Supporting Information

Superfast-Response and Ultrahigh-Power-Density Electromechanical Actuators Based on Hierarchal Carbon Nanotube Electrodes and Chitosan

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S1. Experimental details

Preparation of the electrolyte layer. As received chitosan powder (Sinopharm Chemical Reagent Co., Ltd., 250 mg) was added to 2 % acetic acid solution (10 mL) and the mixture was stirred at 60 °C for 30 min to obtain a homogenous solution. Then as received ionic liquid $EMIBF_4$ (Alfa Aesar Inc., 600 ul) and glycerol (1.25 ml) was added to the above chitosan solution and stirred at 60 °C for another 30 min. Subsequently, a portion (1.2-1.8 ml) of the resulting solution was cast in a plastic mold (25 × 25 mm).

Assembly of the bimorph configured actuators. After standing at room temperature for about 6-8 hours, the half-dried electrolyte layer membrane was carefully peeled off and sandwiched with two pieces of as-grown freestanding SWNT films as electrode layers via a facile and effective hot-press process (70 °C, 150 N for 2 min). Subsequently, the final actuator was aged at room temperature for one day, dried under a reduced pressure at 60 °C for one day, then cut into strips with the same dimensions (20 × 2 mm) for further characterizations. The thicknesses of the actuator strips are in the range of 40-60 μm, which were pre-determined by the high-resolution SEM images of their cross sections.

Characterization. SEM was carried out using a Hitachi S5200 SEM system. Mechanical tests were performed in a TA Instruments Dynamic Mechanical Analyzer Q800 on specimens with the same gauge dimension (10 × 2 mm) and strain rate (1 %/min). Electrical conductivities were evaluated using a Keithley 4200-SCS under ambient condition. Bending displacements were measured using a Keyence LK-G 80 laser displacement sensor under an AC electrical field supplied by a function signal generator. The free length (vertical distance from the fixed end) of the recorded point was 7-17 mm.

Calculation. The strain ε and stress σ generated in the electrode layers as well as the corresponding work density E_{sp} and power density P_{sp} were estimated by the following equations[1,2]:

$$\varepsilon = 2t\delta / (l^2 + \delta^2); \quad \sigma = Y\varepsilon; \quad E_{sp} = Y_\rho \varepsilon^2 / 2; \quad P_{sp} = 2fY_\rho \varepsilon^2$$

Where t, δ, l are the thickness, displacement, free length of the actuator, Y_ρ is the specific modulus of the electrode layers, and f is the driving frequency of the applied potential.

S2. Displacement-voltage curve under triangle wave potential

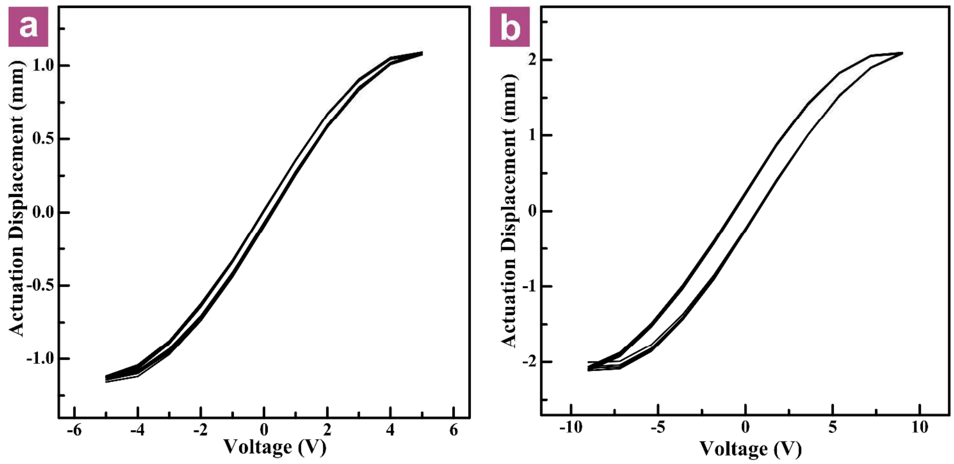

Figure S1. Displacement loops under 1 Hz triangle wave potentials of ±5 V (a) and ±9 V (b).

When triangle wave voltages of 1 Hz are applied to the actuator, the displacement-voltage curves show that the displacement is proportional to the voltage and the displacement loops exhibit little hysteresis even at higher voltages (9 V), indicating the fast response of these devices.

S3. **Derivation of the equation (1) - electromechanical damping (EMD) model**

The electromechanical actuation displacement of the actuator under square wave stimulation can be modeled by corresponding electrochemical charging and discharging processes, thus the displacement dependence on time can be described as follows[3]:

$$\delta_1(t) \propto Q(t) = \delta_0(1 - e^{-\frac{t}{\tau}}) \tag{S1}$$

Where δ_0 represents the displacement at infinite time, while τ represents the electromechanical actuation response time constant of the actuator. A smaller τ means a faster electromechanical bending response, implying faster rates of charge injection and ion migration.

The sinusoidal damping vibration excited by the instant actuating force arising from the fast charge injection and ion migration can be described with the following function[4]:

$$\delta_2(t) = A_0 e^{-\beta t} \sin[2\pi f_d (t + t_c)] \tag{S2}$$

Where A_0, t_c, β and f_d are the initial amplitude, initial phase, exponential decay rate and the damped natural frequency of the cantilever-type actuator, respectively.

Therefore, the whole bending process (up and down) can be described by combining the time dependent charging (discharging) model with the sinusoidal damping model. Here we name it Electromechanical Damping (EMD) model, in which the time dependent displacement can be expressed by following function:

$$\delta(t) = \delta_1(t) + \delta_2(t) = \delta_0(1 - e^{-\frac{t}{\tau}}) + A_0 e^{-\beta t} \sin[2\pi f_d (t + t_c)] \tag{1}$$

Where the meanings of the parameters are the same with that in separate models.

With the fitting damped natural frequency and exponential decay rate, the damping ratio ξ of this underdamped system can be given by:

$$\xi = \frac{\beta}{\sqrt{(2\pi f_d)^2 + \beta^2}} \tag{S3}$$

Additionally, the undamped natural frequency f_0 of the cantilever actuator can be calculated by:

$$f_0 = \sqrt{f_d^2 + (\frac{\beta}{2\pi})^2} \quad \text{or} \quad f_0 = \frac{f_d}{\sqrt{1-\xi^2}} \tag{S4}$$

S4. Fast response analyses for 2 and 5 Hz with the EMD model

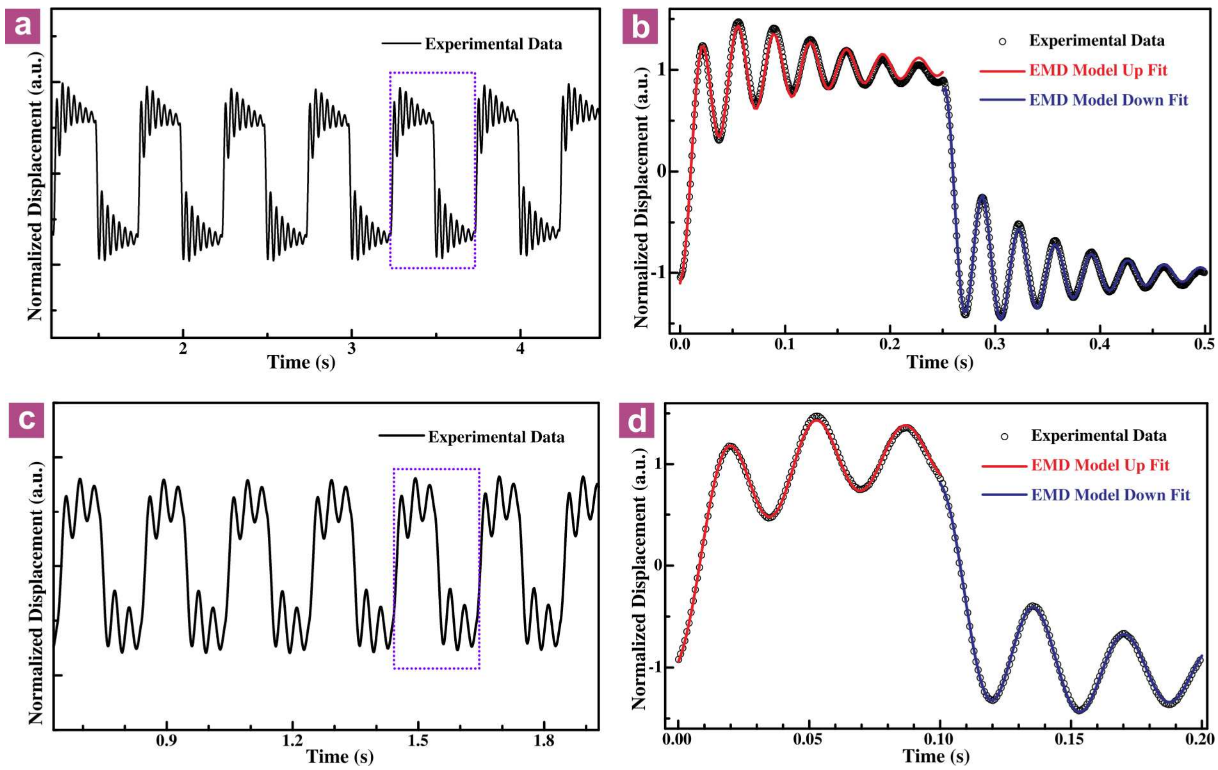

Figure S2. Normalized displacements of the same actuator under ±4 V square wave stimulation at frequencies of (a) 2 Hz and (c) 5 Hz, respectively. (b) and (d) are the extracted periods from the rectangular zones with dotted line frames in (a) and (c) for model analyses. Black open circles represent the experimental data while the red and blue solid lines are the minimum Reduced Chi-Sqr fitting lines using the EMD model (equation (1)). For clarity, the experimental data are adopted one point every 9 points. The fitting lines show perfect agreements with the experimental data.

Table S1. The comparisons of parameter values at different driving frequency.

t_0[a] ms	β[a] Hz	f_d[a] Hz	ξ[b] —	f_0[b] Hz	Frequency Hz
19	9.28	28.8	0.051	28.8	1
18	10.01	29.0	0.055	29.0	2
16	9.63	29.2	0.052	29.2	5

[a]Fitting results using the equation (1).

[b]Deriving results with the equations (S3) and (S4).

From Table S1, the response time constant τ of the actuator is 16-19 ms, the damping ratio of this underdamped system is calculated to be 0.05, and the natural frequency of the cantilever-type actuator is estimated to be 29 Hz which is corresponding to the fitting resonant frequency 29.7 Hz in Fig. 3b. In conclusion, the fitting and deriving parameter values for 2 and 5 Hz are well consistent with that for 1 Hz, further prove the high accuracy of our EMD model, and indicate its potential for evaluating other damping behaviors in mechanical systems based on kinds of different actuation mechanisms.

S5. A video for an actuator flapping at resonant frequency of 30 Hz with magnitude about 4 mm

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