Supporting information for

NIR responsive and conductive PNIPAM/PANI nanocomposite hydrogels with high stretchability for self-sensing actuators

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1. Synthesis of F127DA

F127DA was synthesized according to the references [1, 2]. Pluronic F127 (20 g) was dissolved in 150 mL dichloro-methane under stirring with nitrogen protection. Triethylamine (3.22 g) was then added to the mixture, followed by placing the reaction in an ice bath. Acryloyl chloride (2.6 mL) was slowly diluted with 50 mL dichloromethane and added dropwise into the mixture over 30 minutes. After 2 days of reaction in the ice bath, the mixed solution was filtered and precipitated with petroleum ether and finally dried in vacuum at room temperature. The product was characterized by ^{1}H NMR spectrum (AVANCE III 400 MHz, Bruker, Switzerland). As seen from the spectrum, the peak of 1.21 ppm is attributed to the proton of -CH_3 at the PPO unit, and the peaks of 5.8-6.4 ppm are attributed to the proton of -CH=CH_2 in the acryl unit. The acrylation degree of F127DA is calculated by the ratio of acryl protons of F127DA (-CH=CH_2, 5.8–6.4 ppm) to methyl protons in poly(propylene oxide) groups (–CH_3, 1.1 ppm), which is more than 90%.

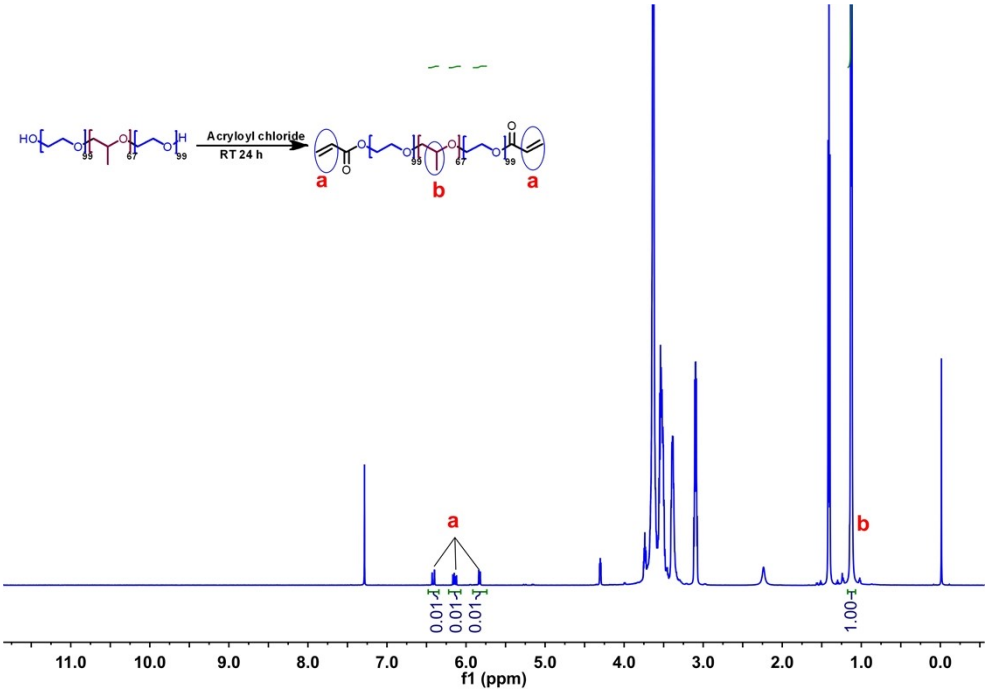

Figure S1. ^{1}H NMR spectrum of the F172DA.

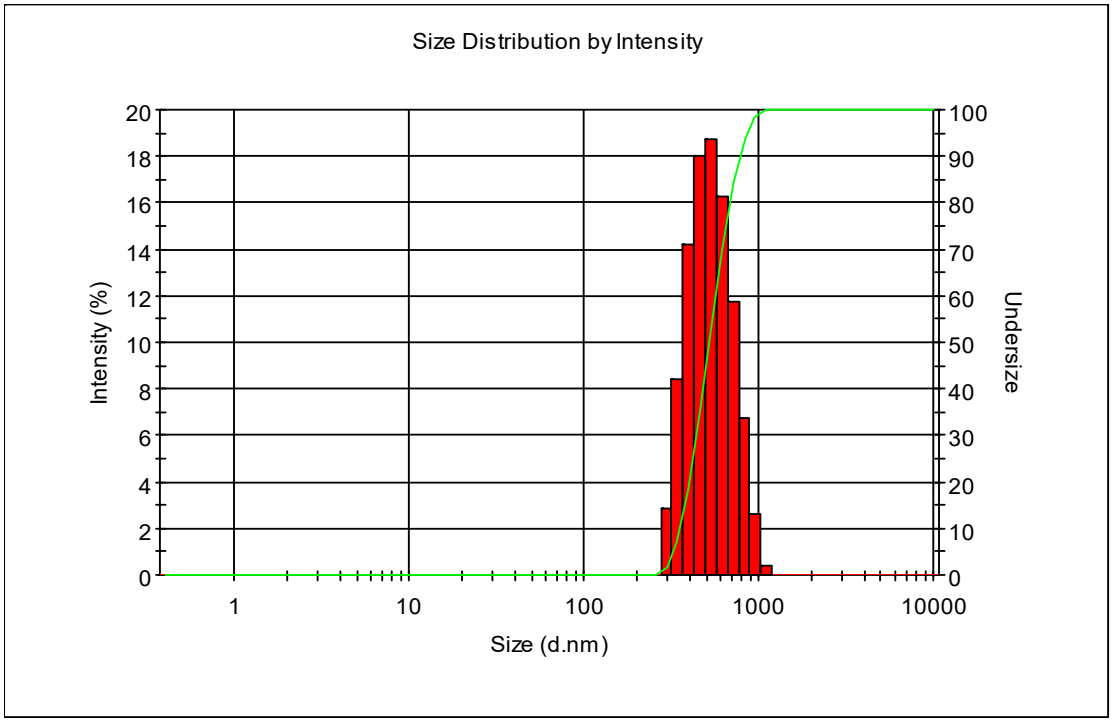

Figure S2. Hydrodynamic radii distributions of F127DA micelles in water solution (15 mg/mL), indicating the F127DA micelles was of 400~500 nm.

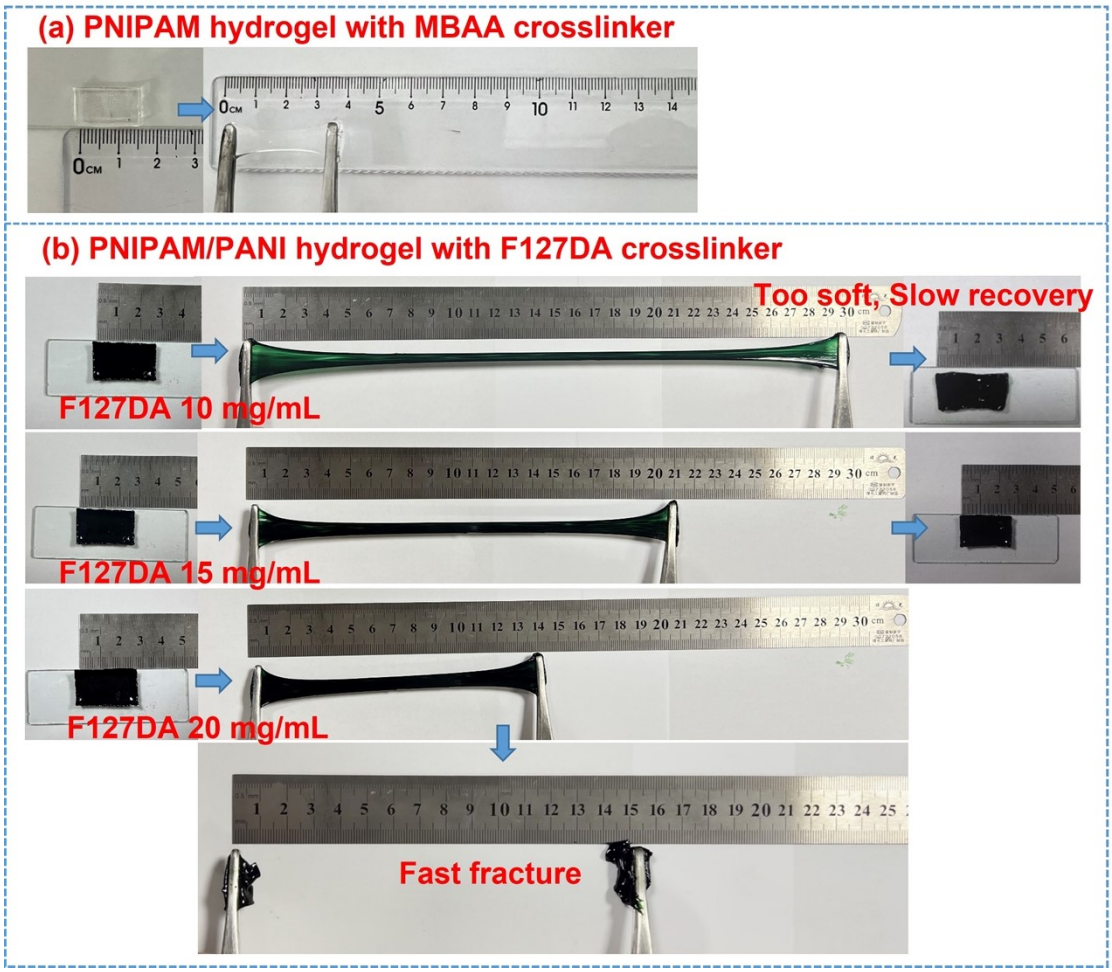

Figure S3. Demonstration of the flexibility and stretchability of the PNIPAM hydrogel with conventional chemically crosslinker *N*,*N*'-methylenebis(2-propenamide) (MBAA) (a) and the PNIPAM/PANI hydrogels with different concentration of F127DA crosslinker (b).

The PNIPAM hydrogel with the conventional MBAA crosslinker was too weak and fragile to stretch up. In contrast, The PNIPAM/PANI hydrogel with the F127DA crosslinker was extensible. With 10 mg/mL F127DA, the PNIPAM/PANI hydrogel can be stretched from the original 3 cm to 30 cm. However, it showed slow recovery speed and was unable to return to its original state after removing the stress. The sample with 15 mg/mL F127DA can be stretched from the original 3 cm to 21 cm. And it can quickly

recover after removing the stress. When the F127DA concentration was increased to 20 mg/mL, the hydrogel could only be stretched to only 14 cm and got vulnerable to break. Considering the real application, the sample with F127DA concentration of 15 mg/mL was chosen in the actuation performance.

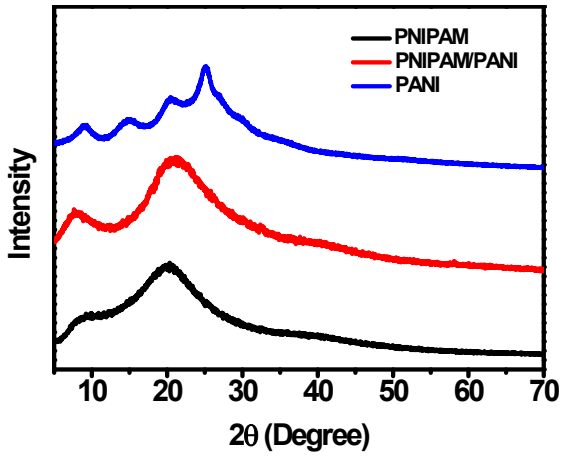

Figure S4. The XRD pattern of the pure PANI, PNIPAM and the PNIPAM/PANI hydrogel after freeze drying.

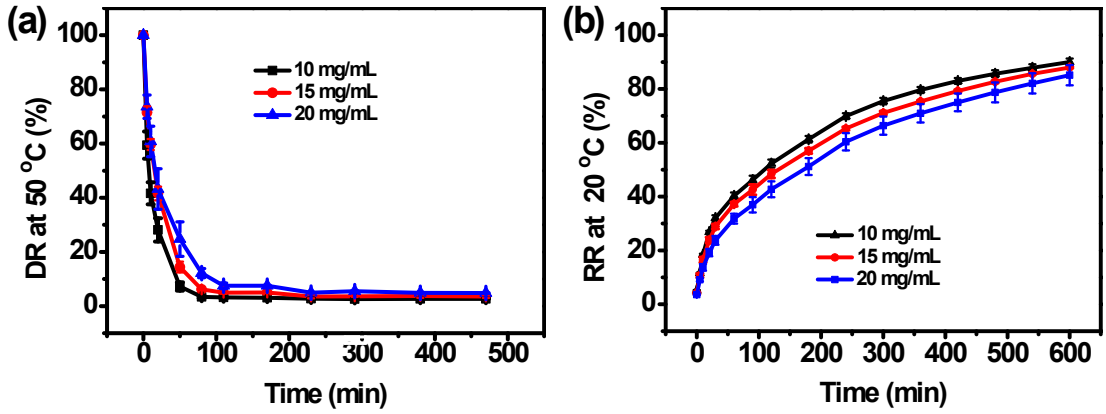

Figure S5. (a) Deswelling kinetics at 50 °C and (b) reswelling kinetics at 20 °C for the PNIPAM/PANI hydrogel with various F127DA concentration.

The deswelling and reswelling process was presented in **Figure S4**. Speciffically, in the deswelling process, a total swollen sample at 20 °C was immersed in to 50 °C hot

water, its weight alteration was recorded at a pre-determined time interval, The deswelling ratio (DR) is calculated by equation: DR (%) = $(W_{t50} - W_d)/(W_0-W_d) \times 100\%$, where W_{t50} is the weight of the sample at the testing time at 50 °C, and W_0 the weight of the total swollen sample at 20 °C. Afterwards, the sample was reswelled again in 20 °C water. The reswelling ratio (RR) is calculated by equation: RR (%) = $(W_{t20} - W_d)/(W_0-W_d) \times 100\%$, where W_{t20} is the weight of the reswelling sample at a specific time at 20 °C, and other terms are the same as defined above. The results in **Figure S5a** indicates the water retention drops quickly in the first 60 min, indicating the majorty percentage of the absorbed water was eliminated. The highest amount of released water was approximately 98% reaching the equilibrium conditions after 480 min. During the reswelling in 20 °C water, all samples took ~400 min to reach an 80% reswelling equilibrium, which indicates the fast dewatering but slow recovery nature.

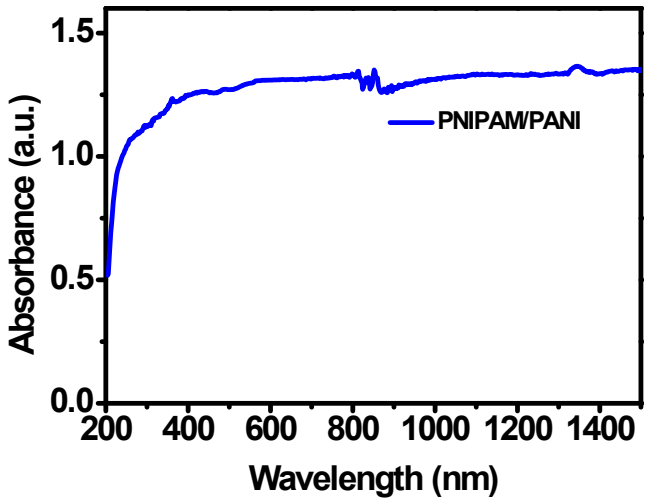

Figure S6. The UV-Vis-NIR spectrum of the PNIPAM/PANI hydrogel.

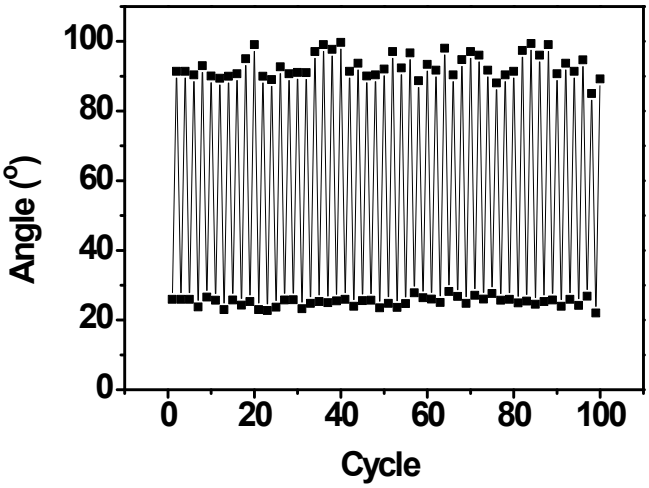

Figure S7. Bending/recovery cycle test of 100 times of the composite hydrogel actuator with NIR irradiation (2 W).

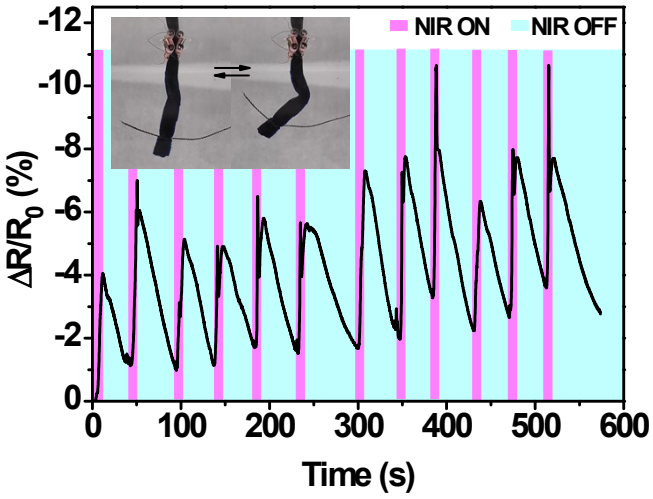

Figure S8. The reversibility of the relative resistance changes of the hydrogel actuator as a function of irradiation time of 808 nm NIR light (2W).

Table S1. Comparative response rate of the PANI/PNIPAM hydrogel the previously described PNIPAM-based hydrogels.

samples	NIR power/power density	Response rate (°C/min)	Containing metal	Ref.
Poly(NIPAM-co-β-CD)/CNT	1.25 W/cm^2	6.5	No	[1]
Graphene/PNIPAM	4.0 W/cm^2	7.25	No	[3]
GO/PNIPAM	0.58 W/cm^2	8	No	[4]
WS_2/PNIPAM	1.0 W/cm^2	12	Yes	[5]
MoO_2/Laponite/PNIPAM	0.8 W/cm^2	17.5	Yes	[6]
$Ti_3C_2T_x$/PNIPAM	0.8 W	2.5	Yes	[7]
CPPY/CPDA/PNIPAM	1.5 W	4.76	No	[8]
PNIPAM/PANI	2.0 W	12.64	No	This work

Table S2. Comparison of our work with other reported flexible hydrogel-based strain sensors.

Sample	GF value	Sensing range (%)	Response time (s)	Ref.
PNIPAM/PANI/	4.83	500	0.15	This work
F-PNIPAAm/PANI	3.92	120	0.4	[9]
PSS/UPy/PANI	3.4	300	-	[10]
P(AAm-co-HEMA)/PANI	1.48	300	0.2	[11]
HPAAm/CS-c-MWCNT	3.2	500	0.18	[12]
PAM/carrageenan/PANI	6	400	-	[13]
PAAm-oxCNTs	3.39	700	0.30	[14]
HP(AAm/AA)-CS-Fe^{3+}	3.621	500	-	[15]

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