Supporting Information

Experimental section

1. Materials. Poly (vinylidene difluoride) (Kynar HSV900) was obtained from a commercial supplier in China (Dowgwan Fuqiao Plastic Technology Co., Ltd). Polydimethylsiloxane (PDMS), Fe particles (diameter: ~3 μm), and *N, N*-dimethylformamide (DMF) were purchased from Sinopham Chemical Reagent Co. Ltd., China. The images and videos were recorded with a smart phone (Smartisan R1). All mechanical data were collected on an electronic tensile testing machine (HY-0580, purchased from Shanghai Hengyi Precise Instrument Limited Company). A electromagnetic device was assembled, in which the magnetic coils and current control system were purchased from an on-line shop (https://www.taobao.com/).

2. Preparation of PVDF single layer. We first prepared PVDF solution by dissolving 1 g of PVDF powder into 20 mL of DMF in a 100 mL beaker with vigorous stirring at 90 °C for 4 hours. We then cast the solution on a pre-cleaned glass slide (75 mm × 75 mm × 3 mm) and shifted it onto a heating plate. The glass slide was heated at 80 °C for 1 h to give rise to a single-layer PVDF film. The PVDF film was kept on the glass slide for the next use.

3. Plasma treatment. The PVDF surface was treated by a plasma cleaner (PDC-32G-2). A rotary pump was connected to achieve a base pressure of 1×10^{-3} mbar. Oxygen was supplied by airflow. The treating time was 5 min for each sample.

4. Preparation of bilayer of PVDF/PDMS@FePs. The PVDF films on a glass slide were respectively treated with oxygen plasma for 5 min to introduce hydroxyl groups on the surface. PDMS@FePs mixture was then casted on the plasma-treated PVDF surface by a spin-coating process. The spin rate varied from 300 to 900 rpm to prepare thickness-different PDMS@FePs layers. After the spin coating, the glass slides with PVDF/PDMS@FePs were heated in a vacuum oven at 80 °C for 5 hours to give dried bilayers of PVDF/PDMS@FePs. Please note here that the PDMS@FePs mixtures were prepared by adding different amount of Fe particles (0.5 g, 1.0 g, 1.5 g) to the mixture of 5 g PDMS and 0.5 g curing agent.

5. Interfacial force of the bilayer. The tests were carried out in ambient air at room temperature. We estimated the interfacial adhesion force of bilayers through a standard 90° peeling test on a mechanical tester, utilizing a 50 N load cell. We adhered PVDF side of the bilayer on a glass surface by a dual adhesive tape, and the PDMS@FePs layer was covered with a rigid backing, so that the tested results could reflect the real interfacial force. All the 90° interfacial tests were proceeded at a peeling rate of 50 mm/min. The interfacial forces over the width of the bilayer strip were recorded with the peeling displacement.

6. Thermal infrared analysis. An AFLIF thermal infrared camera (C2, FLIR Systems Inc.) was used to take infrared picture of the bilayer to reflect the temperature change in response to the alternating magnetic fields.

7. Optical microscopy. The cross section of PVDF/ PDMS@FePs bilayer was observed by an optical microscope (ECLIPSE Ci-L) at room temperature, which could reflect the thickness of PVDF and PDMS@FePs layers.

8. Measurement of expansion rate. The neat PVDF film and PDMS@FePs layer were respectively cut to rectangular strips. The expansion rate was measured on thermomechanical analysis instrument (TMA, Q400/Q400EM). A preloading force (0.05 N) was applied to the strip samples, and increased the temperature from 30 to 100 ℃. The expansion ratio of strip was monitored with increasing the temperature while keeping the preloading force constant. All the tests were conducted in the air atmosphere.

9. Mechanical measurement. The single layers of PDMS with increasing content of Fe particles were cut into rectangular strips (30 mm × 5 mm). The strips were clipped on a universal stretching machine and stretched at a speed of 50 mm/min. the stretching process immediately stopped as the strip broke. The strain-stress profiles were then obtained. The Young's modulus values were calculated based on the slope of each profile.

Figures

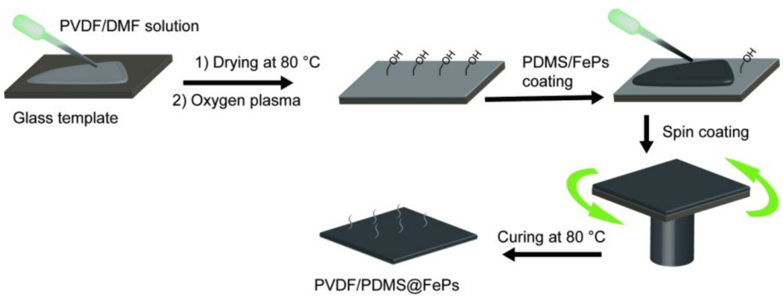

Fig. S1 The manufacturing process of the PVDF/PDMS@FePs bilayer actuators.

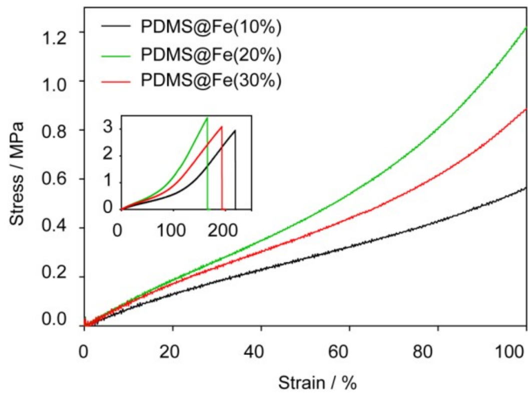

Fig. S2 Mechanical tests of the PDMS@FePs single layer with different FePs content to figure out the FePs content effects on the stiffness of the bilayer.

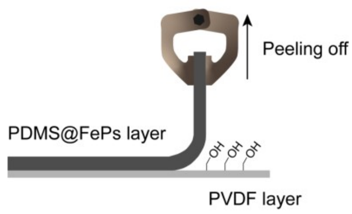

Fig. S3 Sketch map showing how to do a standard 90° peeling test for the bilayer. The result was used to evaluate the interfacial stability during the actuation by magnetocaloric effects.

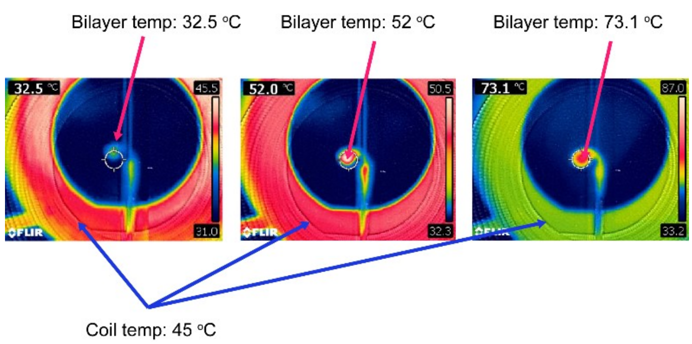

Fig. S4 Upon applying electricity, the temperature of coil would be increased as well. To confirm the temperature of bilayer actuator that came from the magnetocaloric effects of FePs or from the heating coil, a series of real-time thermal images were taken by an infrared thermal imager. After applying electricity, the temperature of coil remained constant at 45 °C, while the temperature of actuator was capable of going from 32.5 to 73.1 °C. This result indicated that the temperature variation of the bilayer actuator was mainly from the magnetocaloric effects of FePs.

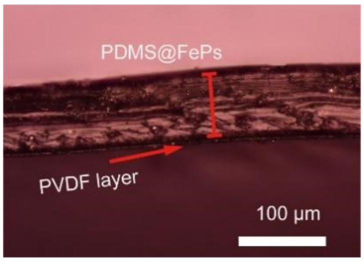

Fig. S5 The microscopic image of the cross section of a bilayer of PVDF/PDMS@FePs.

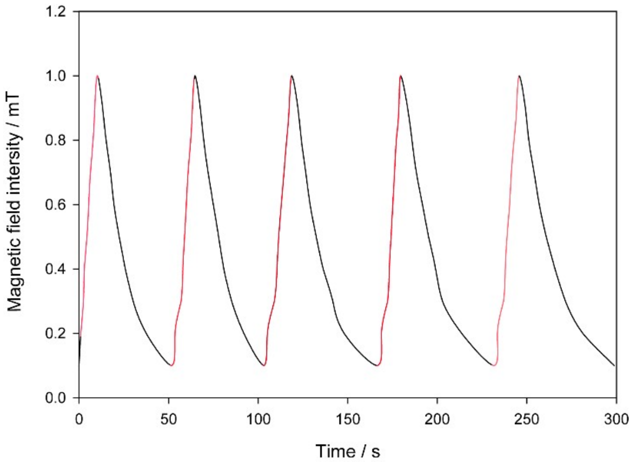

Fig. S6 The magnetic field intensity variation caused by a rectangular sheet iron. As the sheet iron was inserted into the induction coil, the intensity of magnetic field was increased greatly; while it was retracted, the intensity of magnetic field decreased.

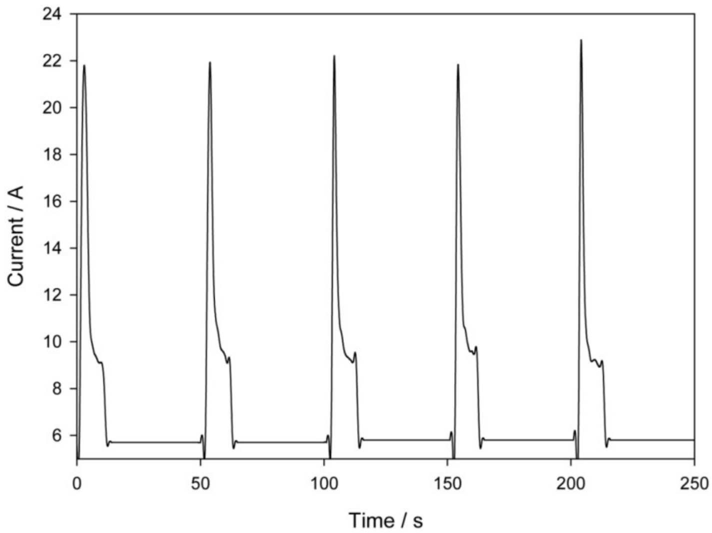

Fig. S7 The profile recording the current change passing through the energized coil at a high power mode (400 W).

Movie Legends

Movie S1. A four-pointed star with structural patterning showing a controllable curling motion driven by magnetocaloric effects. The video was displayed at its five times speed.

Movie S2. A six-pointed star with structural patterning showing a standing motion driven by magnetocaloric effects. The video was displayed at its five times speed.

Movie S3. Reversible actuation of a five-pointed star driven by an intensity-volatile magnetic field. The video was displayed at its five times speed.

Movie S4. Mimicking the movements of human fingers driven by magnetocaloric effects. The video was displayed at its five times speed.

Movie S5. A butterfly-shaped bilayer actuator that mimics the movements of the left/right wings of a butterfly. The video was displayed at its ten times speed.