

**Bio-inspired homogeneous graphene oxide actuator driven by moisture gradients**

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## **1. Experimental section**

### **1.1 Fabrication of GO nanosheets**

Graphite was purchased from Uni-Chem. The GO nanosheets were prepared by expansion, preoxidation, and oxidation according to a previous literature method.<sup>1</sup>

#### **Fabrication of Flexible Free-Standing Uniform GO film**

To prepare the GO film, GO aqueous (5 mL, 5 mg/mL) dispersion was filtrated through an acetate fiber Millipore filter (47 mm in diameter and 0.45  $\mu\text{m}$  in pore size), followed by air drying at room temperature. Finally, the free-standing GO film was peeled off carefully for further use. The thickness of GO films was controlled by adjusting the volume of aqueous dispersion during the filtration process, and a series of GO films with thicknesses varying from 4 to 20  $\mu\text{m}$  were prepared.

### **1.2 Characterization**

The exact widths and the thicknesses were carefully determined by micrometer calipers and scanning electron microscopy (SEM), respectively. SEM images were obtained with SEM (Quanta 400 FEG). X-ray diffraction (XRD) analysis was performed on X'Pert-Pro MPD diffractometer with a Cu K $\alpha$  radiation source at room temperature (the angle measurement accuracy is 0.02°). Atomic Force Microscope (AFM, Dimension Icon) images were obtained in the tapping mode on a Multimode 8 model scanning probe microscope, for which the GO dispersion was drop-casted onto freshly cleaved mica surfaces. XPS analysis was performed on a Shimadzu Axis scope, using Mg-K $\alpha$  as radiation resource.

### **1.3 Testing of the moisture actuation behaviors**

The experiments were performed in a home-made humidity chamber (Figure S4), where the relative humidity (RH) can be adjusted from 60% to 100% by saturated aqueous solutions of NaBr, CH<sub>3</sub>COOLi, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, BaCl<sub>2</sub> and K<sub>2</sub>SO<sub>4</sub>, which yielded approximately 57%, 68%, 79%, 90% and 97% relative humidity (RH), respectively. The upper of the chamber has a constant RH of about 20%  $\pm$  4% by anhydrous calcium chloride. In the middle of the humidity chamber, there was a window of a diameter  $\approx$ 3 cm (Figure S1), with a removable cover lid that could be opened to position a film instead. Positioning the film there allowed imposing a well-controlled relative humidity difference ( $\Delta\text{RH}$ ) across the GO film. The bending/unbending process of GO film was record with a digital camera (Canon EOS 70D). The bending curvature of the actuator was determined by fitting the optical image of the curved strip to a circle with a certain size (Fig. S4). GO film flips over moist paper.

Supplementary Movie 1. GO film (12  $\mu\text{m}$ ) flips spontaneously and continuously over moist paper substrate (40  $^{\circ}\text{C}$ ) substrate at room temperature (20  $^{\circ}\text{C}$ ) and humidity (30%).

Supplementary Movie 2. GO film bends upwards and recovery.

Supplementary Movie 3. GO film lifts a cargo at room temperature (20  $^{\circ}\text{C}$ ) and humidity (30%).

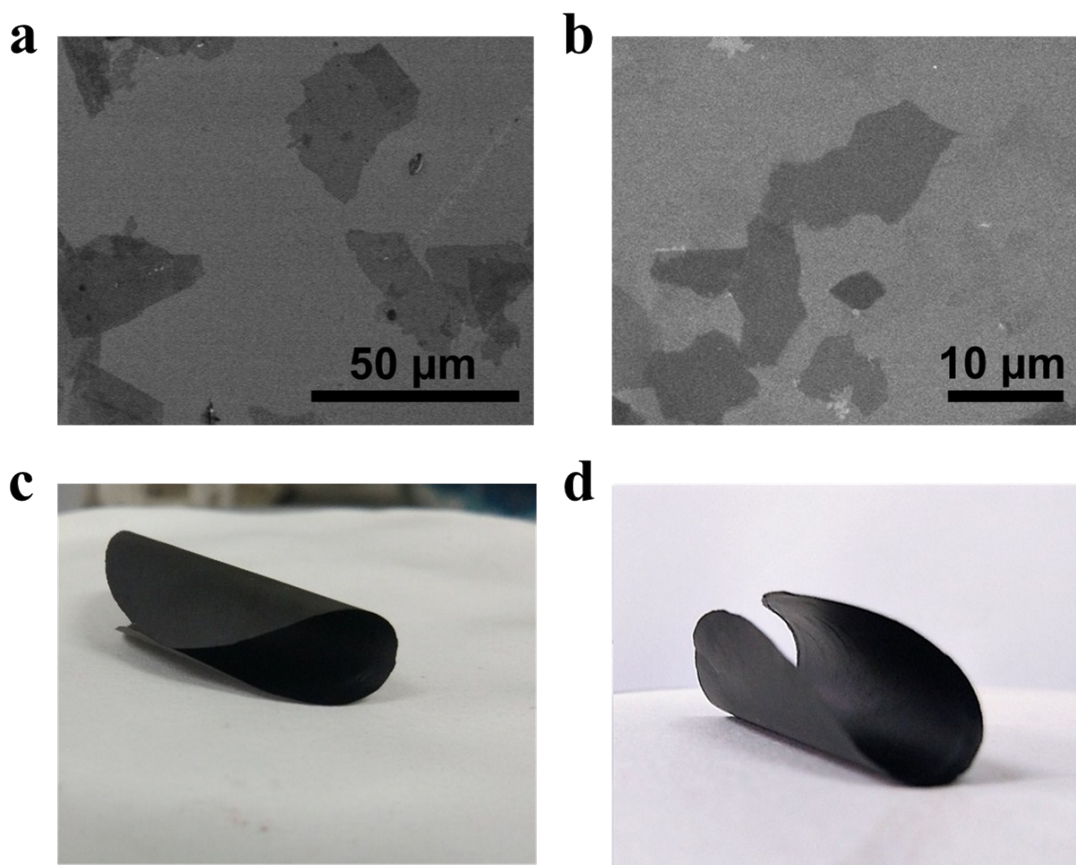


Figure S1. SEM images of GO sheets with the size of (a) 20~30  $\mu\text{m}$  and (b) 6~10  $\mu\text{m}$ . Photographs of GO films with the size of (c) 20~30  $\mu\text{m}$  and (d) 6~10  $\mu\text{m}$  on the moist substrate.

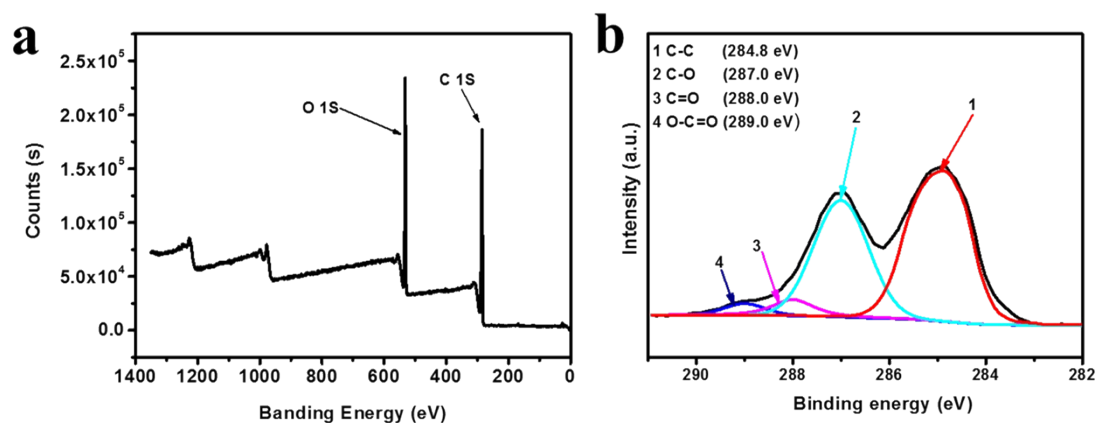


Figure S2. (a) XPS survey spectrum and (b) high resolution of C 1s XPS of GO film.

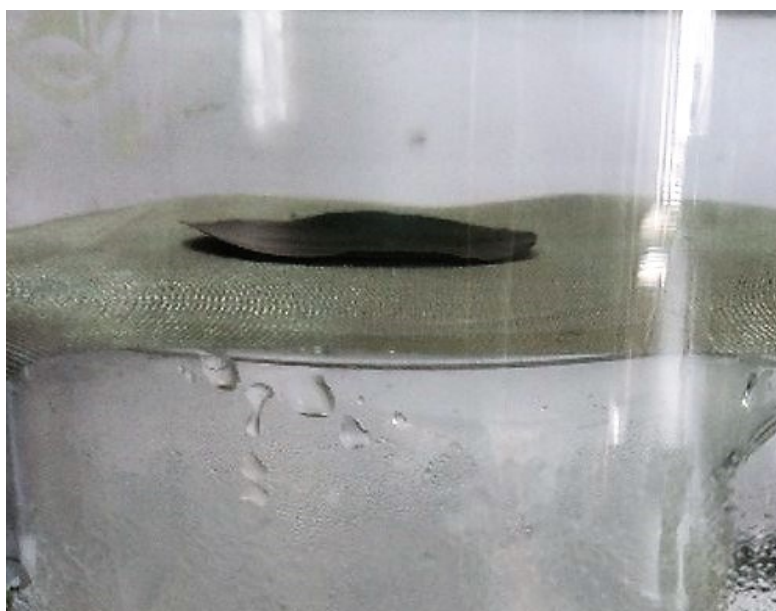


Figure S3. Photograph of GO film in a sealed chamber saturated by water vapor.

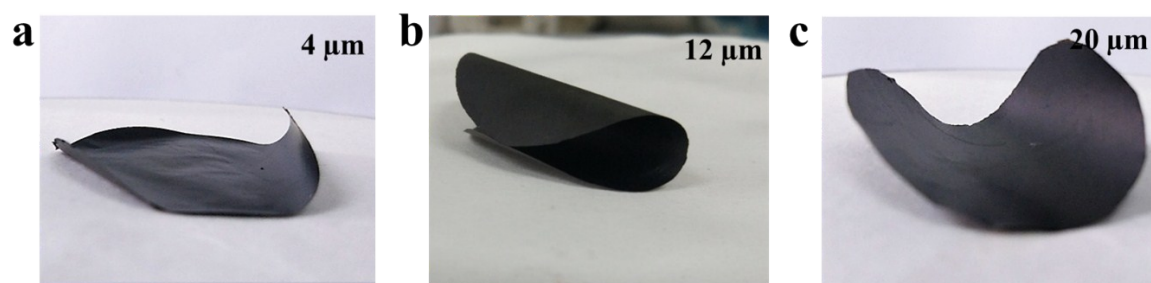


Figure S4. Photographs of GO films on the moist substrate with the thickness of (a) 4 μm, (b) 12 μm and (c) 20 μm.

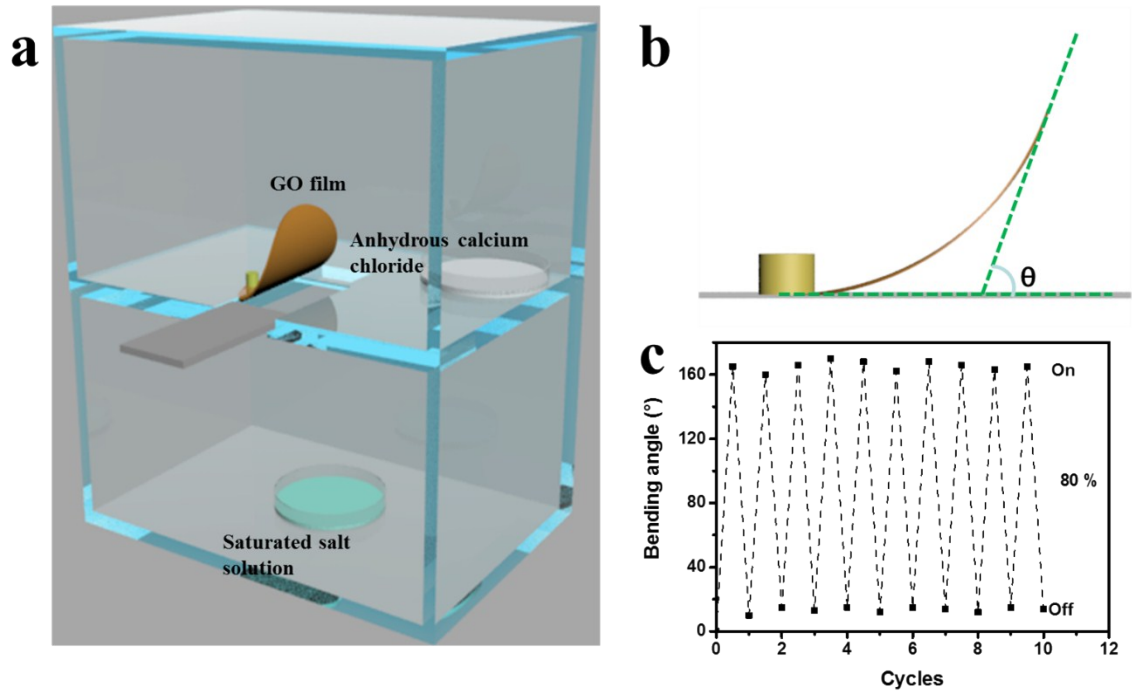


Figure S5. (a) Experimental set-up for measuring the bending curvature of GO film at different relative humidity difference. The relative humidity of the lower part of the chamber was controlled by saturated salt solution. The relative humidity of the upper part of the chamber was fixed at about 20% by anhydrous calcium chloride. The samples were then located at a hole in the middle of the chamber and driven to deflect by the relative humidity difference between the chamber top and bottom. (b) Determination of degree. (c) Reversible and repeated deflection of the GO film (thickness 12  $\mu\text{m}$ ) with the cover lid opened and closed at  $\Delta\text{RH}=80\%$ .

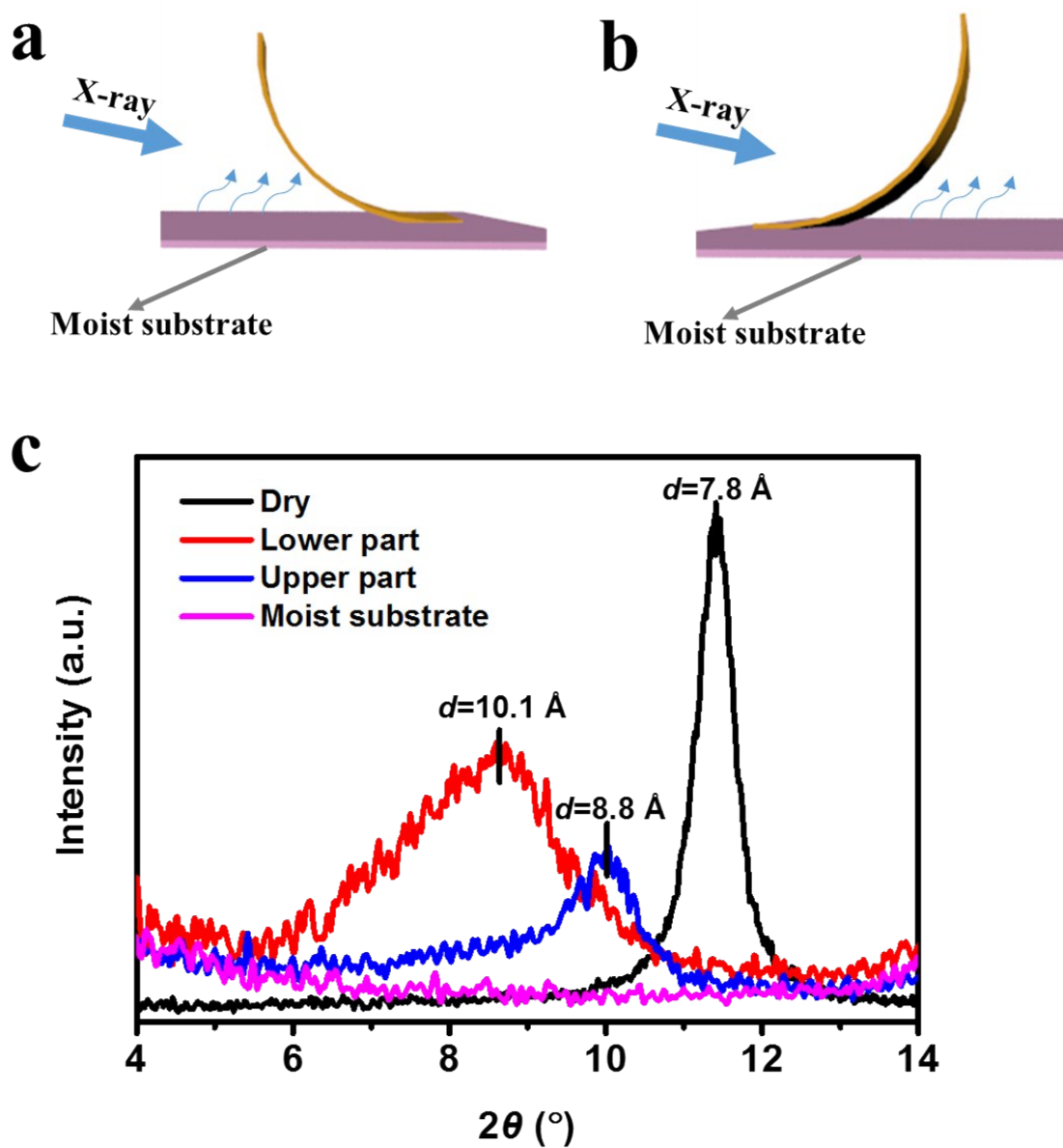


Figure S6. Schematic illustration of XRD measurement in the (a) lower and (b) upper part of GO film. (c) XRD pattern of dry GO film and the different part of GO film bending on the moist substrate, and the corresponding d-spacing.

Renfrence

- 1 Z. Xu , H. Sun , X. Zhao , C. Gao , Adv. Mater. 2013 , 25 , 188.