

Supporting information for publication

## Graphene oxide based moisture-responsive biomimetic film actuators with nacre-like layered structures

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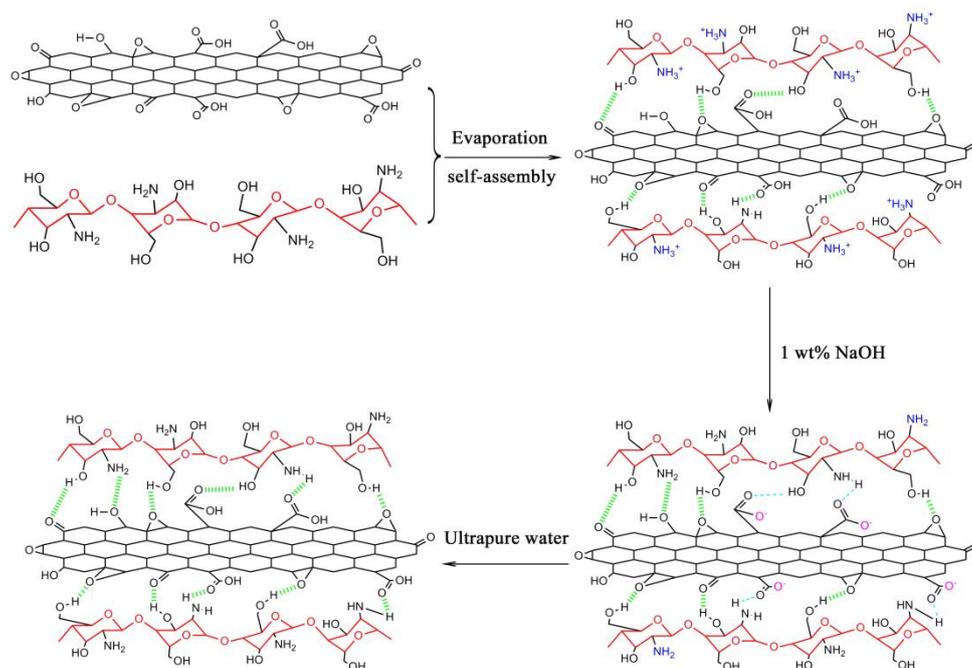


Fig. S1 Schemes for fabricating bio-inspired nacre-like CS/GO composite films.

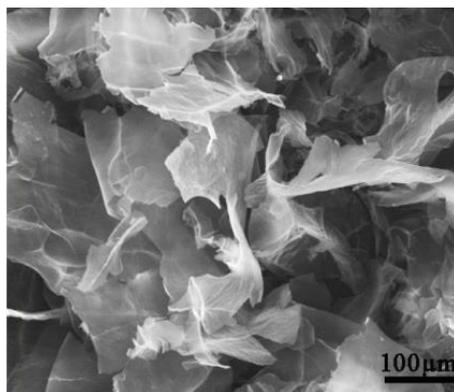


Fig. S2 SEM image of freezing-dried GO.

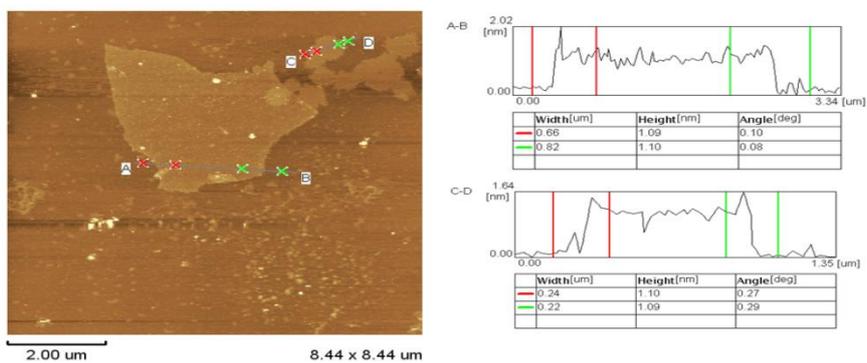


Fig. S3 A typical tapping-mode AFM image of GO sheets on Si wafer.

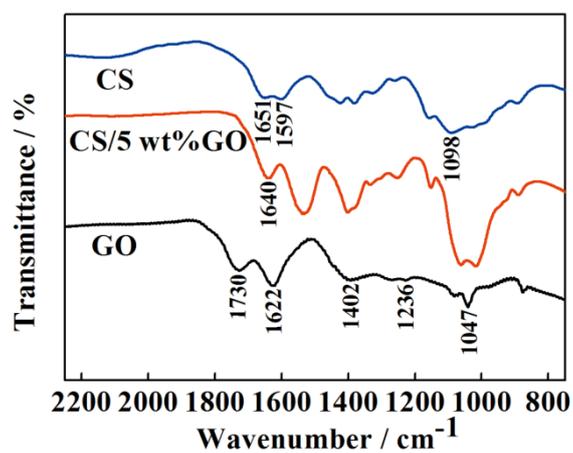


Fig. S4 FTIR spectra of GO, CS and CS/GO.

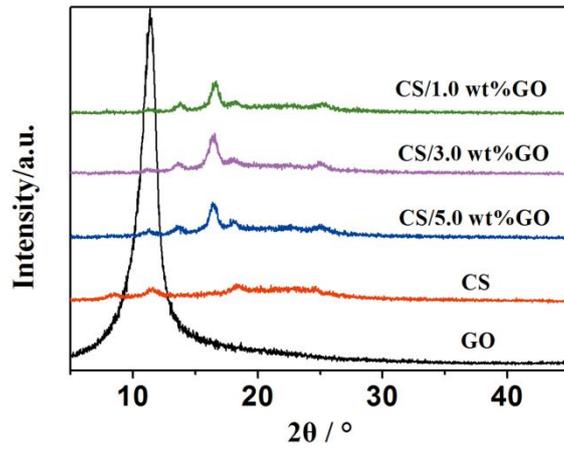


Fig. S5 XRD patterns of CS, GO and CS/GO.

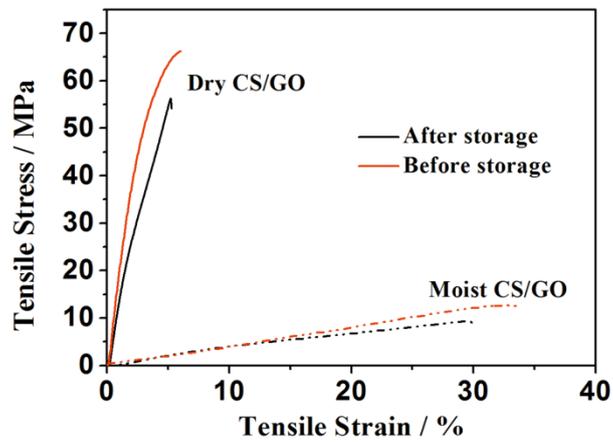


Fig. S6 Typical stress-strain curves of the resulting hybrid films before and after six-month open storage.

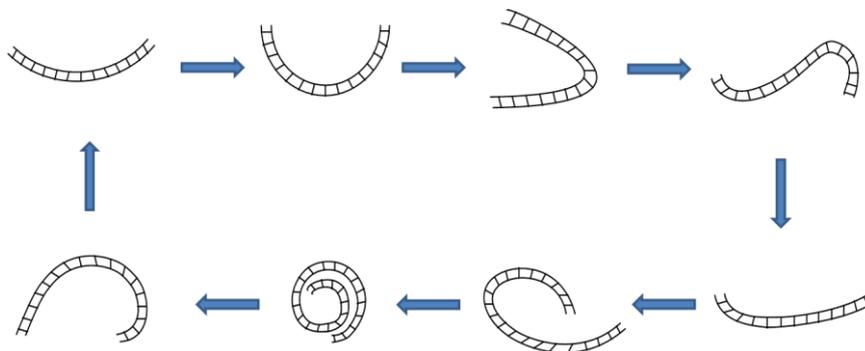


Fig. S7 Eight representative sketches of humidity-induced locomotion of CS/GO composite film.

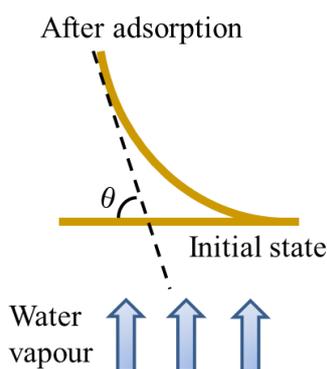


Fig. S8 Schematic showing the definition of bending angle ( $\theta$ ).

## Supplementary methods

**SEM image analysis.** The obtained GO sheets exhibited a translucent yarn-like and highly wrinkled surface structure. SEM image (Fig. S2) showed that GO was fully exfoliated into individual sheets with lateral dimensions ranging from several micrometer to several tens of micrometer.

**AFM image analysis.** The AFM image is shown in Fig. S3. GO could be easily dispersed in water by ultra-sonication to form a stable dispersion composed of fully exfoliated, individual GO sheets. It is clear from this figure that the GO nanosheets were of irregular shape with uniform thickness ( $\sim 1.1$  nm) according to cross-section analysis.

**FTIR spectrum analysis.** The FTIR spectrum of GO helps to illustrate the existence of plentiful oxygen-containing groups. As can be seen in Fig. S4, the GO absorption peaks are located at ca.  $1730$ ,  $1622$ ,  $1402$ ,  $1237$  and  $1047$   $\text{cm}^{-1}$ , which attribute to carbonyl (C=O) stretching vibration, C=C stretching vibration, carboxyl C-O stretching vibration, C-OH stretching vibration, and epoxy C-O stretching vibration, respectively. CS shows three typical absorption peaks centered at ca.  $1651$ ,  $1569$  and  $1098$   $\text{cm}^{-1}$ , which correspond to the C=O stretching vibration, the deformation bending of N-H, and the stretching vibration of C-O, respectively. Compared with pure CS and GO film, the peak at ca.  $1730$   $\text{cm}^{-1}$  belonging to carbonyl group disappears in the spectra of CS/GO hybrid film. Furthermore, the peak at ca.  $1651$   $\text{cm}^{-1}$  related to amide group in CS gradually shifts to  $1640$   $\text{cm}^{-1}$  in composite film. These can be ascribed to the synergistic effect of hydrogen bondings and the electrostatic interactions between polycationic CS and negatively charged GO.

**XRD patterns analysis.** As can be seen in Fig. S5, the XRD patterns of CS exhibits three characteristic diffraction peaks around  $2\theta = 8.6^\circ$ ,  $11.7^\circ$  and  $23.7^\circ$ . The first two peaks correspond to the hydrated crystalline structure, and the diffraction peak at higher  $2\theta$  angle indicates the existence of an amorphous structure. After addition of GO, apart from the three characteristic peaks of CS mentioned above, a new obvious diffraction peak can also be observed around  $2\theta = 17^\circ$ , which indicates the increased crystalline structure by the incorporation of GO. However, the typical diffraction peak at  $2\theta = 11.9^\circ$  of GO disappears, indicating the formation of fully exfoliated structure of GO sheets in the polymer matrix. In addition, the CS/GO hybrid films with GO contents in the range 1.0-5.0 wt % have almost the same XRD patterns, reflecting that they have similar crystalline structures.

**Mechanical properties analysis.** To investigate the effect of storage time on the properties of CS/GO hybrid films, the mechanical behaviors of CS/GO layered hydrogel films after six-month open storage were investigated by tensile tests at room temperature. The typical stress-strain curves are shown in Fig. S6. It is interesting to see that both tensile strength and elongation at break of the CS/GO hybrid film show no obvious reduction after six months, giving the conclusion that this nanocomposite film exhibits good stability.

## Supplementary movies

Movies are provided as .avi files.

Movie S1. A 60- $\mu\text{m}$ -thick CS/GO film is flipping over a porous substrate. The water temperature is maintained at  $40^\circ\text{C}$ .

Movie S2. A 47- $\mu\text{m}$ -thick CS/GO film ( $5\text{ cm} \times 5\text{ cm}$ ) is flipping over a porous substrate. The water temperature is maintained at  $40^\circ\text{C}$ .

Movie S3. Driven by humidity gradients, a strip of CS/GO film is rapidly bending upwards; a 60- $\mu\text{m}$ -thick CS/GO film is flipping over one's hand.

Movie S4. A 4.8 mg, 37- $\mu\text{m}$ -thick CS/GO film is lifting a cargo 50 times heavier than itself; a 15 mg, 43- $\mu\text{m}$ -thick film loaded with a small cargo 10 times heavier than itself is flipping over a porous substrate.

Movie S5. Driven by humidity gradients, a motor based on CS/GO film is moving forward with a speed of  $2.5\text{ cm s}^{-1}$ .

## Supplementary References

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