Supporting information

Mussel-Inspired PAM-PDA/Ga^{3+} Hydrogels with Antibacterial, Adhesive and Self-healable Properties for Wearable Strain Sensors

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Experimental

1. Chemicals and Materials

Dopamine hydrochloride (DA, 98%), acrylamide (AM, 99%) and gallium nitrate hydrate ($Ga(NO_3)_3$, 99%) were purchased from Shanghai Macklin Biochemical Co. Ltd. APS (98.0%) was purchased from Sinopharm Chemical Reagent Co. Ltd. N, N′-methylenebis(acrylamide) (MBAA, 98%) and N, N, N′, N′-tetramethylethylenediamine (TEMED, 99%) were obtained from Shanghai Zhongqin Chemical Reagent Co. Ltd. 1,1-Diphenyl-2-picrylhydrazyl (DPPH, 98%) was purchased from Beijing Soleibao Technology Co. Ltd. All reagents were used as received without further purification.

2. Preparation of PAM-PDA/Ga^{3+} hydrogels

Initially, dopamine hydrochloride (DA, 0.0128 g), ammonium persulfate (APS, 0.16 g), and a specified amount of gallium nitrate hydrate ($Ga(NO_3)_3$) were dissolved

in 8 mL PBS aqueous solution (pH=11). The mixture was stirred for 30 min to form a stable polydopamine/gallium (III) nanoparticles (PDA/Ga^{3+} NPs) solution. Subsequently, acrylamide (AM, 1.6 g), N, N'-methylenebis(acrylamide) (MBAA, 0.01 g), and N,N,N',N'-tetramethylethylenediamine (TEMED, 50 μL) were added to the above solution under continuous stirring for 10 min. Finally, the precursor solution was transferred into a polypropylene mold and allowed to polymerize at room temperature for 4 h to obtain PAM-PDA/Ga^{3+} hydrogel.

3. Characterizations

The morphology of the samples was characterized using an Ultra Plus field emission scanning electron microscope (FE-SEM, Zeiss, Germany), and the elemental composition was analyzed by energy-dispersive X-ray spectroscopy (EDS). Chemical composition was evaluated via attenuated total reflection Fourier-transform infrared spectroscopy (ATR-FTIR) on an FTS-3000 spectrometer (Digilab, USA). UV-visible (UV-vis) absorption spectra were recorded using a UV-8453 spectrophotometer (Agilent, USA). The particle size distribution and zeta potential of PDA solution were determined using a Zetasizer Nano ZS (Malvern Panalytical, UK).

4. Mechanical Property Measurement

The test samples were fabricated into rectangular specimens with dimensions of 70 mm × 10 mm × 2 mm (length × width × thickness). Uniaxial tensile tests were performed using an electronic universal testing machine (EZ-Test, SHIMADZU) equipped with a 500 N load cell at a crosshead speed of 100 mm/min. The Young's modulus was determined from the linear elastic region (5-15% strain) of the stress-strain curves. The toughness was calculated by integrating the area under the stress-strain curve.

5. Adhesion Property Test

The adhesion properties were evaluated using an electronic universal testing machine (AGS-X, SHIMADZU) equipped with a 100 N load cell. Shear adhesion tests

were conducted at a crosshead speed of 50 mm/min. For testing, the PAM-PDA/Ga^{3+} hydrogel was first sandwiched between various substrates including polyethylene (PE), polyurethane (PU), glass, titanium (Ti) and wood with the contact area of 1 cm × 1 cm. Different weights were then applied to the overlapping region for varying durations to ensure proper contact. Finally, the bonded specimens were subjected to shear testing to measure the adhesion strength between the hydrogel and substrates.

6. Self-healing Performance Test

Hydrogel specimens with the size of 70 mm × 10 mm × 2 mm were first bisected with a clean incision. The cut surfaces were carefully aligned and allowed to heal for predetermined time. Mechanical and electrical recovery properties were then characterized using an electronic universal testing machine (AGS-X, Shimadzu) equipped with a 500 N load in conjunction with a digital source meter (2450, Keithley).

7. Antibacterial Property Test

The antibacterial properties were evaluated using both colony counting and SEM observation methods. For bacterial preparation, E. coli and Staphylococcus aureus were cultured in Luria-Bertani (LB) medium at 37°C until reaching logarithmic growth phase, then diluted to 1×10^6 CFU/mL with sterile PBS. Sterilized PAM-PDA/Ga^{3+} hydrogel samples (300 μL) were placed in 48-well plates at 37°C for 6 h. Next, the incubated hydrogel sample was gently washed three times with sterilized deionized water to remove loosely bound bacteria. Then, Surface-adhered bacteria were dislodged by ultrasonication in 1.5 mL LB medium to obtain the bacterial suspension and 60 μL suspension was inoculated on the LB agar plate along with incubating again for 16 h to count the colony forming units. Finally, bacterial inhibition efficiency was calculated based on colony forming unit (CFU) counts. Bacterial suspensions (1×10^8 CFU/mL, 300 μL) were co-cultured with samples at 37°C for 6 h. Samples were washed three times with PBS to remove loosely attached bacteria. Bacterial fixation was performed using 1 mL glutaraldehyde solution (2.5%) for 4 h. Samples were dehydrated through an ethanol gradient series (30%, 50%, 70%, 90%, 100%). Morphological changes were

observed by scanning electron microscopy (SEM).

8. Sensing Property Test

The resistance signals of PAM-PDA/Ga^{3+} hydrogel were characterized using a benchtop digital source meter (2450, Keithley, China). Real-time resistance measurements were recorded during mechanical deformation and human motion detection. The relative resistance change ($\Delta R/R_0$) was calculated according to the formula: $\Delta R/R=(R-R_0)/R_0$, where R_0 and R is respectively the initial and strain resistance. The conductivity of the PAM-PDA/Ga^{3+} hydrogel was calculated using the formula: $\sigma=L/RS$, where σ is the conductivity of the PAM-PDA/Ga^{3+} hydrogel, R is the resistance value, L is the length, and S is the cross-sectional area. The gauge factor (GF), defined as the ratio of the relative change in electrical resistance ($\Delta R/R_0$) to the applied mechanical strain (ε), i.e., GF = $(\Delta R/R_0)/\varepsilon$. To evaluate the strain-sensing performance of the PAM-PDA/Ga^{3+}-1 hydrogel, we calculated its GF values across different strain stages by measuring the $\Delta R/R_0$ under varied applied strain.

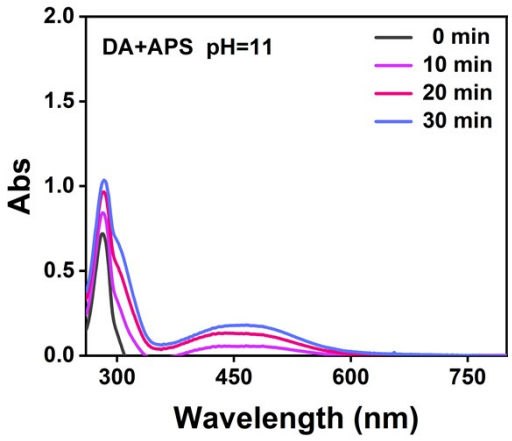


Figure S1. The UV-vis adsorption spectrum of the mixture solution of DA and APS for different times.

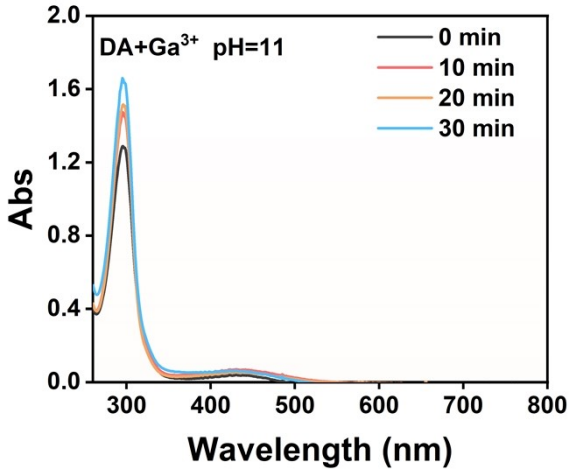


Figure S2. The UV-vis adsorption spectrum of the mixture solution of DA and $Ga(NO_3)_3$ for different times.

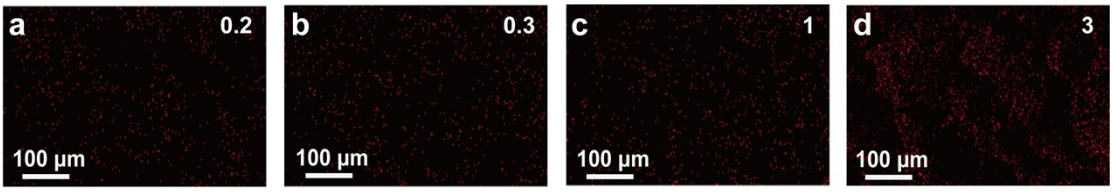


Figure S3. The energy dispersive spectroscopy mapping of Ga^{3+} ions.

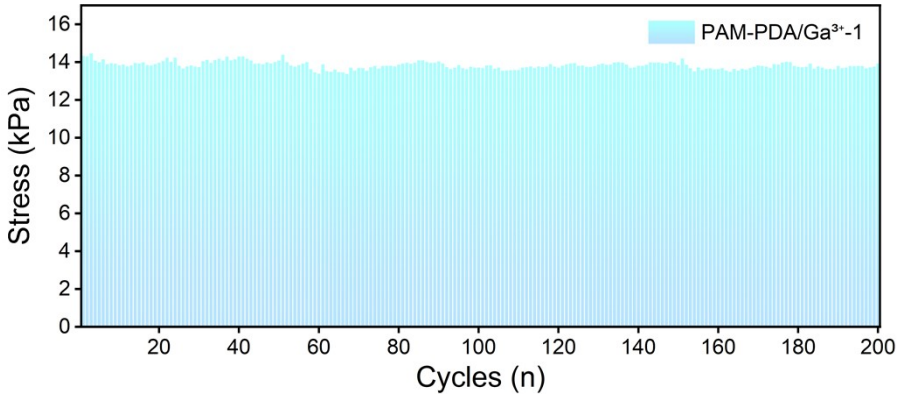


Figure S4. The stress changes for PAM-PDA/Ga^{3+}-1 hydrogel subjected to 200 tensile cycles at 60% strain.

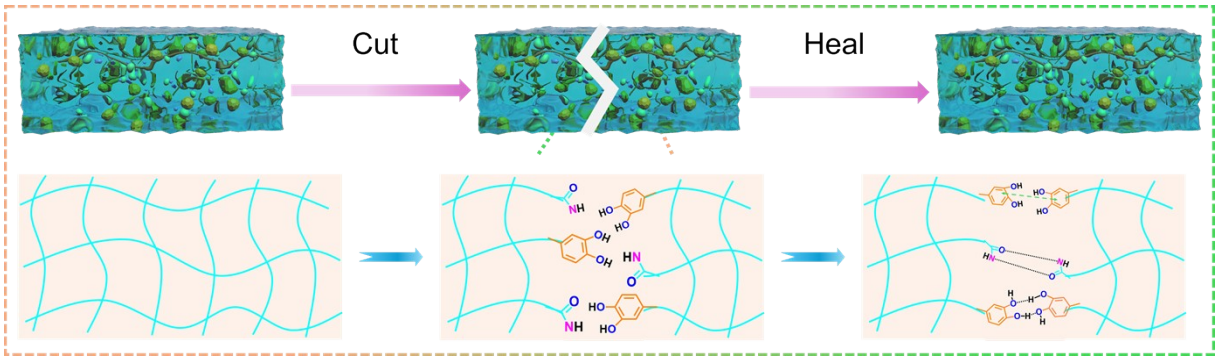


Figure S5. Schematic illustration of the proposed self-healing mechanism of PAM-PDA/Ga^{3+}-1 hydrogel.

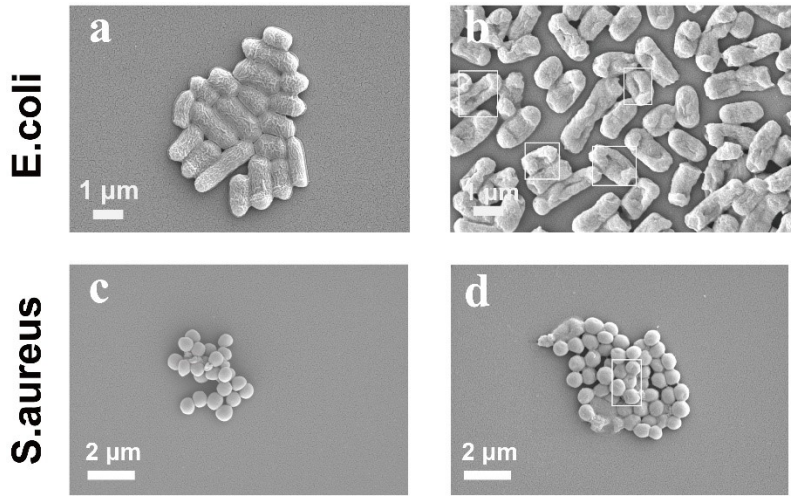


Figure S6. Scanning electron microscopy (SEM) images showing morphological changes of E. coli cultured with PAM-PDA hydrogel (a) and PAM-PDA/Ga^{3+}-1 hydrogel (b). Corresponding SEM images of S. aureus cultured with PAM-PDA hydrogel (c) and PAM-PDA/Ga^{3+}-1 hydrogel (d).

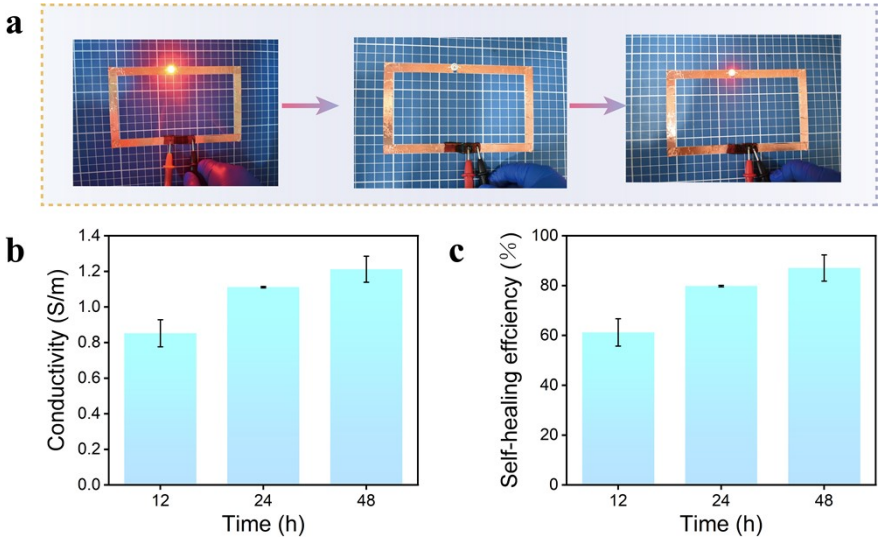


Figure S7. (a) The demonstration of electrical/conductive self-healing performance. (b) Conductivity of PAM-PDA/Ga^{3+}-1 hydrogel after different healing times. (c) Quantitative evaluation of self-healing efficiency.

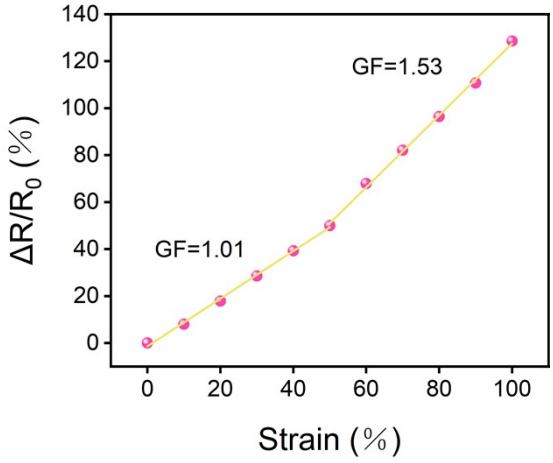


Figure S8. Changes in relative resistance of PAM-PDA/Ga^{3+}-1 hydrogel at different strain stages.

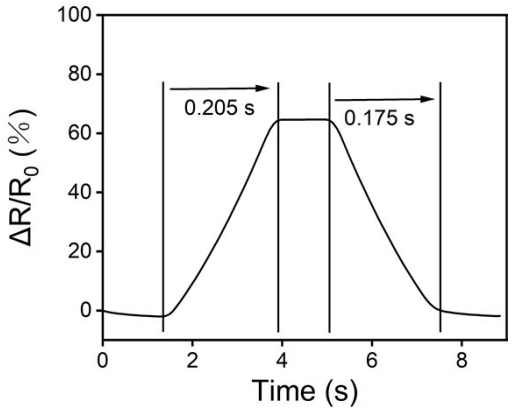


Figure S9. Response and recovery times of PAM-PDA/Ga^{3+}-1 hydrogel.

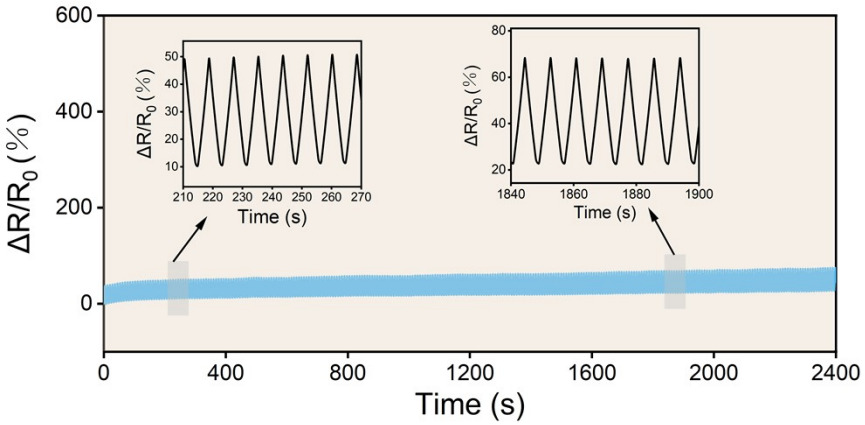
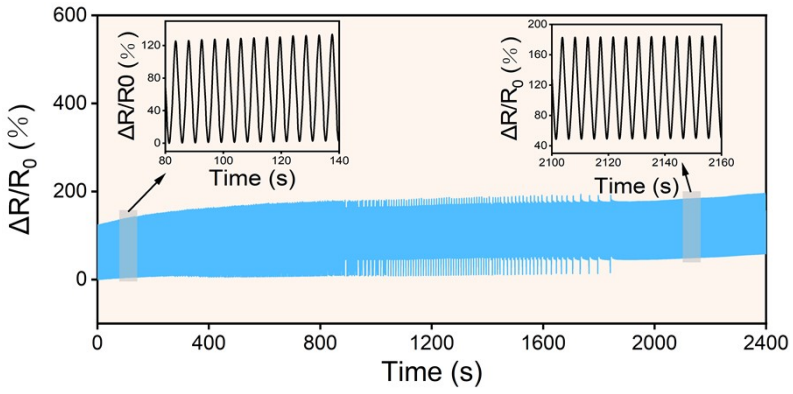


Figure S10. The cyclic stability test indicates the relative change in resistance after 300 tensile cycles at 50% strain (temperature: 25 °C, humidity: 75%).



FigureS11. The cyclic stability test indicates the relative change in resistance after 300 tensile cycles at 50% strain (temperature: 30 °C, humidity: 30%).

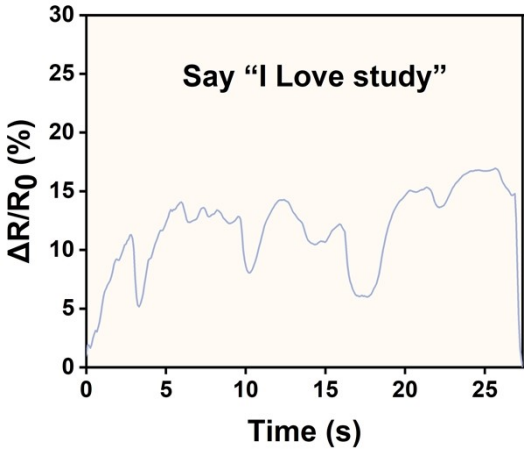


Figure S12. Changes of relative resistance of saying "I love study" when adhering PAM-PDA/Ga^{3+}-1 hydrogel on the throat.

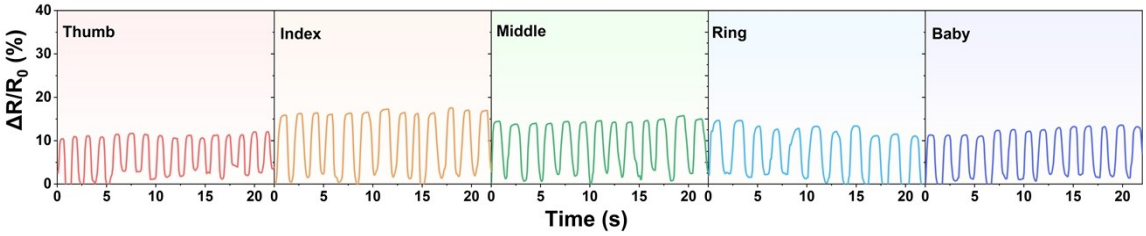


Figure S13. The relative resistance changes of PAM-PDA/Ga^{3+}-1 hydrogel adhered on five fingers upon holding on apple.