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# Supporting Information

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Electrodeposited Superhydrophilic-Superhydrophobic Composites for Untethered Multi-Stimuli-Responsive Soft Millirobots

*Zhiqiang Zheng, Jie Han, Sinan Ozgun Demir, Huaping Wang, Weitao Jiang, Hongzhong Liu and Metin Sitti**

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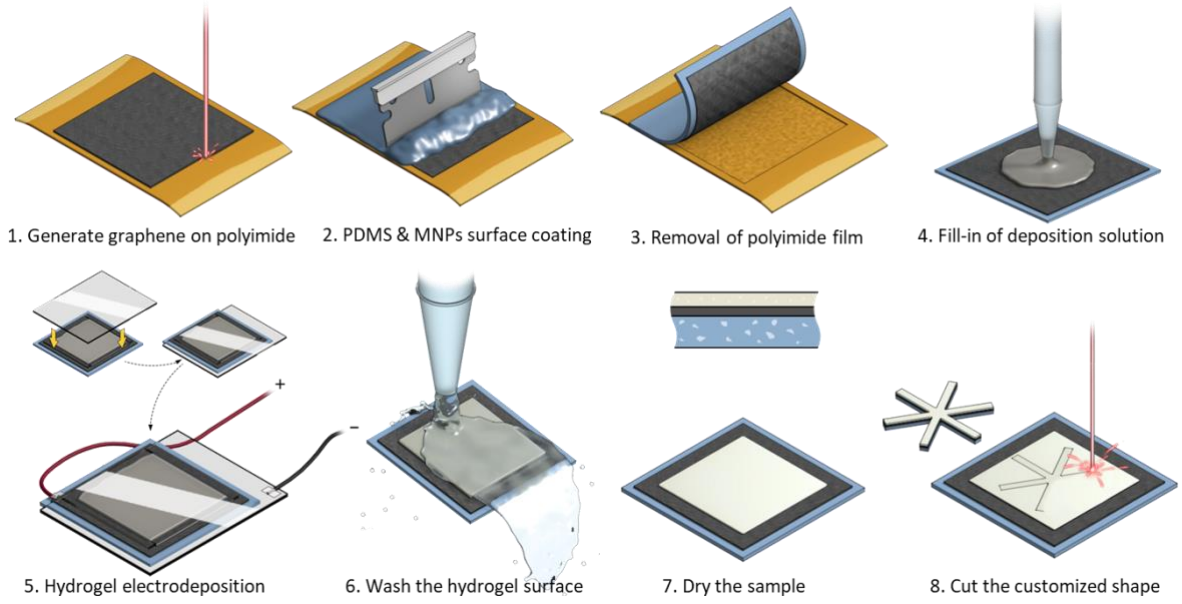


**Figure S1**. Schematics of the MSR fabrication process.

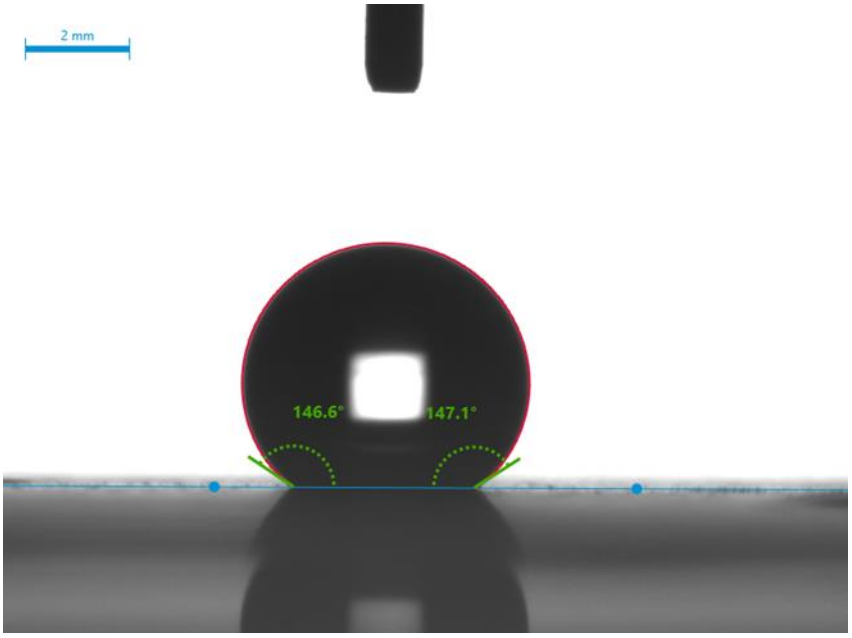


**Figure S2**. Water contact angle of the LIG-coated PDMS. 4 μL water droplet was used to test the contact angle of the surface.

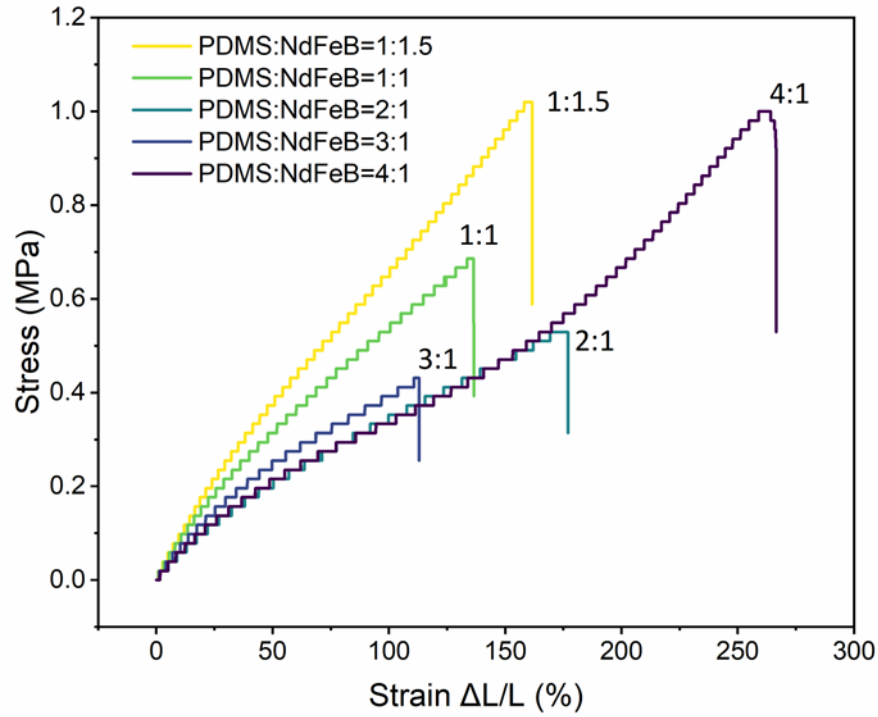


**Figure S3**. Tensile test of the composite stripe structures with PDMS and NdFeB microparticle ratio of 1:1.5, 1:1, 2:1, 3:1 and 4:1.

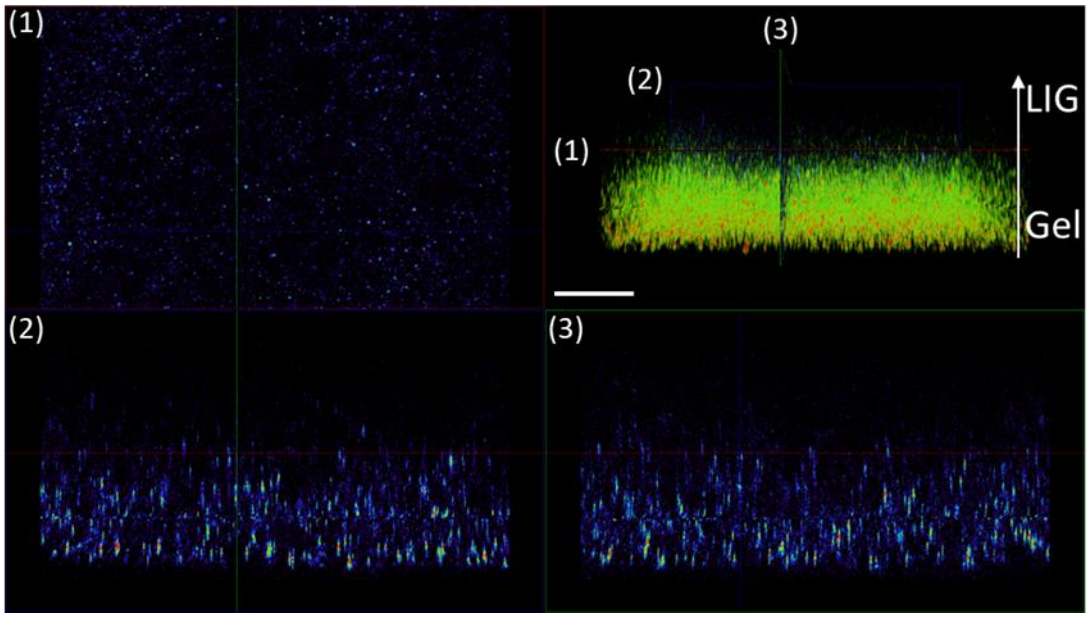


**Figure S4**. The three section views fluorescent images of hydrogel interface. This image is scanned along the surface of hydrogel to the LIG. Scale bar: 100 μm.

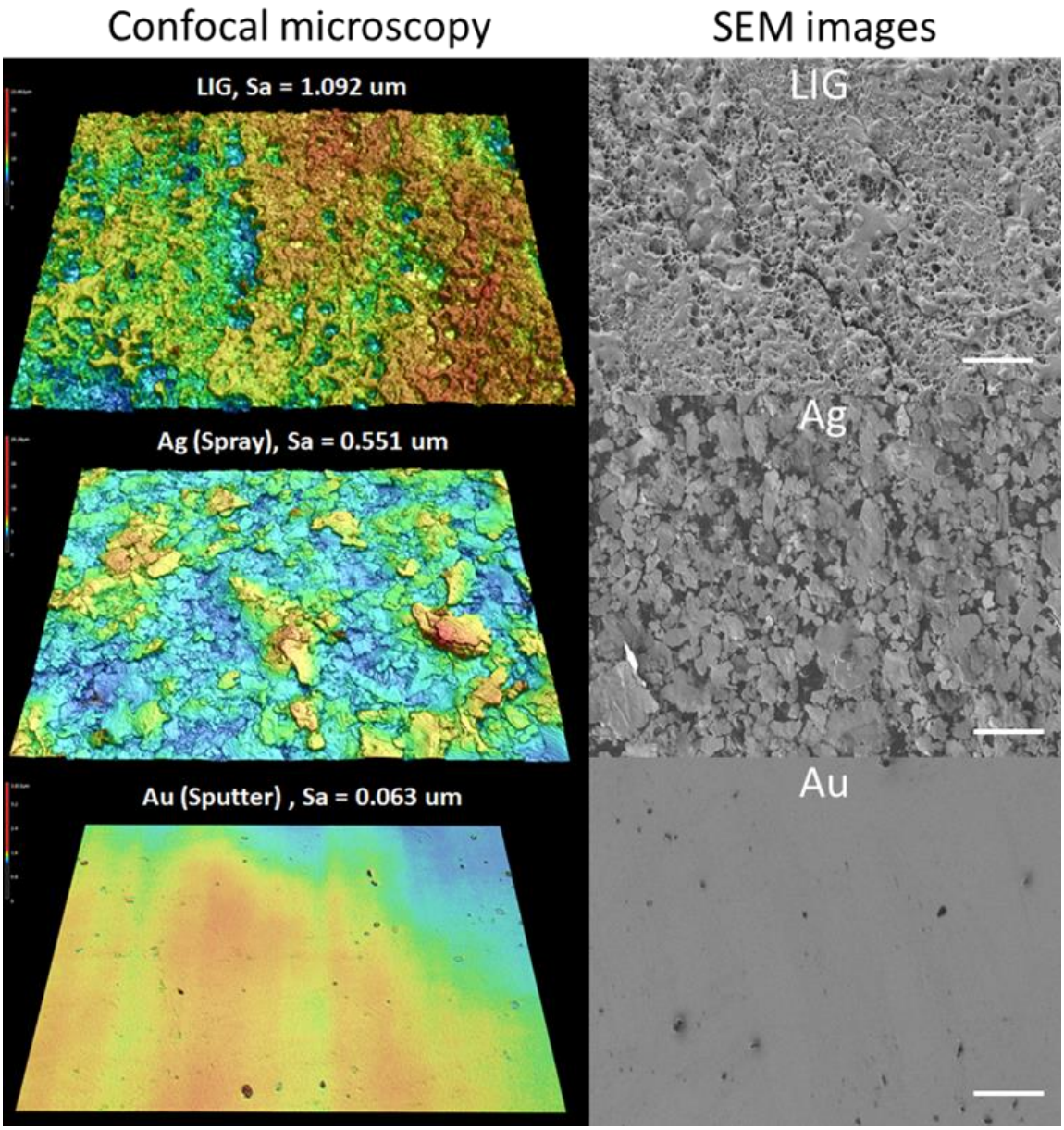


**Figure S5**. The confocal and SEM images of LIG, sprayed silver (Ag), and sputtered gold (Au) surface. Sa represents the arithmetical mean height of the surface. Scale bar: 20 μm.

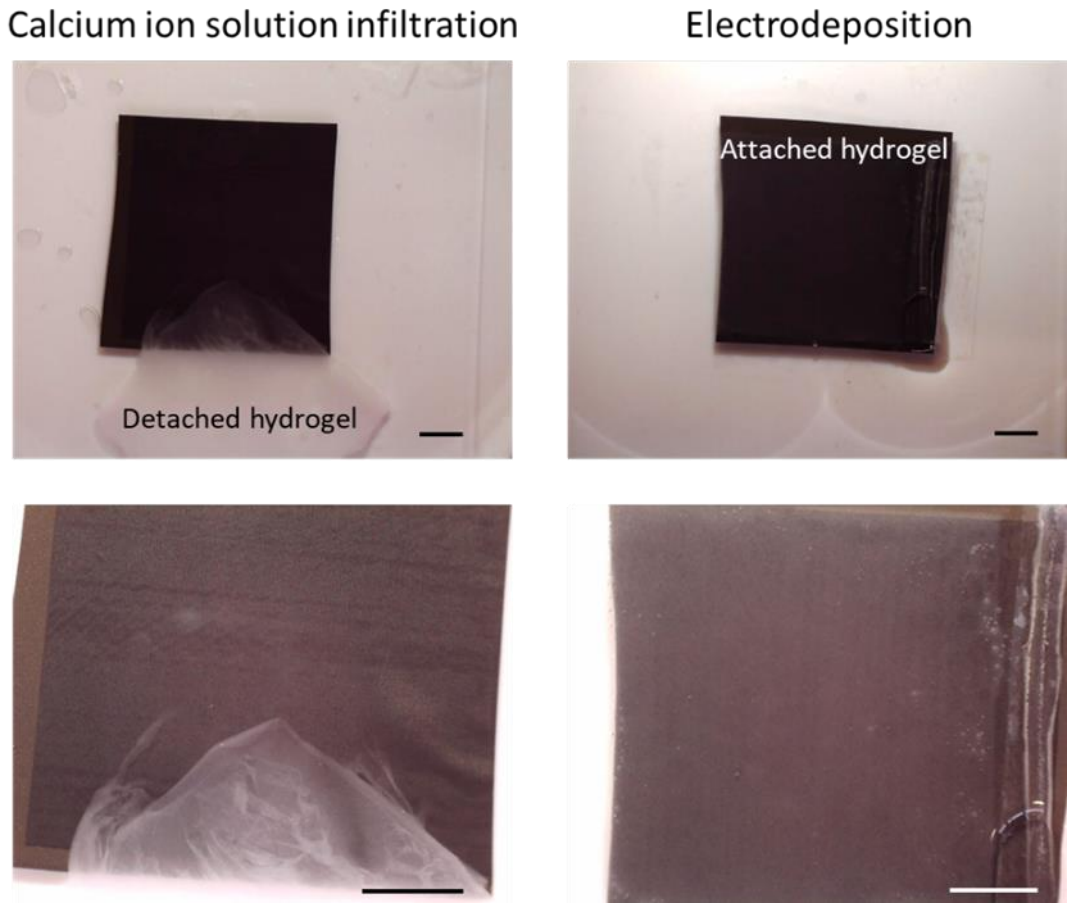


**Figure S6**. Calcium ion solution and electrodeposition gelation result after 1-min washing process. Scale bar: 1 cm.

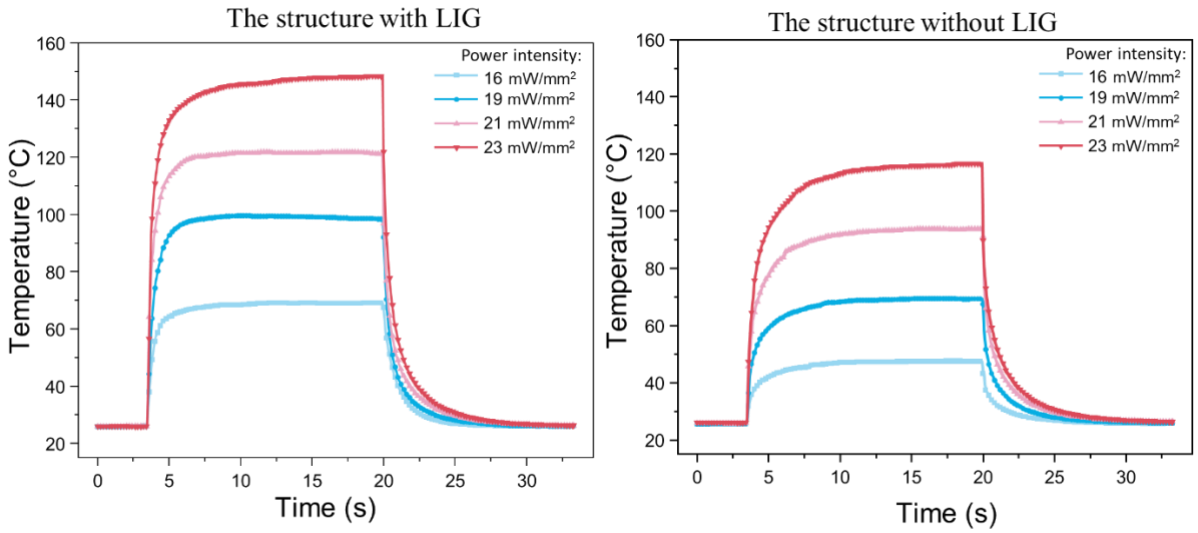


**Figure S7**. Time-dependent temperature as function of the power intensity. Power intensity produced by laser varying from 23 mW/mm$^2$ to 16 mW/mm$^2$ on PDMS film with and without LIG.

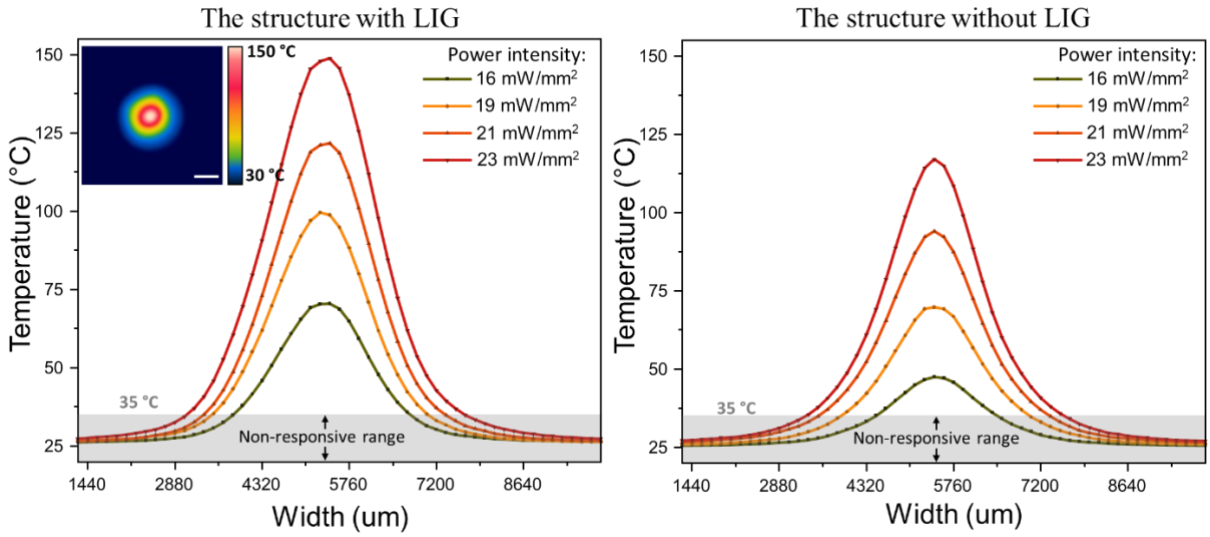


**Figure S8**. Width-dependent temperature as a function of the power intensity. Power intensity produced by laser varying from 23 mW/mm$^2$ to 16 mW/mm$^2$ on PDMS film with and without laser induced graphene.

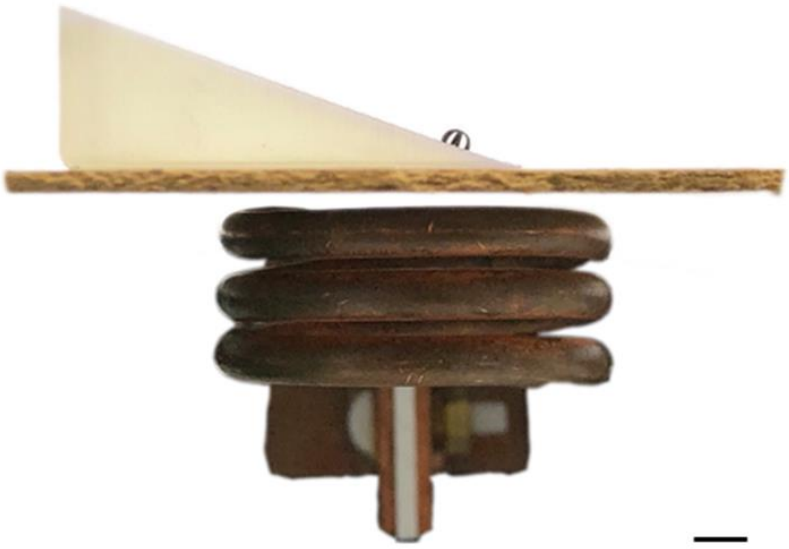

**Figure S9**. Photo of the radio frequncy (RF) magnetic field heating setup. Scale bar: 2 mm. The stage is around 1 mm above the electromagnetic coil.

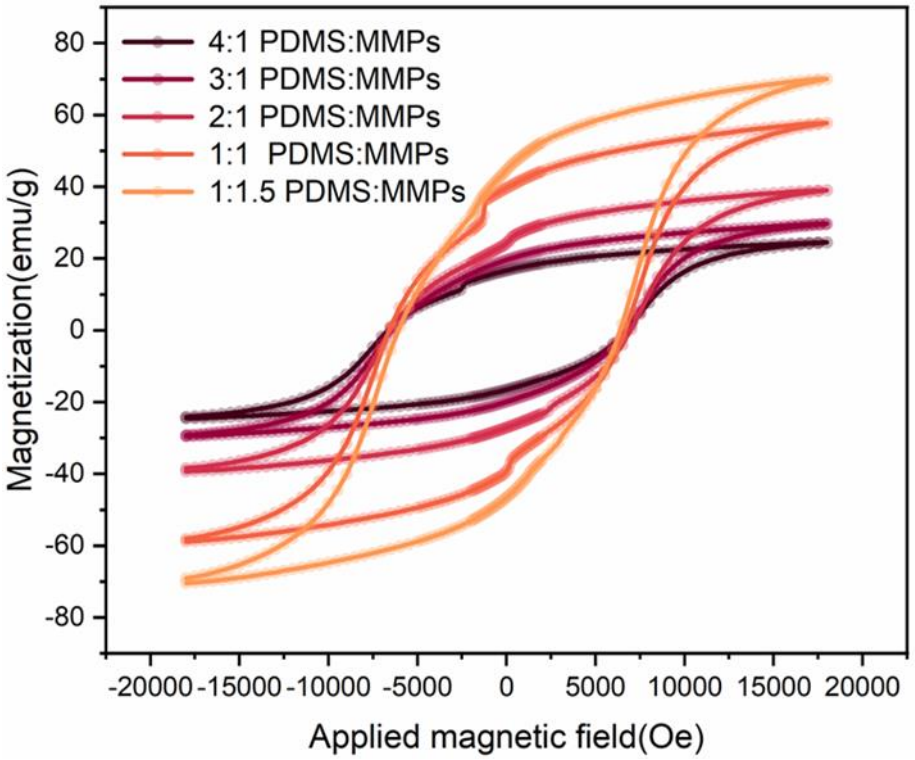


**Figure S10**. Measured magnetic hysteresis curves (four-quadrant BH curves) of the composite materials. The ratios are 4:1, 3:1, 2:1, 1:1, 1:1.5 of PDMS: magnetic microparticles (MMPs) mass ratio.

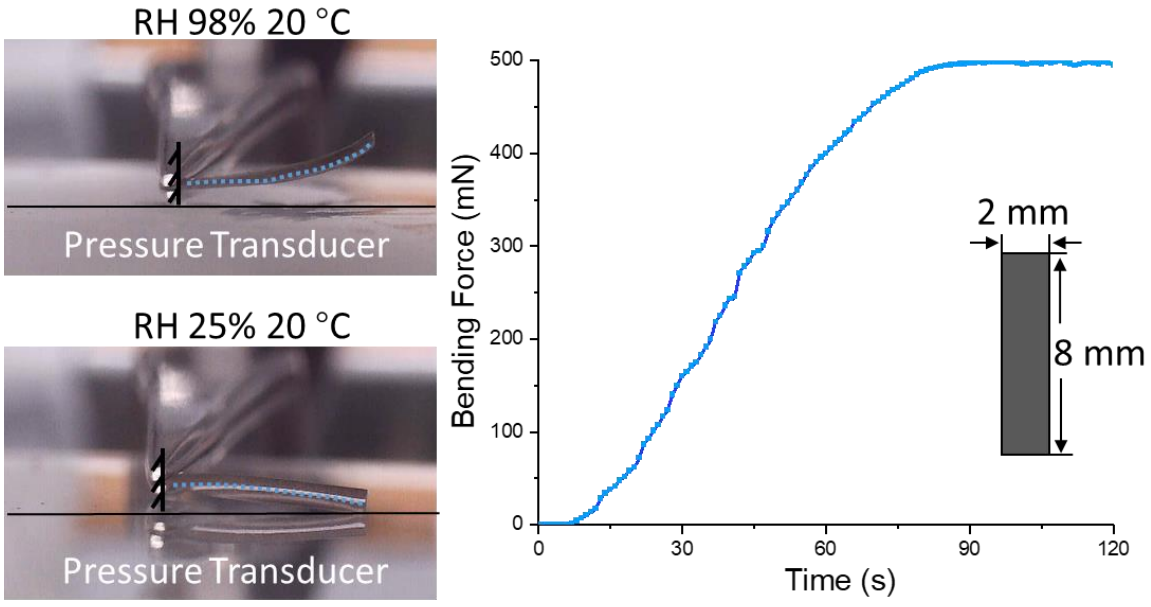


**Figure S11**. Bending force test of the MSR from 98% RH to 25% RH in the room temperature. The size of tested beam structure is 2-mm width and 8-mm length. The maximum bending force is around 500 mN.

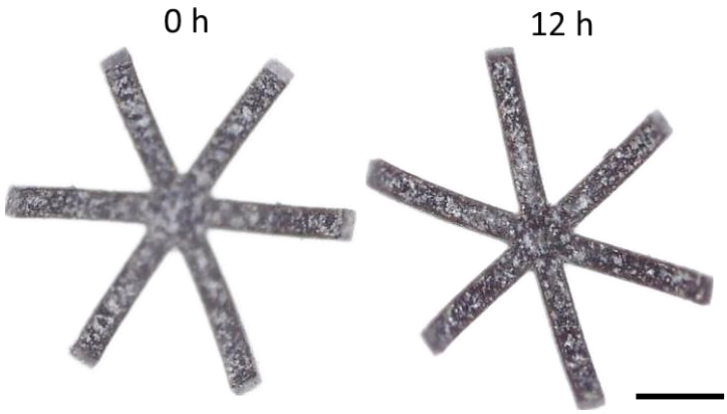


**Figure S12**. Stability test of the MSR in 95% RH for 12 hours. Scale bar: 1 mm.

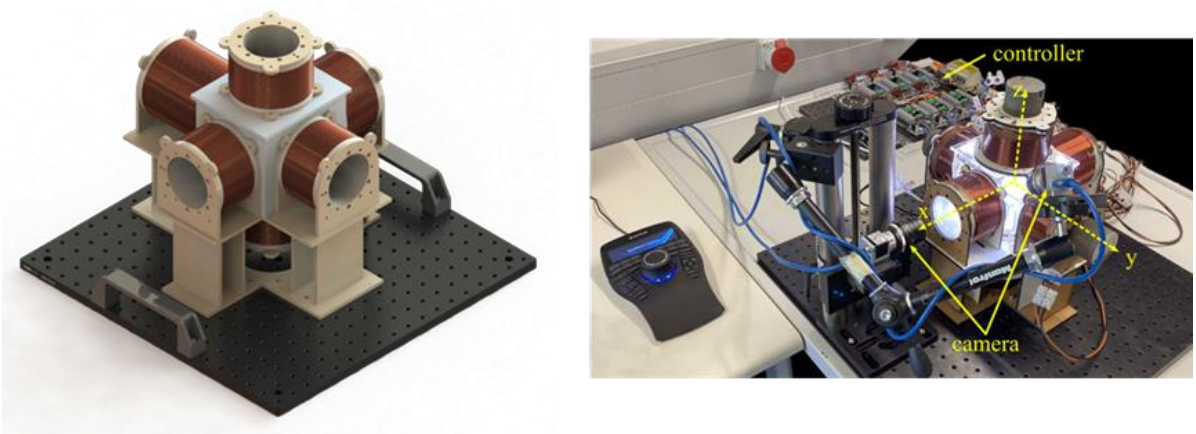


**Figure S13**. The schematic and image of the magnetic coil setup, which is used to apply 3D magnetic fields on the MSRs in the workspace.

## Supplementary Videos

**Video S1.** Confocal microscopic 3D image of fluorescent nano particle loaded hydrogel interface.

**Video S2.** Bending of stripe structure in response to humidity change.

**Video S3.** Shape-morphing and simulation of the stripe structure.

**Video S4.** Hand humidity reaction of the cross structure.

**Video S5.** Shape-morphing of the flower, vine, mimosa and flytrap shaped structure

**Video S6.** Self-propulsion of asymmetric petal-shaped structures.

**Video S7.** MSRs conducting local transformation in response to the laser heating.

**Video S8.** MSRs conducting uniform shape morphing in response to the radio frequency heating.

**Video S9.** MSRs climbing the slope with choppy surface.

**Video S10.** Multimodal motion of snowflake shaped MSR in response to the environmental changes.

**Video S11.** Multimodal motion of stripe shaped MSR in response to the environmental changes.

**Video S12.** Multimodal motion of cross shaped MSR in response to the environmental changes.

**Video S13.** Movement adaptability of MSR in air and liquid.

**Video S14.** Multi-tasking of MSR in response to the environmental changes.