Electronic Supporting Information

Magnetically Directed Soft Actuators Driven by Moisture

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Experimental section

Chemicals and materials

Chemicals including pentaerythritol ethoxylate, pyrrole, and boron trifluoride diethyl etherate (BFEE) were purchased from Sigma-Aldrich. The iron oxide nanoparticles was purchased from Ocean Nano Tech. BFEE and pyrrole were purified by distillation under reduced pressure. The working electrode was made by depositing a 10-nm Ti layer and then a 25-nm Pt layer on a microscopy glass slide.

Synthesis of PPy films

The PPy films with or without iron oxide nanoparticle loaded were electrochemically synthesized. The PPy film was synthesized through electro-polymerization of pyrrole in a one-compartment cell, with a glass coating Pt as the working electrode, a stainless steel slice (Type 304, 0.5 mm thick) as the counter electrode, and the silver wire with a diameter of 2 mm as the quasi reference electrode. The electrochemical process was controlled by employing a 660E electrochemical workstation (CHI).

The electrolyte solution contained boron trifluoride diethyl etherate (BFEE) and isopropanol at a volume ratio of 3:7, with 5% PEE by volume added. The pyrrole concentration was 0.05 M. Before use, the solution was degassed at 100 kPa on a vacuum rotavap for 5 min.

It took about 50 min to deposit a thin layer of PPy film (~ 8 μm, 25 mm × 20 mm) on the platinum electrode under the constant current density of 0.92 mA cm^{-2}. Then the electrode was washed by isopropanol and air-dried. 100 μL iron oxide nanoparticles in chloroform (~ 20 mg mL^{-1}) was evenly added onto the PPy surface dropwise and slowly air-dried. During the drying process, the iron oxide nanoparticles attached tightly to the PPy matrix. In this manner, a thin layer of iron oxide nanoparticles with known mass was formed on the PPy film. Afterwards, the electrode was immersed into the electrolyte solution for PPy deposition (~ 11 μm) for another 70 min. Subsequent deposition of PPy fully impregnated the iron oxide layer, and the final thickness was 18-20 μm. To investigate the effect of the adding amount of iron oxide nanoparticles, different amount iron oxide nanoparticles (25 μL, 50 μL, 150 μL) were added likewise. With repeated iron oxide nanoparticles loading and PPy deposition steps, thicker PPy film can be prepared. After synthesis, the PPy film was rinsed by isopropanol and peeled off the electrode. Similarly, the PPy film without iron oxide nanoparticle loaded was synthesized by one step deposition at 0.92 mA cm^{-2} for 2h, reaching the thickness of 19 μm.

The mass of PPy films with SPIONs were measured by ultramicro analytical balance

(Sartorius). By calculation, the iron oxide nanoparticles (25 µL, 50 µL, 100 µL, 150 µL) in the PPy film were 3.1%, 6.4%, 12.0%, and 17.8% by mass, respectively.

Characterization of PPy film

The infrared spectrum of PPy films with or without SPIONs loaded were recorded by using Thermo Scientific spectroscopy (OMNIC, Nicolet iS5) at room temperature with a humidity RH = 30%. Olympas Optical Raman Microscope (514 nm laser beam, 50× objective, 0.5 mW power) was used to record the films' Raman spectra. The spectra were recorded by accumulating two times for 60 s each time. PPy film surface's morphology was tested by using FE-SEM (JSM-6700F). The TEM images of the iron oxide nanoparticles was taken on a JEOL JEM-2100F field-emission high-resolution transmission electron microscope operated at 200 kV. The magnetic property of PPy films with SPIONs loaded was measured by Magnetic Property Measurement System (Quantum Design, SQUID VSM) with the scanning magnetic ranging from -3 T to 3 T.

Mechanical test of PPy films

The mechanical properties of PPy films was tested on an Instron 3340 testing machine. The polymer films were cut into long stripes (2 cm long, 0.3 cm wide, 18-20 µm thick). The effective length of the sample between the machine's two clamps were 7 mm and the tensile rate was 1 mm/min. The experiment was carried at 25 °C with a humidity of 30%.

Flipping motion of PPy films

For the measurement of PPy films' flipping motion frequency, films were placed on a piece of water-wetting nonwoven paper. The saturated water vapor pressure was tuned by placing the paper on a heater with adjustable temperature settings. One flipping cycle refers the film flipped from one face to the other face.

Theoretical analysis of the PPy film's water-induced expansion-contraction cycle

We did a theoretical thermodynamic analysis of the film's water-induced expansion-contraction cycle and came up with the following two equations:

$$\frac{E \times d}{R} < \frac{(-\Delta G_{cycle} \times \rho)}{M} \quad (1)$$

$$\frac{E \times d^3}{2R^2} > f_{ad} \quad (2)$$

E, d and R are the elastic modulus, thickness and curvature of the buckled actuator, respectively. ρ and M are the density and molecular weight of water, respectively. ΔG_{cycle} is the molar Gibbs free energy change of absorbed water during one expansion-contraction cycle. f_{ad} is the adhesive force coefficient between PPy films and moist substrates. Given a certain E, R and f_{ad}, equations 1 and 2 roughly define a **theoretical maximum and minimum limit on the required thickness of the actuator to perform**

fast locomotion. See our previous paper (Science, 2013, 339, 186-189) for more details about the theoretical analysis.

Directional control of PPy film

For controlling the motion of PPy films, a NdFeB magnet was placed at a certain distance away from the film. A PPy film without SPIONs and a SPIONs-PPy film with the same size and shape were placed on a 30 °C wet nonwoven paper simultaneously. Driven by the water vapor, both PPy films moved around on the wet paper. While the motion direction of the PPy film without SPIONs was random, the motion direction of the SPIONs-PPy film was toward the NdFeB magnet.

Targeted transportation of cargo by PPy film

For the cargo delivery experiment, an 8.28-mg SPIONs-PPy film (12.0 wt% SPIONs) was used. Copper sheets (~ 41.8 mg, nearly 5 times weight of the PPy film) were attached to corners of the PPy film, as is shown in (Figure S4). In this way, the loaded film's gravity center still lied in film's geometry center. The wet nonwoven paper as substrate was set at 35 °C. The loaded film was placed 5 cm away from the NdFeB magnet.

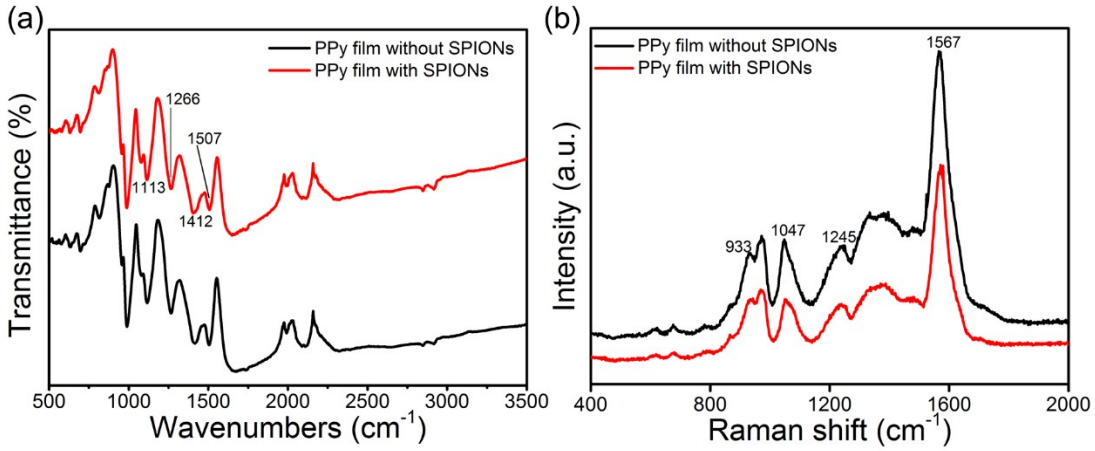

Fig. S1 The FTIR spectra (a) and Raman spectrum (b) of PPy films with or without SPIONs.

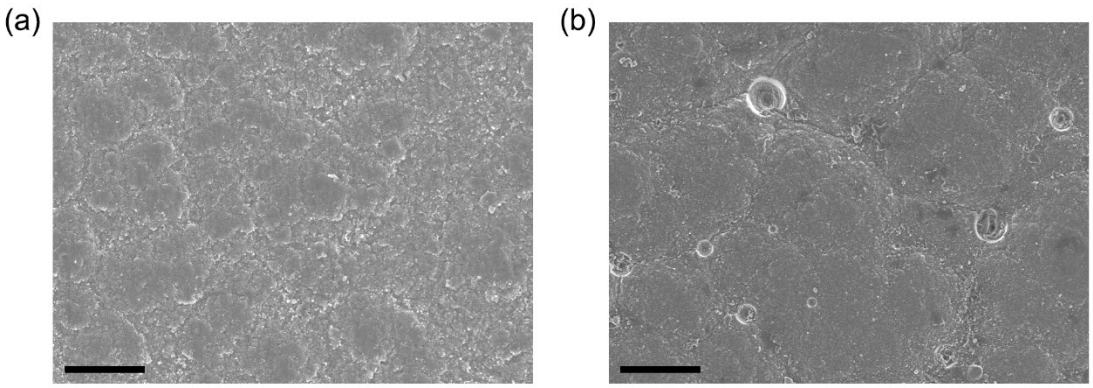

Fig. S2 SEM images of a PPy film without SPIONs (a) and a SPIONs-PPy film (b). Scale

bar: 5 μm.

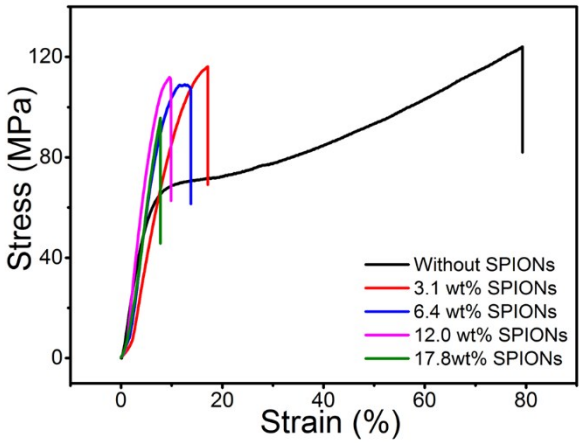

Fig. S3 Stress-strain curves of PPy films with or without SPIONs. The number indicates the weight percentage of SPIONs in the PPy films.

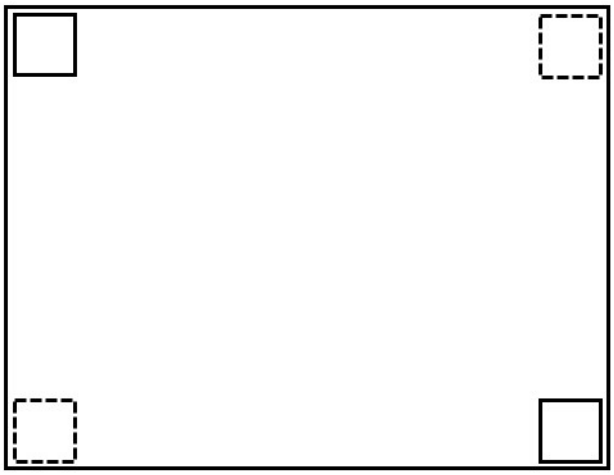

Fig. S4 Pattern of copper sheets attachment to corners of the SPIONs-PPy film. The film is expressed as the rectangular block. Four attached copper sheets are illustrated as small square: two (solid lines) are placed on the top face of the film and the other two (dashed lines) are on the bottom face.

Movie S1. A SPIONs-PPy film (12 wt% SPIONs) is rotating up to 400 rpm driven by a magnetic stirrer.

Movie S2. Directional control of the motion of SPIONs-PPy film (12 wt% SPIONs). The SPIONs-PPy film is moving towards a NdFeB magnet, while the PPy film without SPIONs is moving randomly.

Movie S3. A SPIONs-PPy film (12 wt% SPIONs) remains stationary state on a 30 °C dry substrate 3-cm away from a NdFeB magnet.

Movie S4. A SPIONs-PPy (12 wt% SPIONs) film performs random walk on a 30 °C wet substrate, without NdFeB magnet.

Movie S5. Targeted transportation of 5-time-heavier cargo by a SPIONs-PPy film. Copper sheets (total weight 41.80 mg) were attached to corners of an 8.28-mg SPIONs-PPy film (12.0 wt% SPIONs). The SPIONs-PPy film was placed on a 35 °C wet

substrate. A NdFeB magnet was placed 5 cm away from the film.