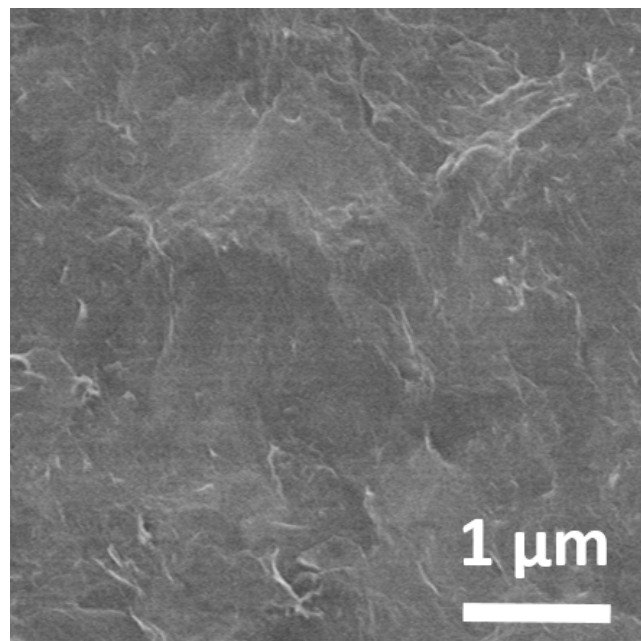
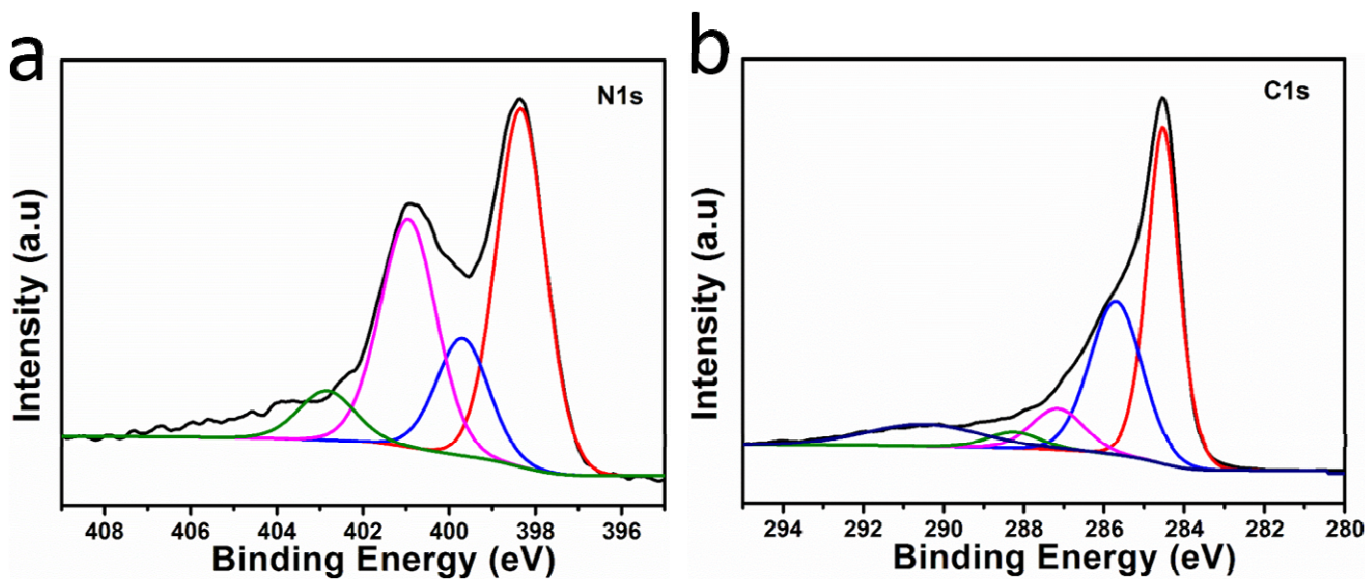


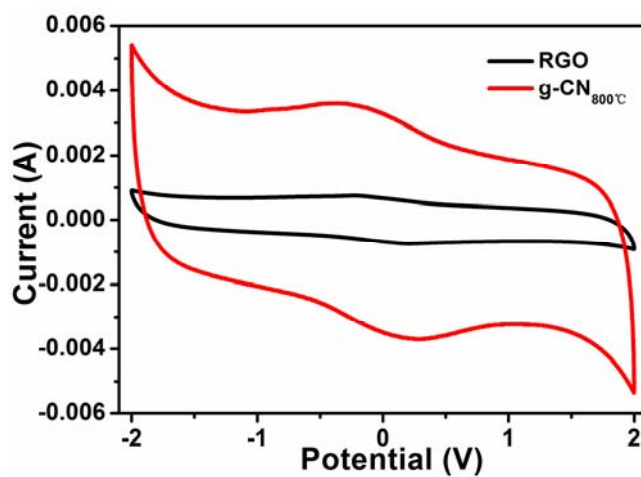
Supplementary Fig. 1 | Process of preparation g-CN. Glucose, urea and CNTs go through a two-step thermal treatment according to the process.



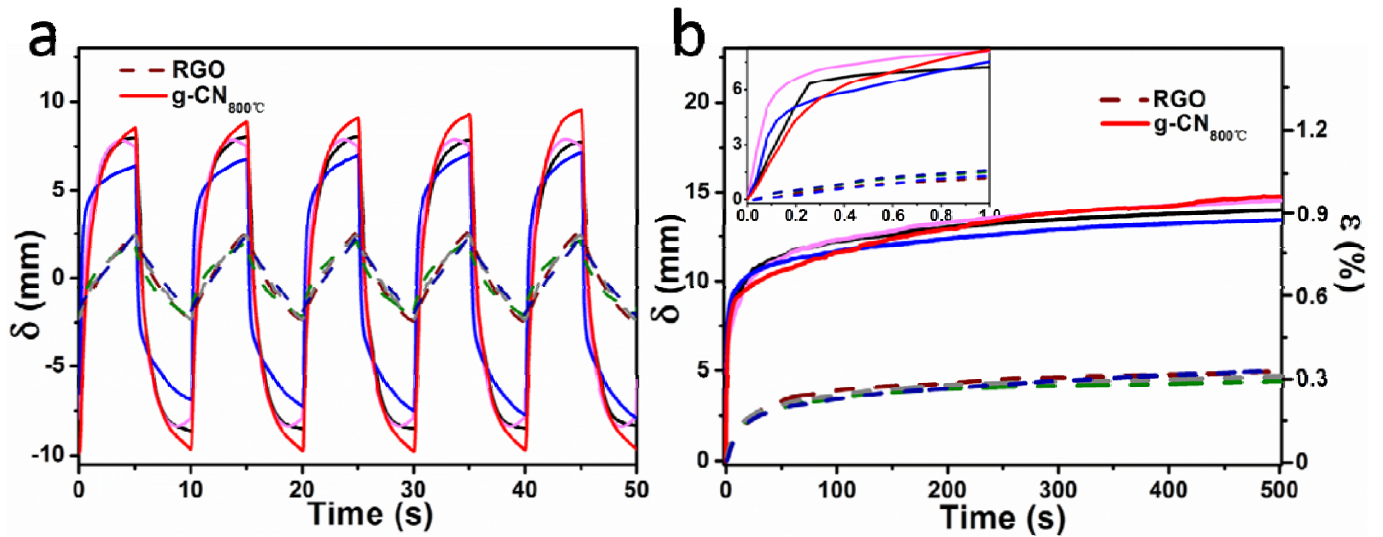
Supplementary Fig. 2 | Surface morphology of RGO. SEM image of RGO, scale bar 1 μm.



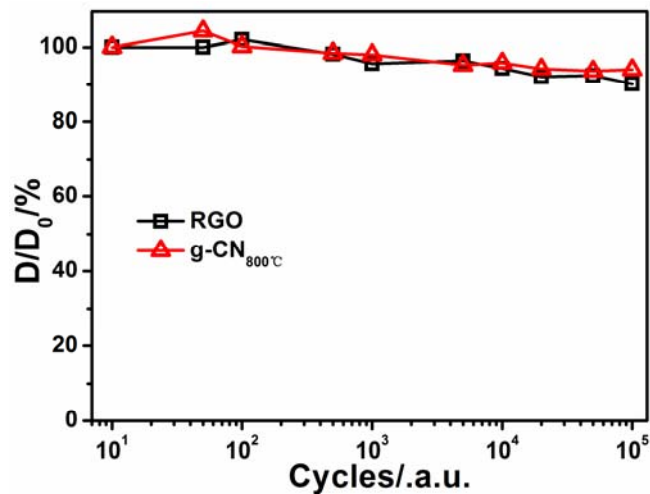
**Supplementary Fig.3 | XPS analysis of g-CN.** High resolution N1s (a) and C1s (b) XPS spectra of g-CN<sub>800°C</sub>.



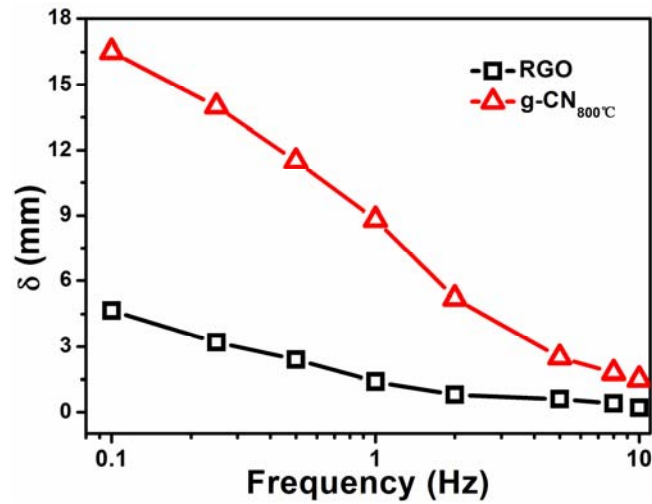
**Supplementary Fig. 4 | Charge storage behaviors of electrochemical actuators.** Cyclic voltammetry (CV) curves for actuators at 100 mV s<sup>-1</sup> sweep rate.



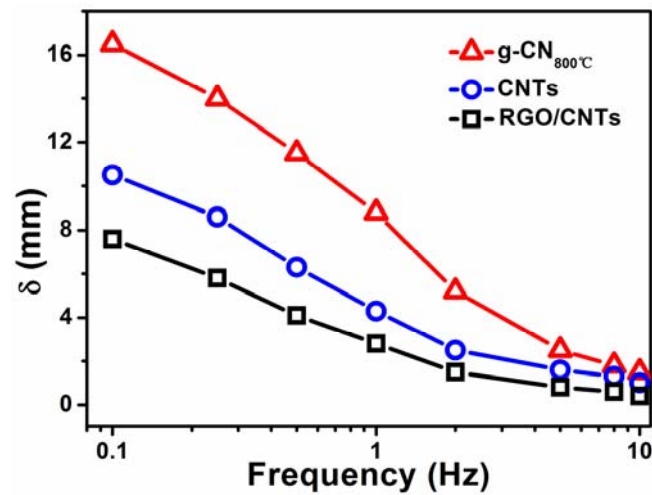
**Supplementary Fig. 5 | Bending performance of four pairs of actuator samples. (a)** Bending response of actuators at  $\pm 3$  V, 0.1 Hz. **(b)** Time-dependent displacement of actuators under 3 V. The dash lines stand for RGO actuators and solid lines represent g-CN<sub>800°C</sub> actuators.



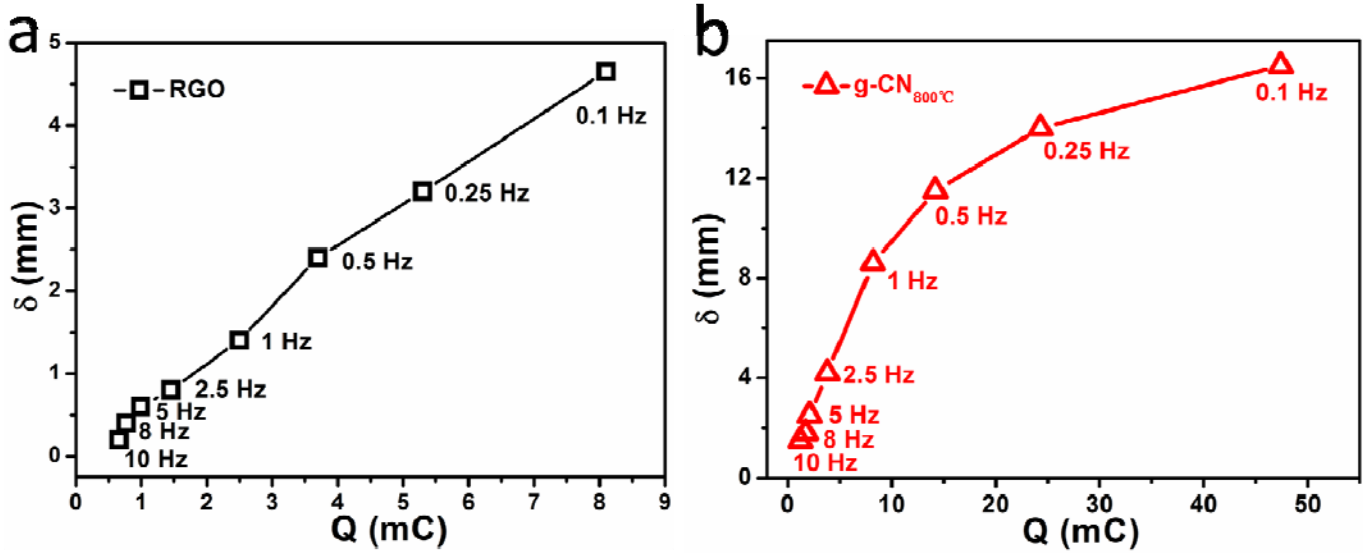
**Supplementary Fig. 6 | Actuation stability of electrochemical actuators.** Cycling test for RGO and g-CN<sub>800°C</sub> actuator under  $\pm 3$  V and 1 Hz applied square wave voltage stimulation,  $D_0$  represents the initial displacement value for tenth cycles.



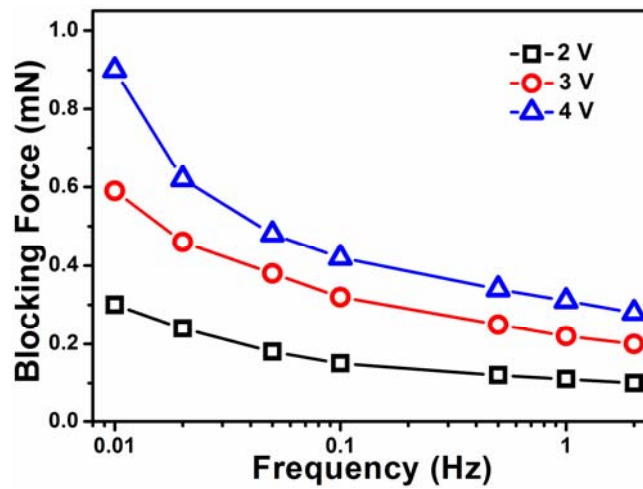
**Supplementary Fig. 7 | Wide frequency response of actuators.** Actuation performance of wide frequency response (0.1~10 Hz) of RGO and g-CN<sub>800°C</sub> actuators.



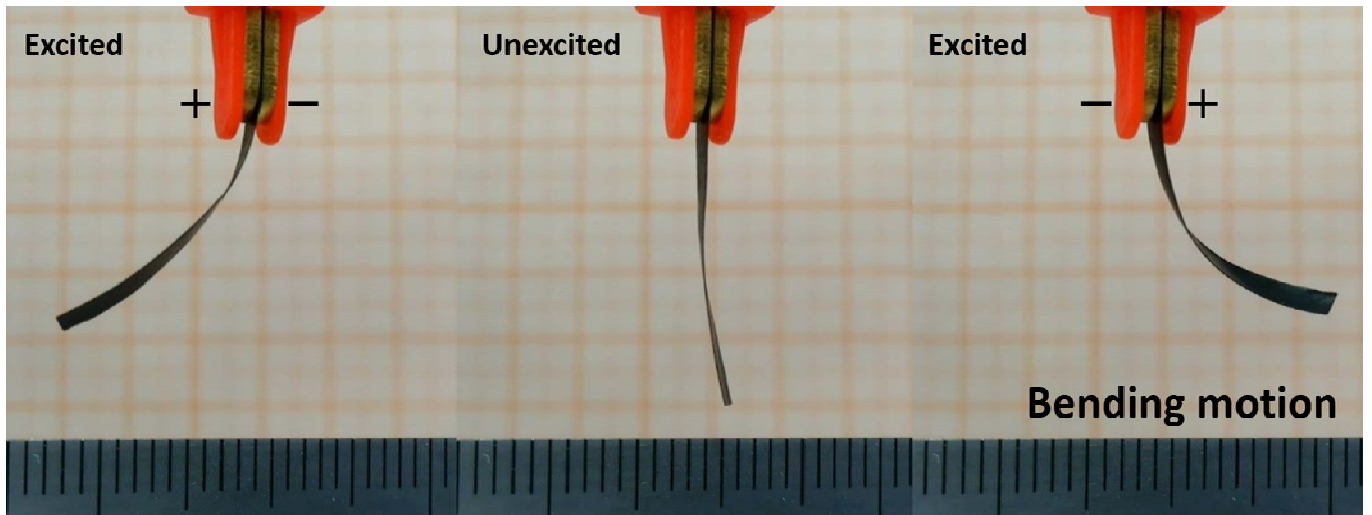
**Supplementary Fig. 8 | The comparison of actuation performance.** The actuation performance comparison of g-CN<sub>800°C</sub>, CNTs and RGO/CNTs electrode based actuators.



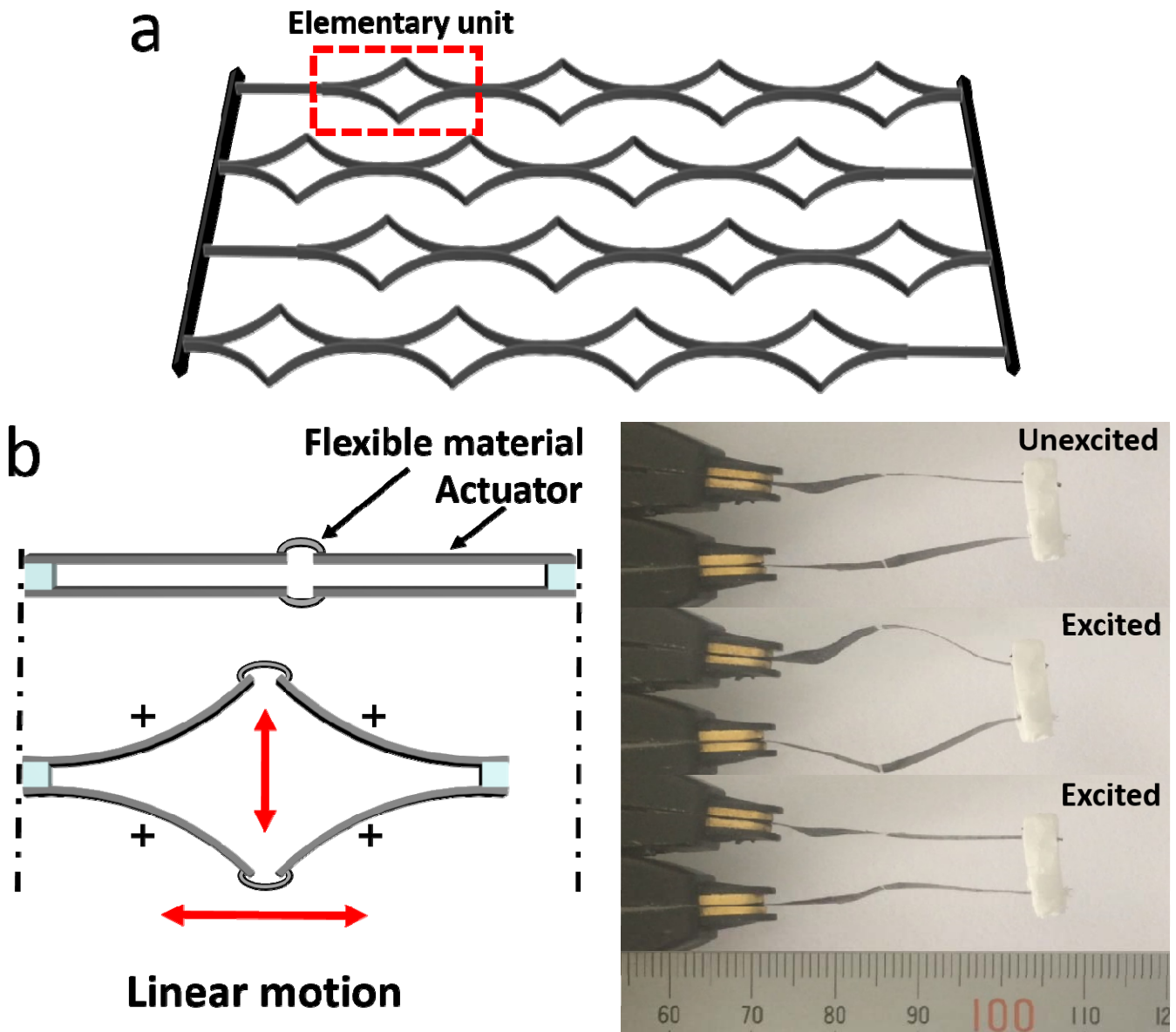
**Supplementary Fig. 9 | The relationships between bending displacements and charge injections.** Actuation bending displacements along with charge injections under wide frequency response (0.1~10 Hz) of actuators. (a) RGO and (b) g-CN<sub>800°C</sub>.



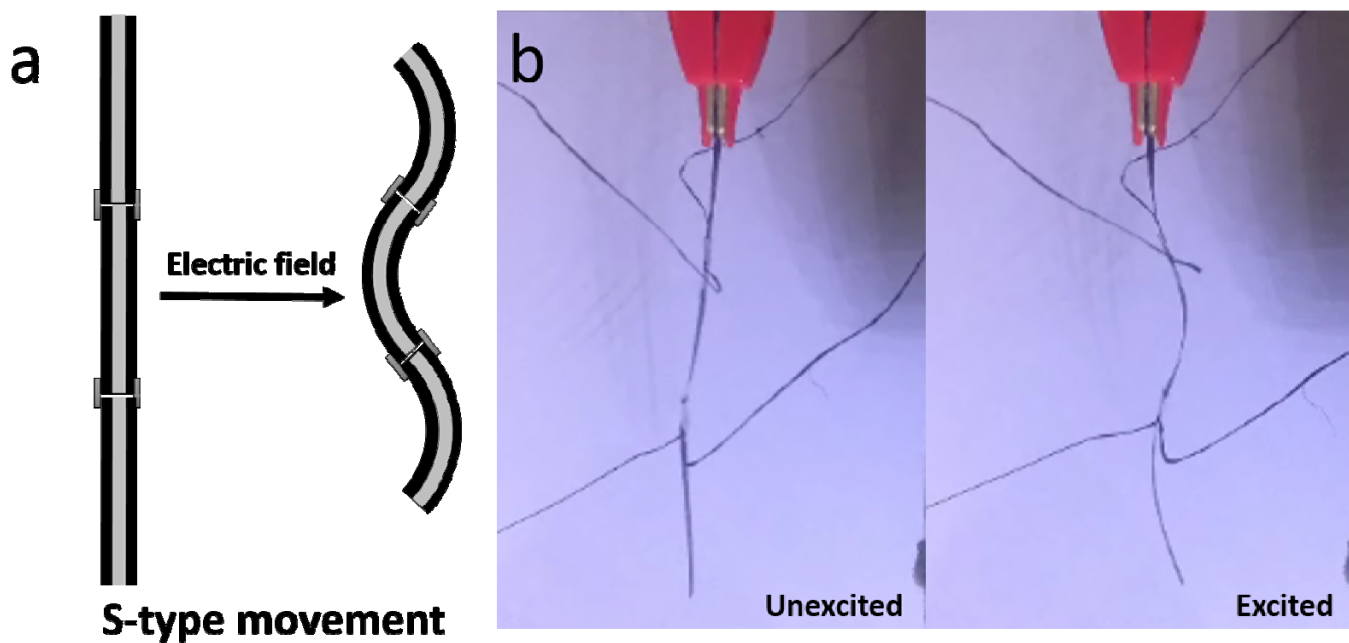
**Supplementary Fig. 10 | The mechanical force output of actuator.** Blocking force of g-CN<sub>800°C</sub> actuators under different applied voltages and frequencies stimulation.



**Supplementary Fig. 11 | Bending movement of actuator.** Bending motion of g-CN electrode based actuator under the frequency of 0.1Hz.



**Supplementary Fig. 12 | Schematic illustration of a linear motion actuator. (a)** Structure of a linear actuator; **(b)** An elementary unit of the proposed linear actuator and its movement under electric stimulus.



**Supplementary Fig. 13 | Schematic illustration of an actuator with S-type movement. (a)** Structure of the S-type actuator; **(b)** Photographs of the S-type movement actuator under electric stimulus.

**Supplementary Table 1 | A summary of pore size distribution and its contribution to SSA**

Materials	SSA ( $\text{m}^2\text{g}^{-1}$ )	Micropore (%)	Mesopore (%)		Macropore (%)
		<2 nm	2-4 nm	4-50 nm	>50 nm
RGO	108.91	15.24	30.26	53.94	0.56
g-CN <sub>800°C</sub>	284.3	34.49	16.07	47.21	2.23