

$$G_{S} = \int_{V} w_{s}(r) dV = 2\pi L_{C} \int_{r_{1}}^{r_{m}} t^{1} r w_{s}(r) dr \# (29)$$

#### S2.2 Energy stored in the electric field

In the current configuration, the voltage  $\Phi$  is applied, and the electric displacement  $D_c$  is

$$D_c = \begin{bmatrix} \frac{\Phi \varepsilon}{rln\left(\frac{r_2}{r_1}\right)} \\ 0 \\ 0 \end{bmatrix} \# (30)$$

in which  $\varepsilon$  is the dielectric constant of the material.

The stored electrostatic energy in the elastomer dielectric per unit volume is

$$w_E = -\frac{|D_c|^2}{2\varepsilon} \#(31)$$

The total energy of electrostatic potential energy can be written as

$$G_{E} = \int_{V} w_{E}(r) dV = 2\pi z \int_{r_{1}}^{r_{2}} r w_{E}(r) dr = -\frac{\pi \varepsilon L_{c} \Phi^{2}}{\ln\left(\frac{r_{2}}{r_{1}}\right)} \#(32)$$

For a structure composed of M layers of elastomers, the total electrical energy can be expressed

as

$$G_e = \sum_{m=1}^{M} - \frac{\pi \varepsilon L_c \Phi^2}{\ln\left(\frac{r_{m+1}}{r_m}\right)} \#(33)$$

### S2.3 Energy stored in the KDOM

For the convenience of calculation, the force-displacement curve of KDOM is fitted as a polynomial function, F = F(h). The height of KDOM is  $L_R$  in the reference configuration and  $L_c$  in the current configuration, and the potential energy change of KDOM is

$$G_K = -\int_{L_R}^{L_C} F(h)dh\#(34)$$

#### S2.4 Determination of OHAM's State

The total Helmholtz free energy of OHAM can be written as

$$G = G_S + G_E + G_K.\#(35)$$

G depends on three variables, namely the voltage  $\Phi$ , the longitudinal stretch  $\lambda_z$  and the inner

diameter  $r_1$ . Define the dimensionless quantity hoop stretch as  $\lambda_R = \frac{r_1}{R_1}$ . And for rolled DEA, we suppose  $\lambda_R = 1$ . When the voltage  $\Phi$  is given ( $\Phi = 0$  for the intermediate configuration), the equilibrium is where the actuation results in minimum total energy:

$$\frac{\partial G}{\partial \lambda_z}\Big|_h = \frac{\partial U}{\partial \lambda_R}\Big|_h = 0, \ \frac{\partial^2 G}{\partial \lambda_z^2}\Big|_h > 0, \ \frac{\partial^2 G}{\partial \lambda_R^2}\Big|_h > 0.\#(36)$$

And for rolled DEA, we suppose  $\lambda_R = 1$ . We get the equilibrium using:

$$\frac{\partial G}{\partial \lambda_z}\Big|_h = 0, \ \frac{\partial^2 G}{\partial \lambda_z^2}\Big|_h > 0\#(36)$$



Fig. S8 Stroke of OHAM-QZ5 during cycling test at 1200 V and 125 Hz which is the resonance frequency of a new specimen.

# **Supporting References**

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