Supporting Information for

NIR-mediated Deformation from a CNT-based Bilayer Hydrogel

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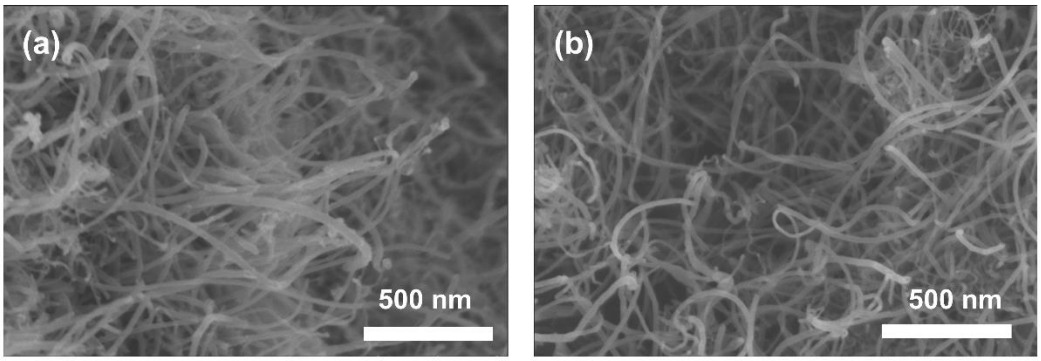

Figure S1. SEM photographs of original (a) and modified (b) CNTs. The scale bar is 500 nm

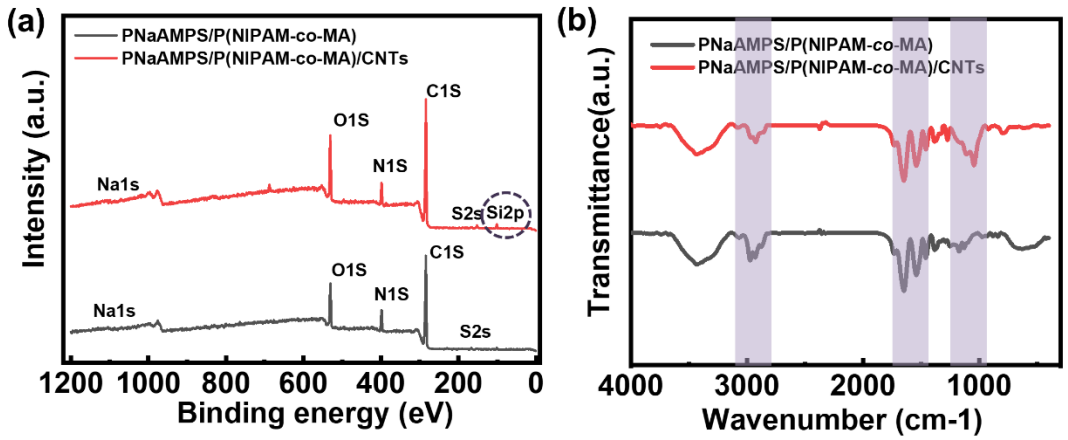

Figure S2. The microstructure characterization of dried PNaAMPS/P(NIPAM-co-MA) and PNaAMPS/P(NIPAM-co-MA)/CNTs hydrogel (CNTs content, 0.2 wt%). (a) XPS spectroscopy, (b) FTIR spectroscopy

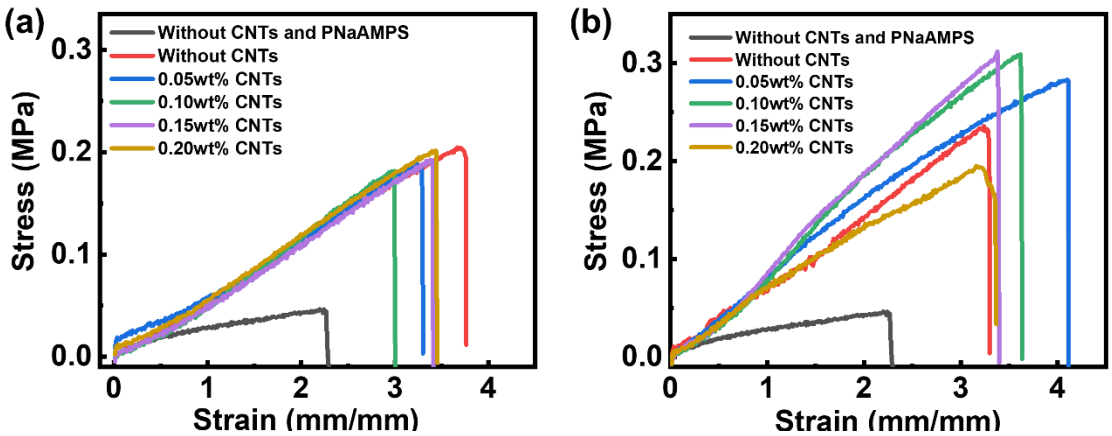

Figure S3. Tensile Stress-Strain curves of hydrogels(P(NIPAM-co-MA) hydrogel, PNaAMPS/P(NIPAM-co-MA) hydrogel and PNaAMPS/P(NIPAM-co-MA)/CNTs hydrogels) (a) hydrogels added with original CNTs (b) hydrogels added with modified CNTs

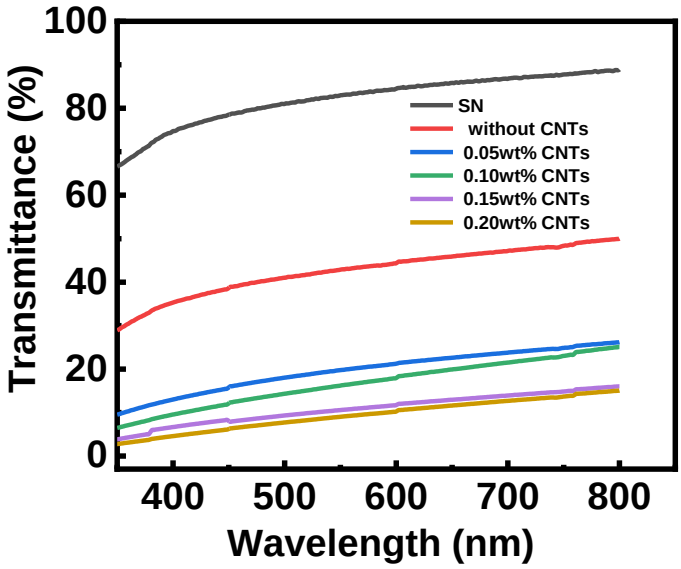

Figure S4. Transmittance curve of all hydrogels(PNaAMPS/P(NIPAM-co-MA), PNaAMPS/P(NIPAM-co-MA)/CNTs and SN: P(NIPAM-co-MA))

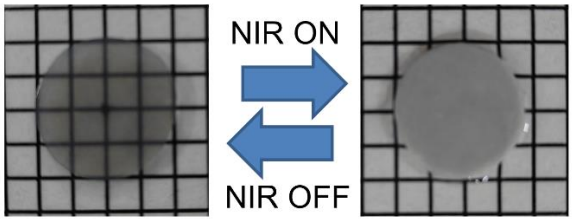

Figure S5. Optical photos of the phase transition behavior of PNaAMPS/P(NIPAM-co-MA)/CNTs hydrogels(CNTs content：0.20 wt%) under near-infrared light irradiation

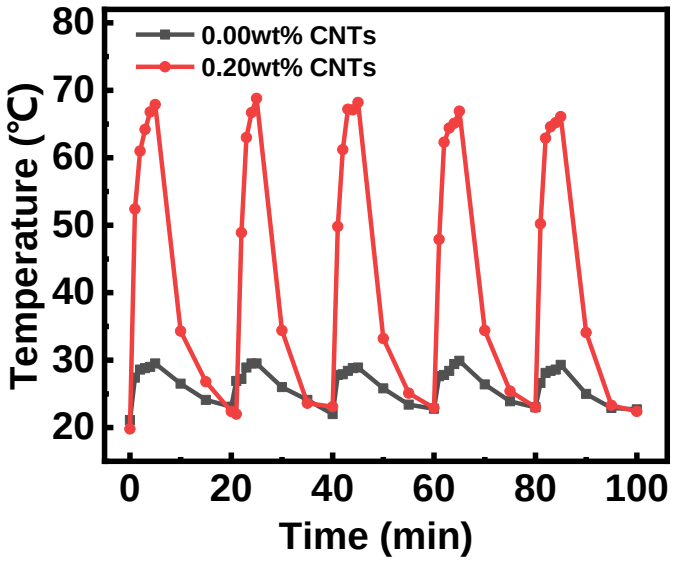

Figure S6. Cyclic reversible photothermal conversion behavior of PNaAMPS/P(NIPAM-co-MA)/CNTs hydrogels(Control group: PNaAMPS/P(NIPAM-co-MA) hydrogel)

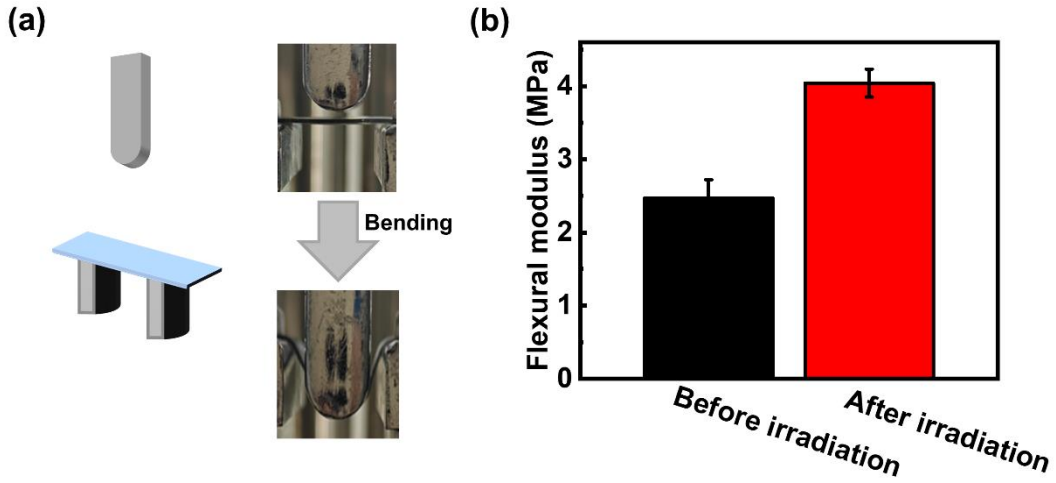

Figure S7. Three-point bending test of PNaAMPS/P(NIPAM-co-MA)/CNTs hydrogels (CNTs content, 0.2 wt%) before and after NIR irradiation. (a) Schematic diagram and optical images for three-point bending test; (b) Flexural modulus

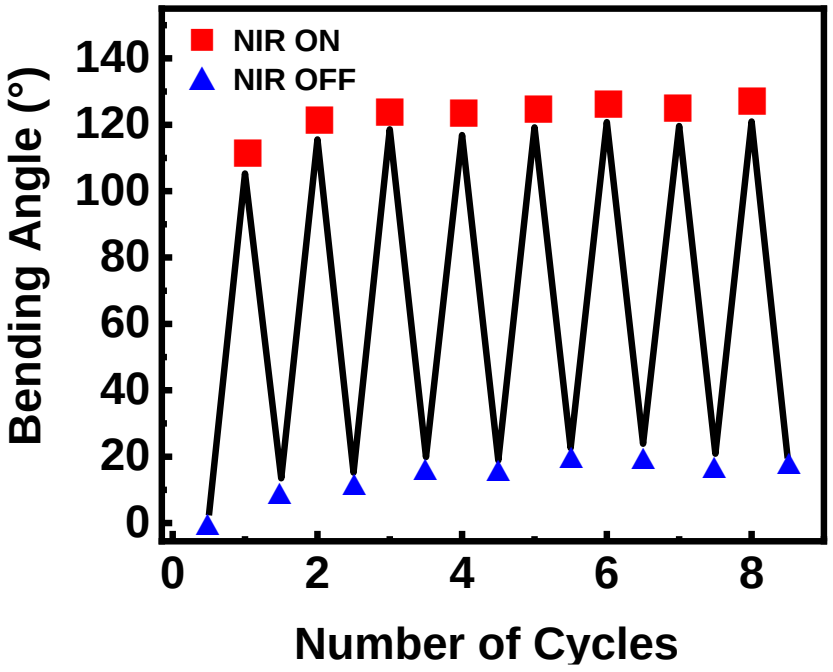

Figure S8. Cyclic reversible bending behavior of bilayer hydrogel(active layer: PNaAMPS/P(NIPAM-co-MA)/CNTs, negative layer: PNaAMPS/P(NIPAM-co-MA)) under infrared light irradiation

Table S1 The elements ratio of original and modified CNTs.

Elements	C	O	Si
Ratio(Original)	92.69%	7.21%	0.00%
Ratio(Modified)	86.24%	9.32%	4.44%

Table S2 Overall performance comparison of PNIPAM-based hydrogel actuators

Material	Structure	Stimuli-response type	Mechanic property	Deformation
This work	Bilayer	Temperature/light	Strengthen	3D
PNIPAM, Gelatin, PTCA, PAM[1]	Bilayer	Temperature/pH	No strengthen	3D
PNIPAM, PEDOT: PSS, Mxene[2]	Bilayer	Mechanic/light	No strengthen	3D
PNIPAM, rGO, ATPE, PLMA, 6APA[3]	Gradient	Temperature/light	No strengthen	3D
PNIPAM, Spiropyran[4]	Bilayer	Light	No strengthen	3D
PNIPAM, HEA, XLG[5]	Gradient	Temperature	No strengthen	2D
PNIPAM, Silicon Rubber[6]	Multilayer	Temperature	Strengthen	3D
PNIPAM, PAM, Nanothylakoid[7]	Bilayer	Temperature/light	No strengthen	3D

Reference of Table S2

[1] Wu, B. Y.; Le, X. X.; Jian, Y. K.; Lu, W.; Yang, Z. Y.; Zheng, Z. K.; Théato, P.; Zhang, J. W.; Zhang, A.; Chen, T. PH and Thermo Dual-Responsive Fluorescent Hydrogel Actuator. *Macromolecular Rapid Communications* **2018**, *40* (4), 1800648.

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