Supporting Information

for

Magnetic field assisted fabrication of quasi-bilayered, multi-responsive and patternable actuators

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1. Experimental

1.1 Materials

Polyvinylidene fluoride (PVDF, M_W~534000) was purchsed from Sigma-Aldrich. Graphite powder (50 mesh, ≥99%), concentrated sulfuric acid (95~98%), potassium permanganate ($KMnO_4$), hydrogen peroxide (H_2O_2, 30%), concentrated hydrochloric acid, ferric chloride hexahydrate ($FeCl_3 \cdot 6H_2O$), and ferrous chloride tetrahydrate ($FeCl_2 \cdot 4H_2O$) were obtained from Sinopharm Chemical Reagent Co., ltd. N, N-Dimethylacetamide (DMAc), acetone, and ammonium hydroxide were purchased from Shanghai Lingfeng Chemical Reagent Co., LTD. All reagents were used without further purification.

1.2 Synthesis of Fe_3O_4 particles decorated graphene oxide (Fe_3O_4@GO)

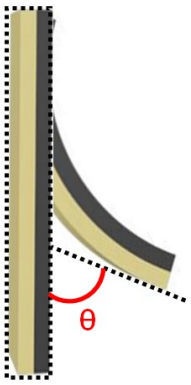
Grahpene oxide (GO) was prepared according to widely adopted Hummers method[1, 2]. For the preparation of Fe_3O_4@GO, GO powder (0.1 g) was first dispersed in 250 mL deionized water and sonicated for 1 h to fully exfoliate GO into single layered sheets. In parallel, 0.1 g $FeCl_3 \cdot 6H_2O$ and 0.2 g $FeCl_2 \cdot 4H_2O$ were dissolved in 15 mL deionized water to obtain a mixed Fe^{3+}/Fe^{2+} solution, the solution was then slowly added into the GO dispersion dropwise, and then mechanically stirred for 1 h. After that, ammonium hydroxide was dropped into the mixture until the pH was increased to ~10, and the color of mixture truned from yellow to black. The solution was heated to 90 °C and maintaied at this temperature for 3 h to complete the reaction. The black powder was collected by magnet and washed by deionized water until neutral, and lyophilization was applied to obtain the final products.

1.3 Fabrication of Fe_3O_4@GO/PVDF composite actuator

For the typical fabrication of Fe_3O_4@GO/PVDF actuator, 0.2 g Fe_3O_4@GO powder was first dispersed in 10 mL DMAc, and sonicated for 30 min until homogeneous dispersion was formed. In parallel, PVDF (1 g) was fully dissolved in 15 mL DMAc solvent with mechanical stirring, and these two solution was mixed together and sonicated for another 30 min. The homogeneous mixture was poured into a Teflon mold and dried in a blasting oven at 80 °C for 12 h. For the quasi-bilayered actuator, a magnet was put under the mold during drying process.

1.4 Characterizations

Transmission electron microscopy (TEM) images were observed by JEOL2100F at a voltage of 120 kV, aqueous dispersion of Fe_3O_4@GO was dropped on copper mesh and dried at the temperature of 25 °C for TEM characterization. The X-ray diffraction (XRD) pattern of Fe_3O_4@GO was collected in a D/max-2550 PC instrument. X-ray photoelectron spectroscopy (XPS) analysis was recorded with a Thermo Escalab-250Xi (America) spectroscopy with an Al KRX-ray source (1487 eV) at a power of approximately 150 W. The cross-sections of Fe_3O_4@GO/PVDF samples were characterized by Field Emission Scanning Electron Microscope (*/S-4800, Japan) at an activation voltage of 5 kV, and energy dispersive spectrometer (EDS) were used to analyze the distribution of elements. Thermal images were obtained using a thermal image camera (FL-IR A400). For measurement of the actuation performance of actuator when stilumated by NIR light or acetone vapour, the bending angle θ is defined as follow:



The actuation forces of actuators generated upon stimulus were recorded by an electromechanical universal material testing machine (MTS C44-104, U.S.). During test, the two ends of actutors were fixed to the fixture, then NIR light or acetone saturated filter paper were applied to the middle part of actuators, the generated forces were recorded by the testing machine. The sizes of specimens were about 30 mm × 10 mm × 60 μm (length × width × thickness).

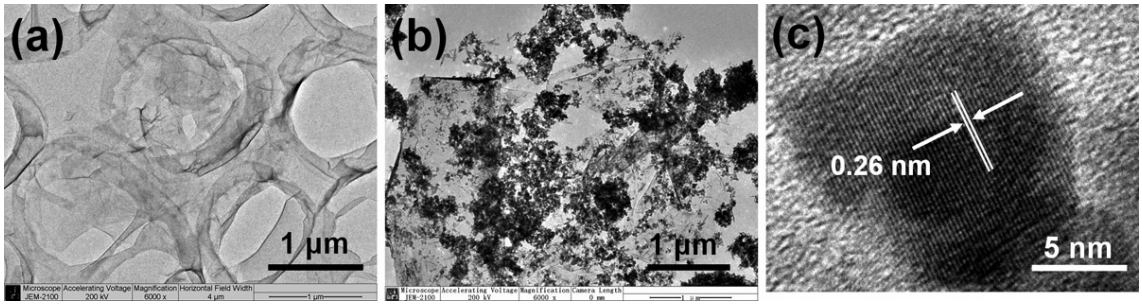


Fig. S1 (a) TEM image of GO. (b) TEM image of Fe_3O_4@GO. (c) Magnified TEM image of Fe_3O_4 nanoparticles.

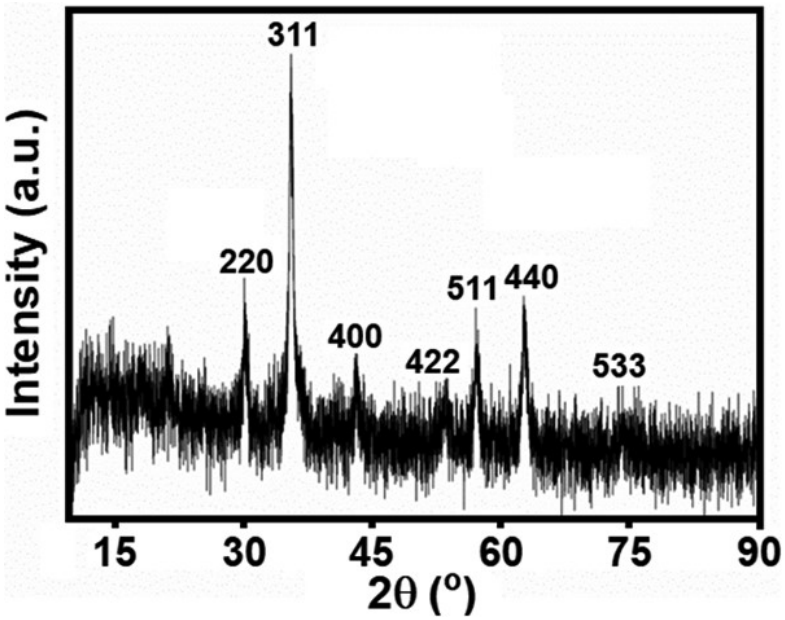


Fig. S2 XRD pattern of Fe_3O_4@GO.

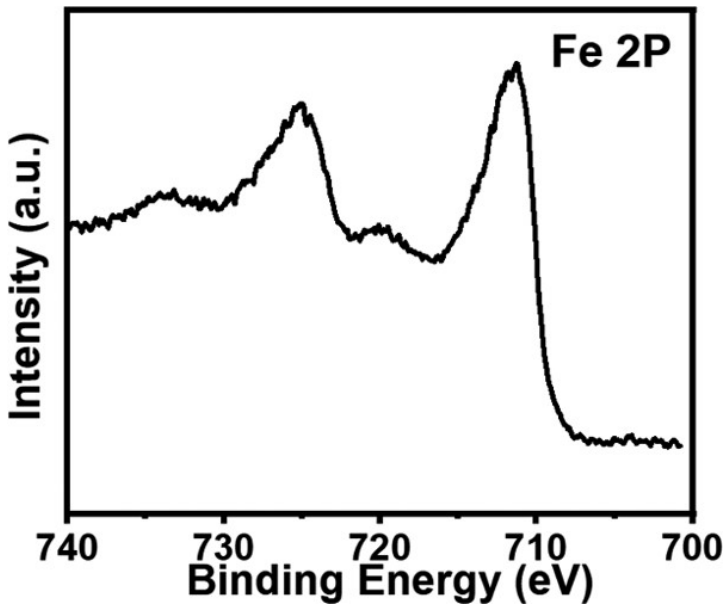


Fig. S3 XPS spectrum of Fe_3O_4@GO in the Fe 2P region.

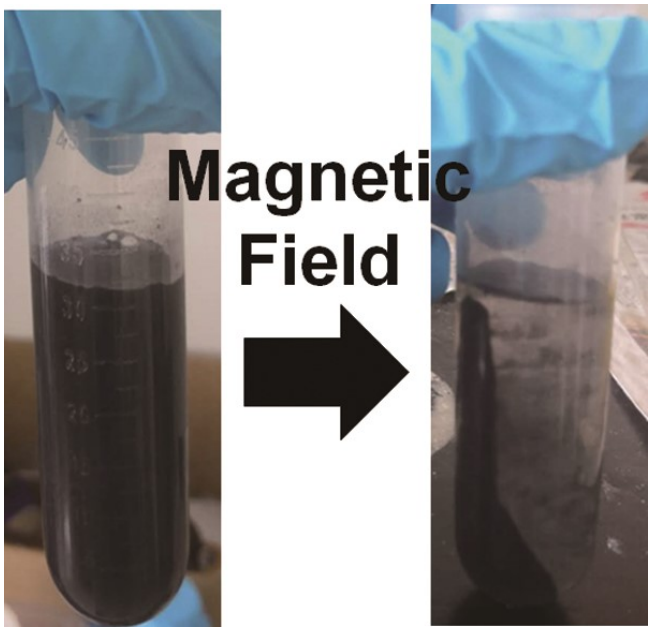


Fig. S4 Photographs of Fe_3O_4@GO under magnetic field.

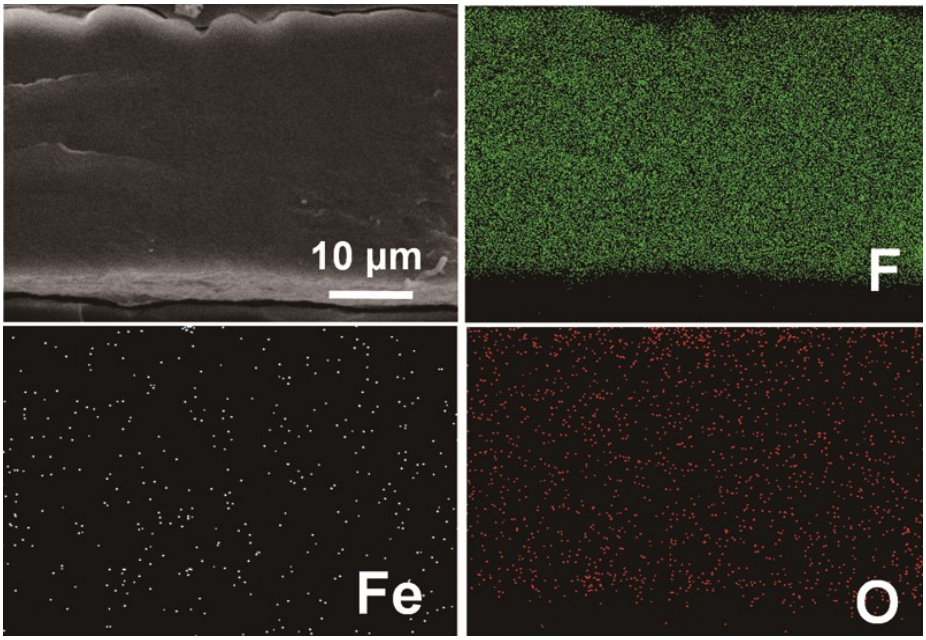


Fig. S5 EDS mapping of distribution of F, Fe, and O elements in Fe_3O_4@GO/PVDF dried without the assisted of magnetic field.

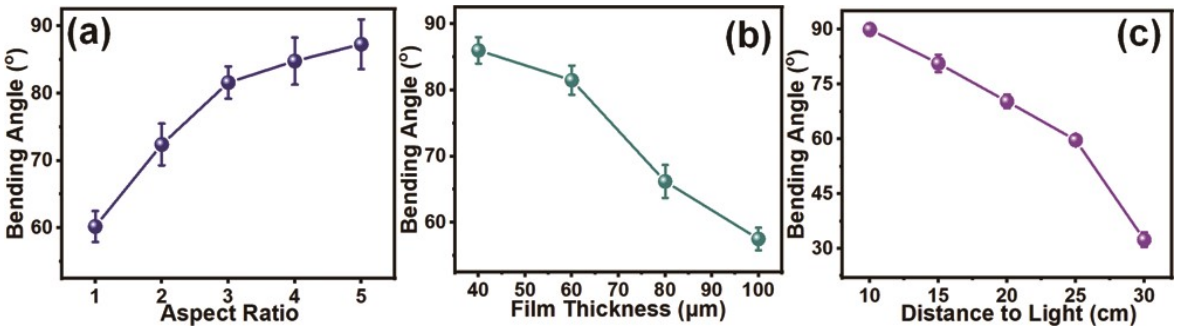


Fig. S6 The effect of (a) aspect ratio (the film thickness and distance to light source are fixed at 60 μm and 15 cm), (b) film thickness (the aspect ratio of actuator and distance to light source are fixed at 3 and 15 cm), and (c) distance to light source (the aspect ratio and thickness of actuator are fixed at 3 and 60 μm) on the bending angle of Fe_3O_4@GO/PVDF actuator.

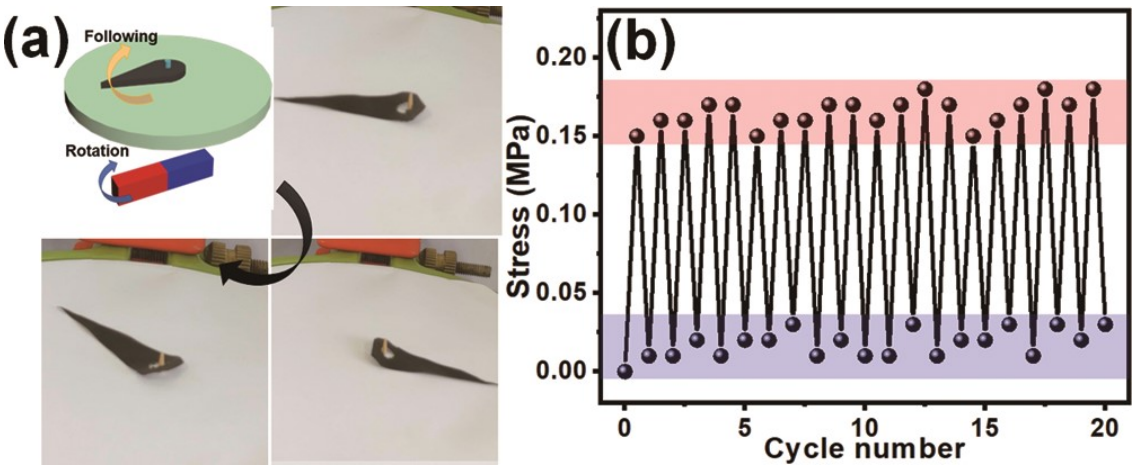


Fig. S7 (a) photographs of the movement of Fe_3O_4@GO/PVDF actuator following a magnet. (b) Stress generated by Fe_3O_4@GO/PVDF actuator under magnetic field with different cycles.

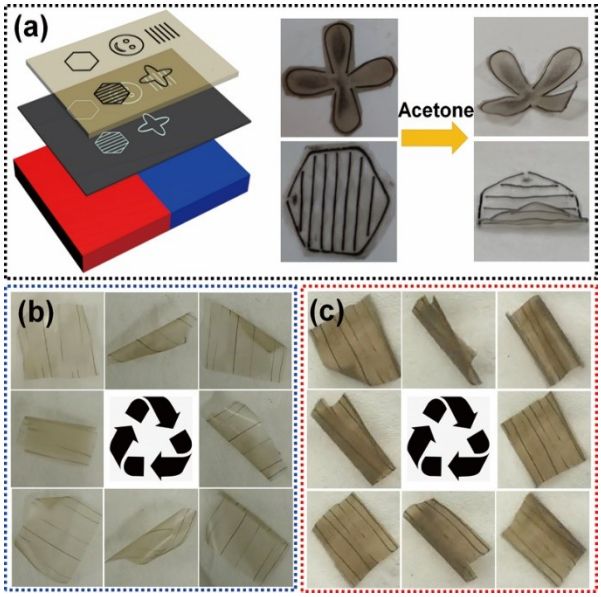


Fig. S8 (a) Illustration of the patterning process of actuator and the deformation of different patterned actuators. (b) Photographs of the continuous flipping process of PVDF film (the line was drawn by pen) on an acetone wetted substrate. (c) Photographs of the continuous flipping process of patterned Fe_3O_4@GO/PVDF film on an acetone wetted substrate.

References

1. W. S. Hummers and R. E. Offeman, *J. Am. Chem. Soc.*, **1958**, *80*, 1339.
2. N. I. Kovtyukhova, P. J. Ollivier, B. R. Martin, T. E. Mallouk, S. A. Chizhik, E. V. Buzaneva and A. D. Gorchinskiy, *Chem. Mater.*, **1999**, *11*, 771−778.