

## Supporting Information:

### Light-driven Untethered Soft Actuators Based on Biomimetic Microstructure Arrays

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Figure S1(a) and Figure S1(c) show the optical images of the fabricated mushroom-shaped micropillars that based on supporting micropillar diameters of  $\sim 35\ \mu\text{m}$  and  $\sim 45\ \mu\text{m}$ , and Figure S1(b) and Figure S1(d) present the corresponding SEM images. Mushroom-shaped microstructures with a supporting micropillar of  $\sim 35\ \mu\text{m}$  in diameter had a height of  $\sim 85\ \mu\text{m}$  and a period of  $\sim 80\ \mu\text{m}$ , and were terminated with circular top plates with a height of  $\sim 3\ \mu\text{m}$  and diameters of  $\sim 44\ \mu\text{m}$  and  $\sim 55\ \mu\text{m}$ . Correspondingly, the mushroom-shaped microstructures with a supporting micropillar of  $\sim 45\ \mu\text{m}$  in diameter had a height of  $\sim 100\ \mu\text{m}$  and a period of  $\sim 100\ \mu\text{m}$ , and were terminated with circular top plates with a height of  $\sim 3\ \mu\text{m}$  and diameters of  $\sim 45\ \mu\text{m}$  and  $\sim 63\ \mu\text{m}$ . The height and period of the micropillars can be adjusted easily by changing the thickness of the resist and the size of the lithography pattern. And the diameter and thickness of the mushroom shaped tips can be changed by modulating the thickness of the PDMS precursor and the inking time. For thinner PDMS precursor layers, the transferred viscous PDMS to micropillar tips was small, and the

corresponding mushroom cap size was thin and small. Specially, when micropillars were fully immersed into the viscous PDMS precursor film, the mushroom caps would connect each other.

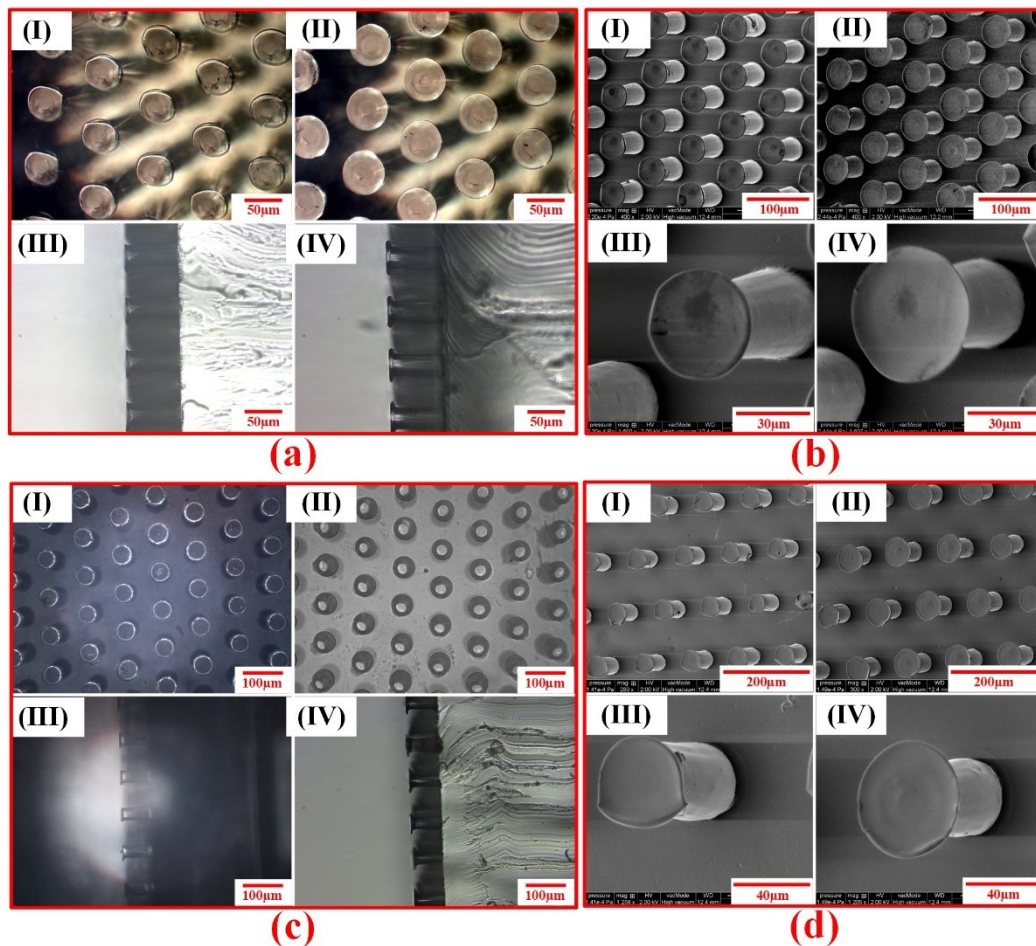


Figure S1. Optical and SEM images of mushroom shaped micropillars based on the supporting micropillar of  $\sim 35 \mu\text{m}$  and  $\sim 45 \mu\text{m}$  in diameter. (a) Optical images of mushroom shaped micropillar with a period of  $\sim 80 \mu\text{m}$  and diameter of  $\sim 35 \mu\text{m}$ : (I) Top view of mushroom terminal with a small size, (II) Top view of mushroom terminal with a big size, (III) and (IV) Side view corresponding to Figure S1(a)I and Figure S1(a)II; (b) SEM images corresponding to optical images in Figure S1(a). (c) Optical images of mushroom shaped micropillar with a period of  $\sim 100 \mu\text{m}$  and diameter of  $\sim 45 \mu\text{m}$ ; (d) SEM images corresponding to optical images in Figure S1(c).

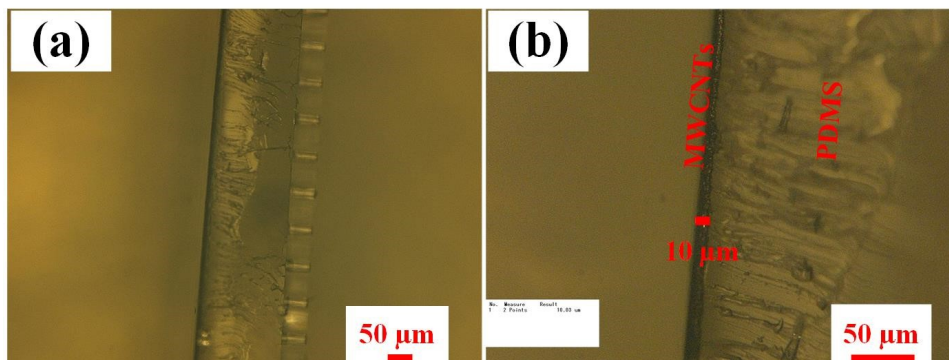


Figure S2. The cross-sectional optical image of PDMS/CNTs bilayer.

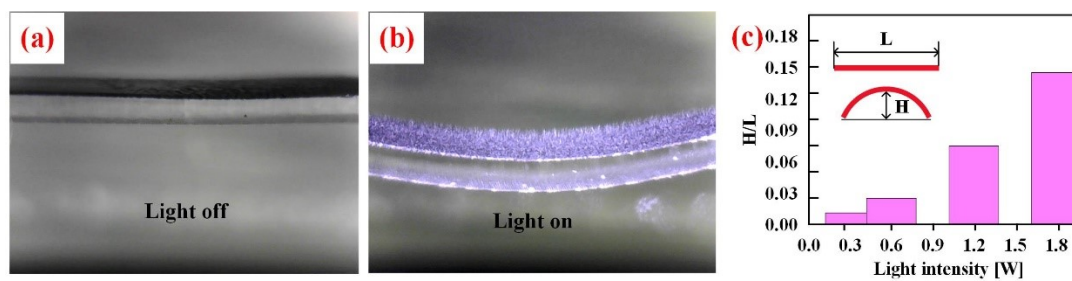


Figure S3. Characterization of the film deformation. (a) Deformation of the film under NIR illumination off state; (b) Deformation of the film under NIR illumination on state; (c) Effect of light intensity on the H/L ratio.