Supporting Information

Coordination Mode Regulated Lanthanide Supramolecular Hydrogel with Tunable Luminescence and Stimuli-Responsive Properties

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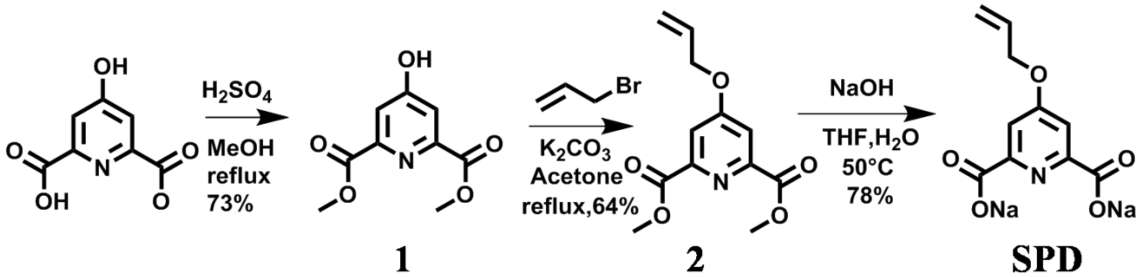
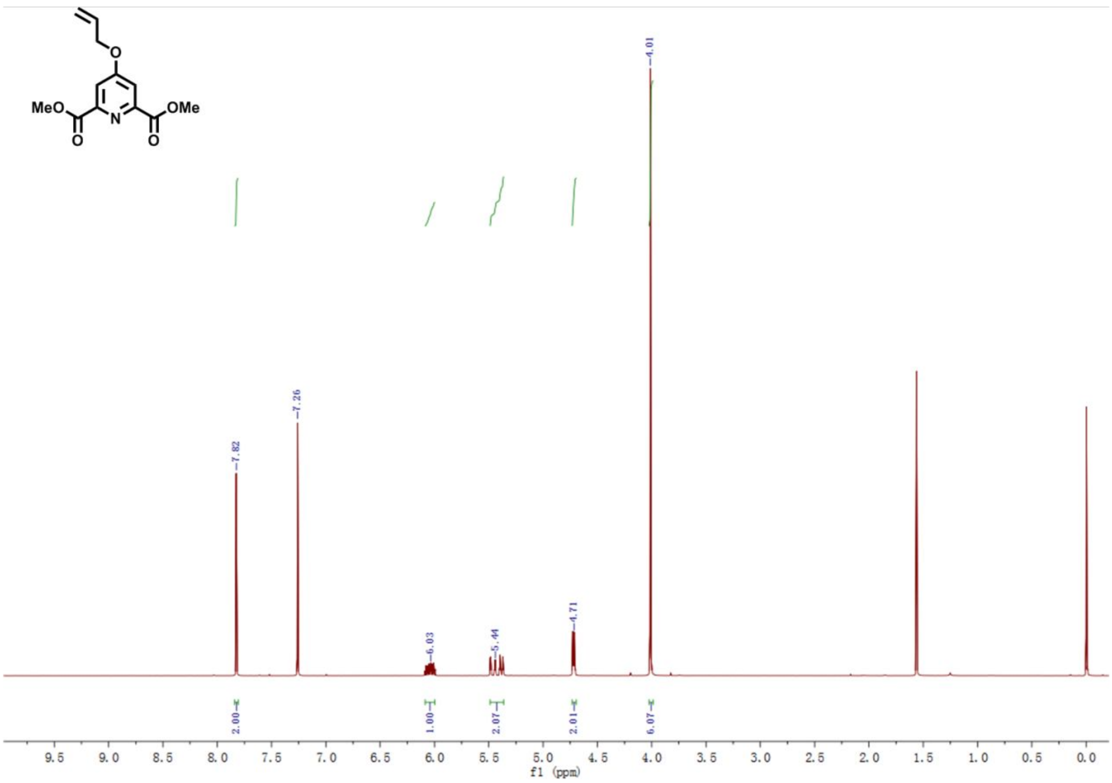
Scheme S1. Synthetic route to compound **SPD**.



Synthesis of compound SPD: Compound **2** (2.2 g, 8.7 mmol) was dissolved in 20 mL THF, then 10 mL aqueous solution of NaOH (1.6 g, 40 mmol) was added. The reaction mixture was stirred at 50 °C for 4 h. The obtained white precipitate was collected by Suction and washed with cold water for 2 times to afford the desired product in 78% yield. ^{1}H NMR (400 MHz, D_2O) δ 7.52 (s, 2H), 6.07 (qd, 1H), 5.39 (dd, 2H), 4.70 (d, 2H). ^{13}C NMR(400MHz, D_2O): δ172.47,166.33, 154.63, 131.84, 118.89, 111.33, 69.15, ppm. HR-ESI-MS: [M-Na]$^-$ calcd for $C_{10}H_7NNaO_5$, 244.0222; found: 244.0198; [M-2Na+H]$^-$ calcd for $C_{10}H_7NO_5$, 222.0402; found: 222.0381.



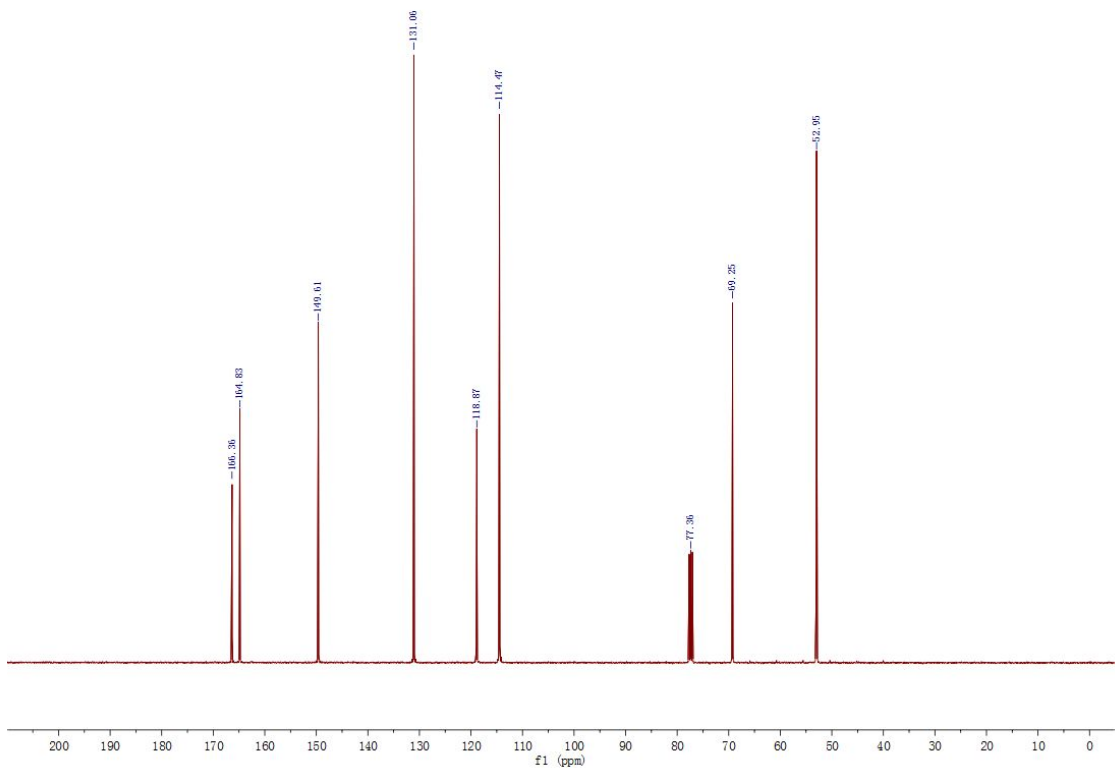


Figure S1. ^{1}H NMR spectra (400MHz, $CDCl_3$) and ^{13}C NMR spectra (100MHz, $CDCl_3$) of compound **2**.

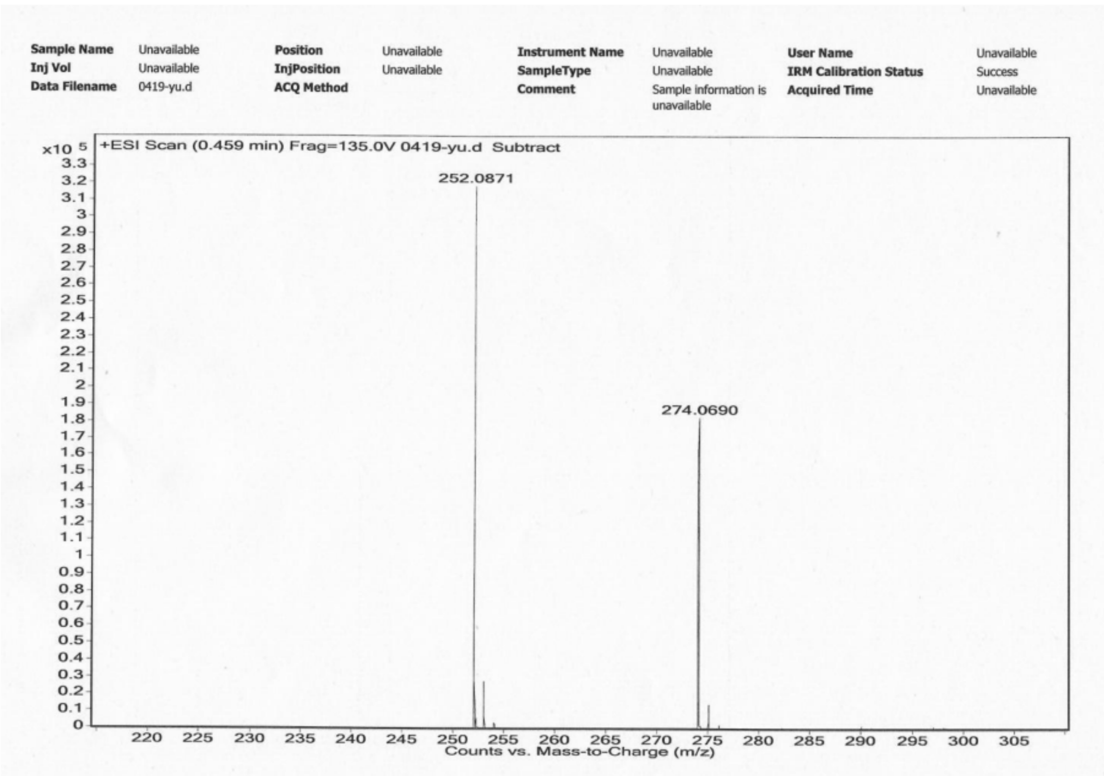


Figure S2.HR-ESI-MS spectra of compound **2**.

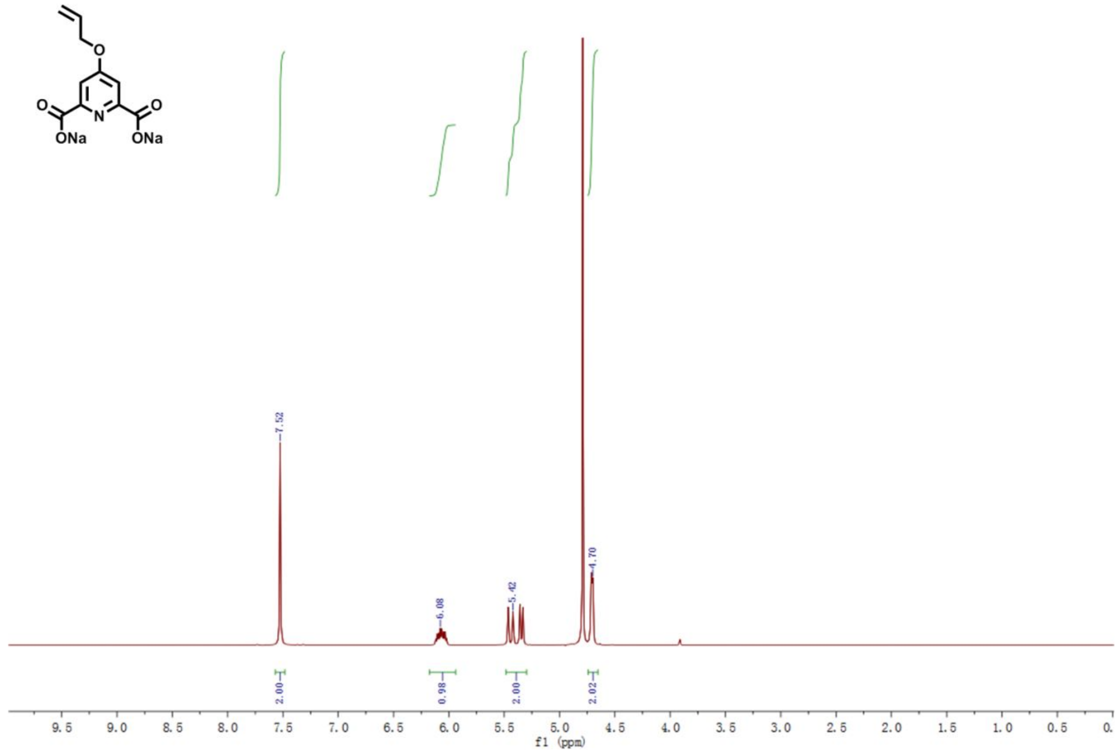


Figure S3.^{1}H NMR spectra of compound **SPD** (400MHz, D_2O).

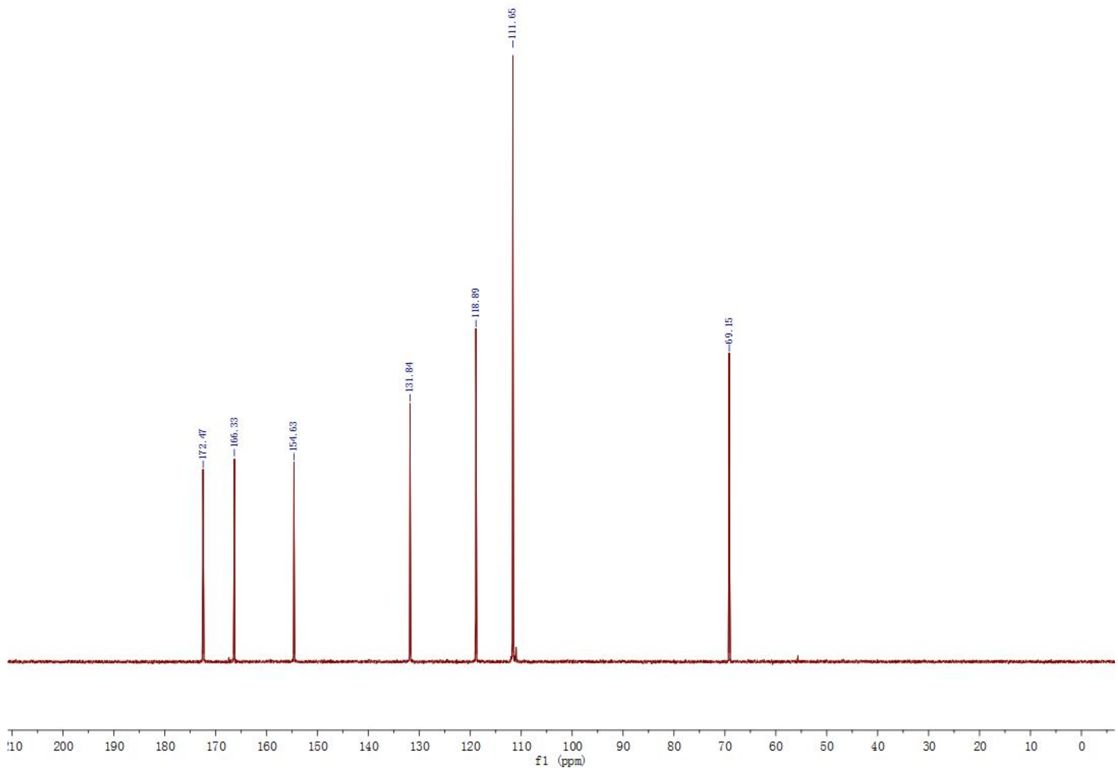


Figure S4.^{13}C NMR spectra of compound **SPD** (100MHz, D_2O).

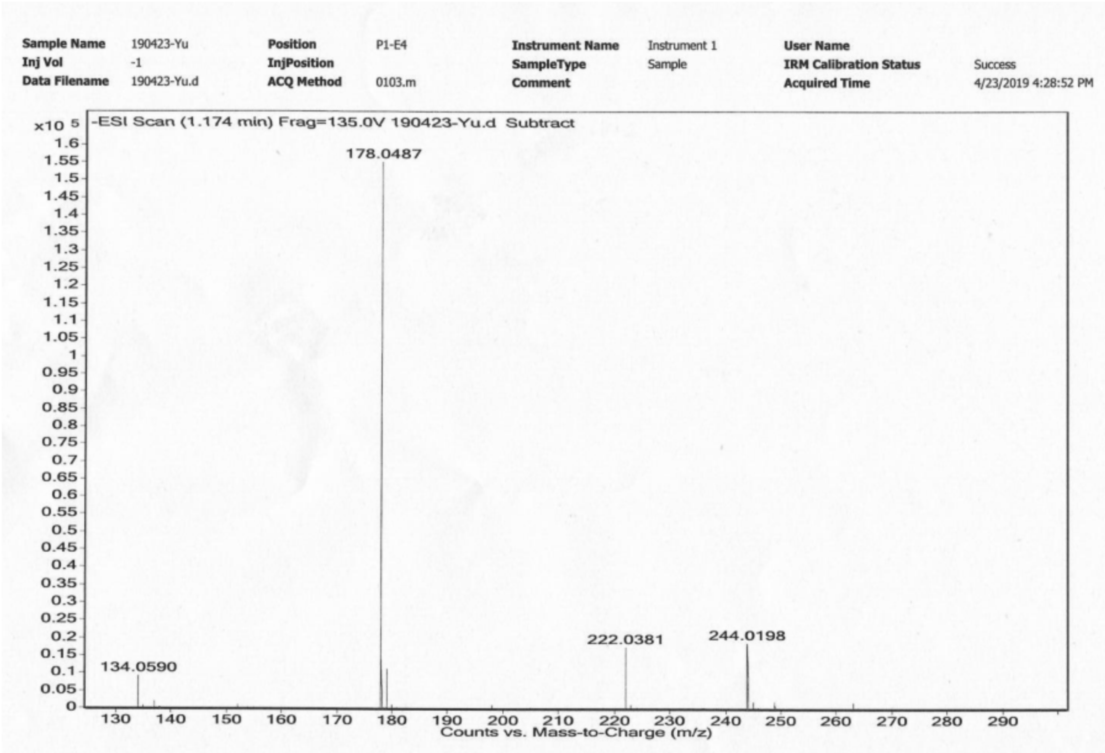


Figure S5. HR-ESI-MS spectra of compound **SPD**.

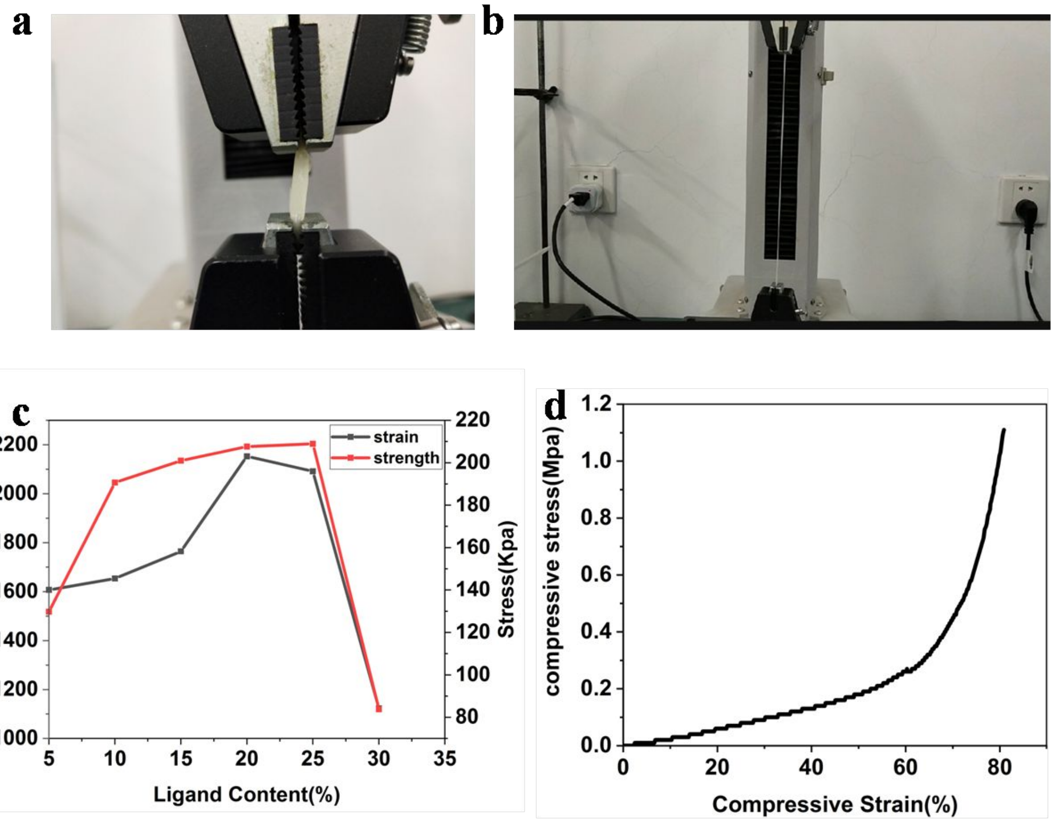


Figure S6. (a) Photo of 0% strain of 20% ligangs content (b) Photo of 2000% strain of 20% ligangs content (c) Stress-strain line chart of different ligangs content (d) compress stress-strain curve of 20% ligands content.

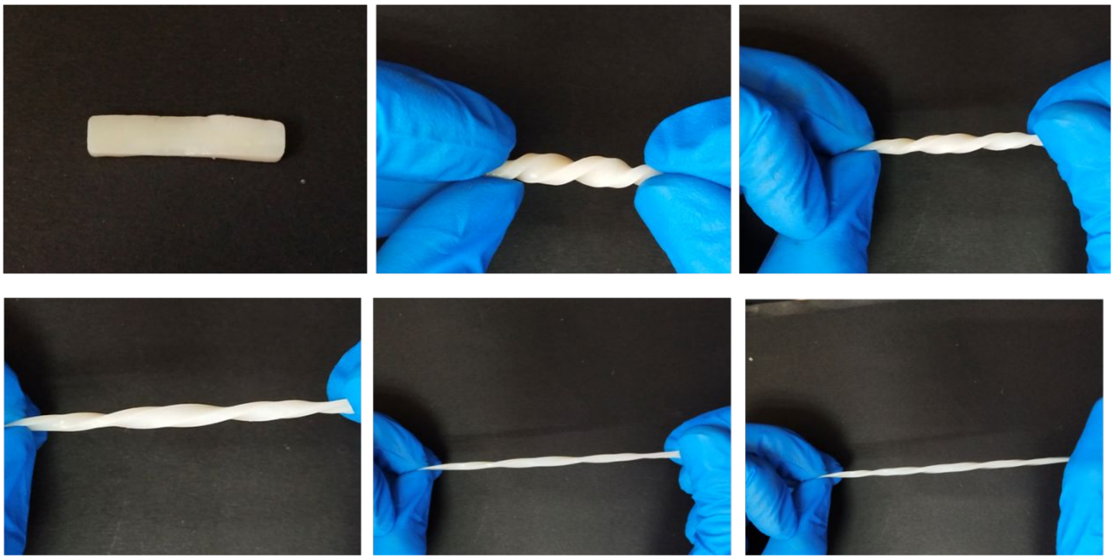


Figure S7. Photos of hydrogel withstand twisting.

Figure S8. Images to demonstrate the high strength of hydrogel by lifting up a 50 g substance.

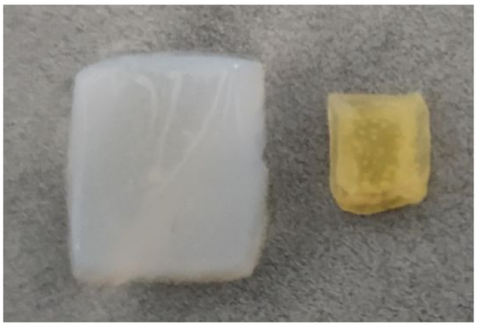


Figure S9. Swelling of the hydrogel in water. Right: the dry gel (28.3 mg); left: the water-swollen gel (285.6 mg).

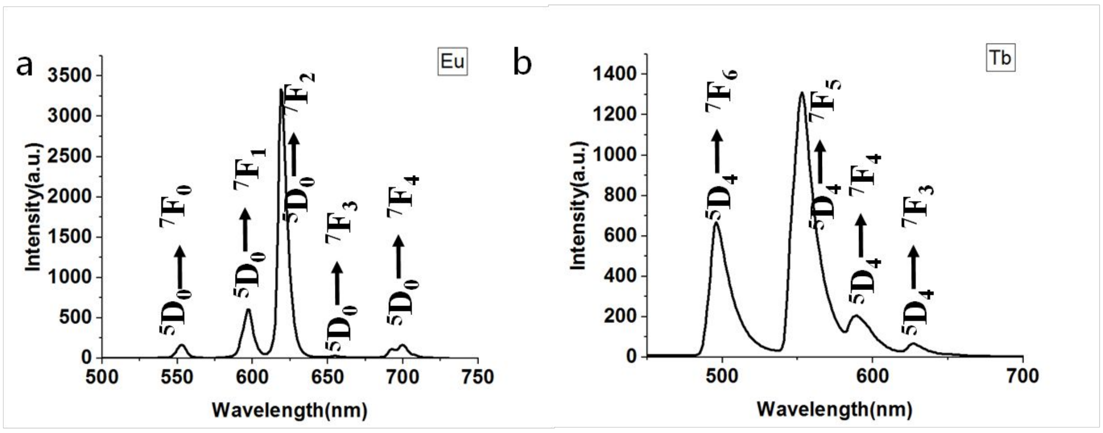


Figure S10. The emission spectra of Eu and Tb centered hydrogels, and their characteristic emission bands.

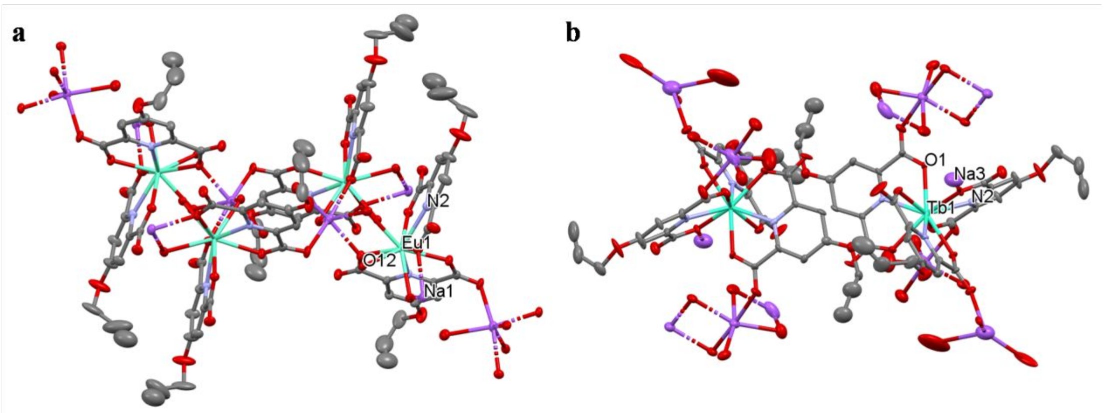


Figure S11. (a) 3D framework of Eu-complex (b) 3D framework of Tb-complex

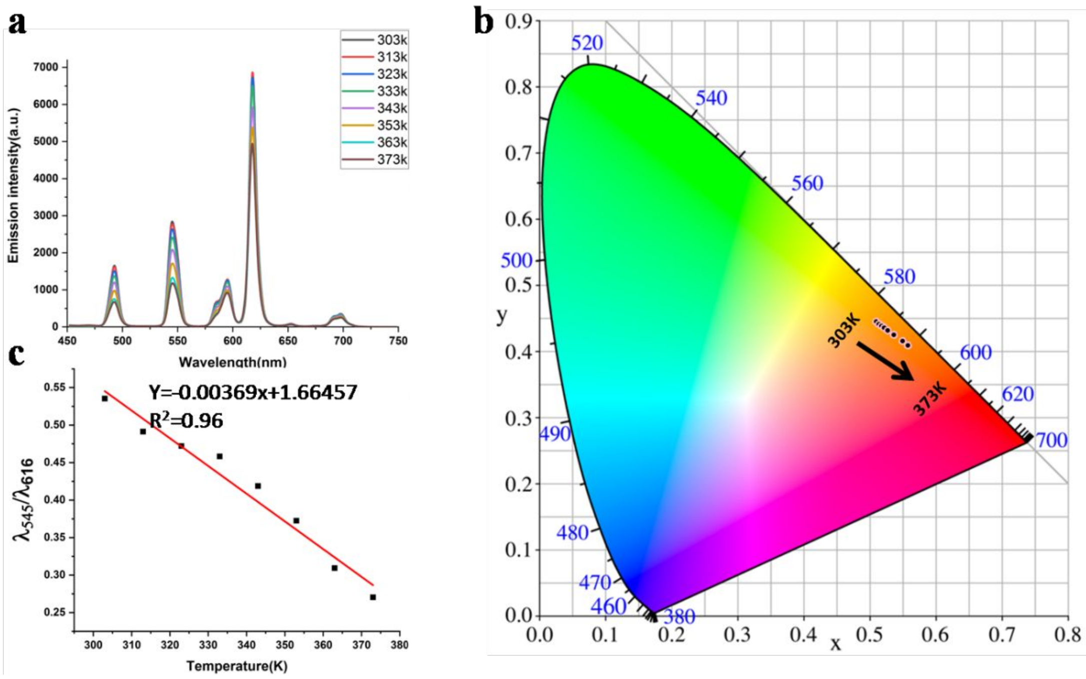


Figure S12. The emission spectra (a) and CIE coordination (b) of the hydrogel treated with different temperature (Eu/Tb=3:2) from 303K to 373K. (c) Relationship between the temperature and the green-to-red emission ratio (I_{544} /I_{616}).

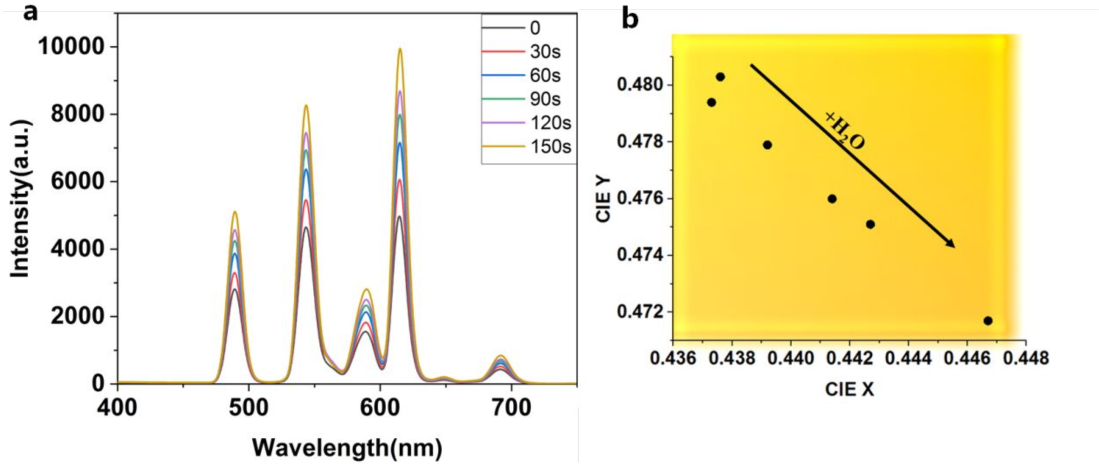


Figure 13. The emission spectra (a) and CIE coordination (b) of the dried gel treated with different time in water.

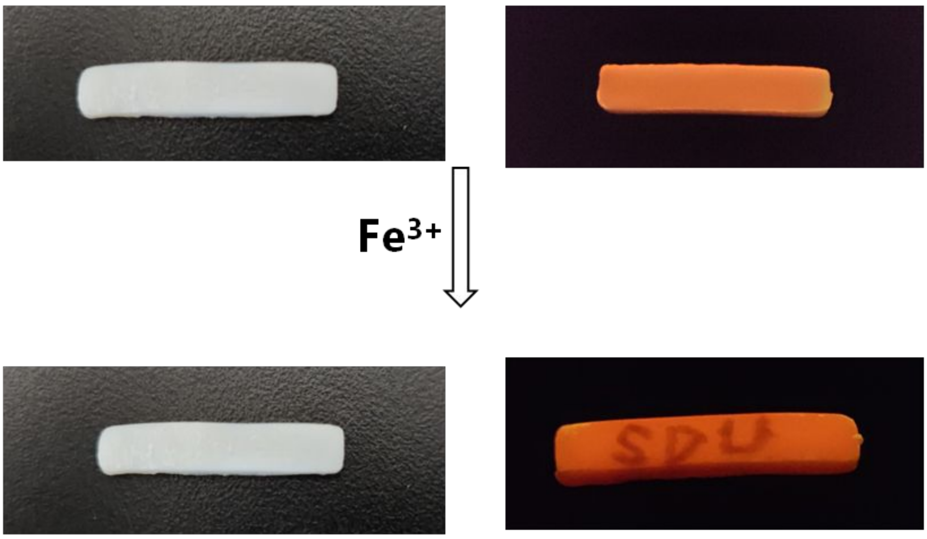


Figure S14. Fe^{3+}-induced fluorescence quenching of the Ln-containing hydrogel. Images showing the change in the fluorescence intensity under daylight and UV light before and after treated with Fe^{3+} by adding 50 mM $FeCl_3$ aqueous solution.

Figure S15. (a) Photographs showing the hydrogel(Eu) was applied from 0% to 2000% uniaxial strain (b) Photographs showing the hydrogel(Tb) was applied from 0% to 2000% uniaxial strain.

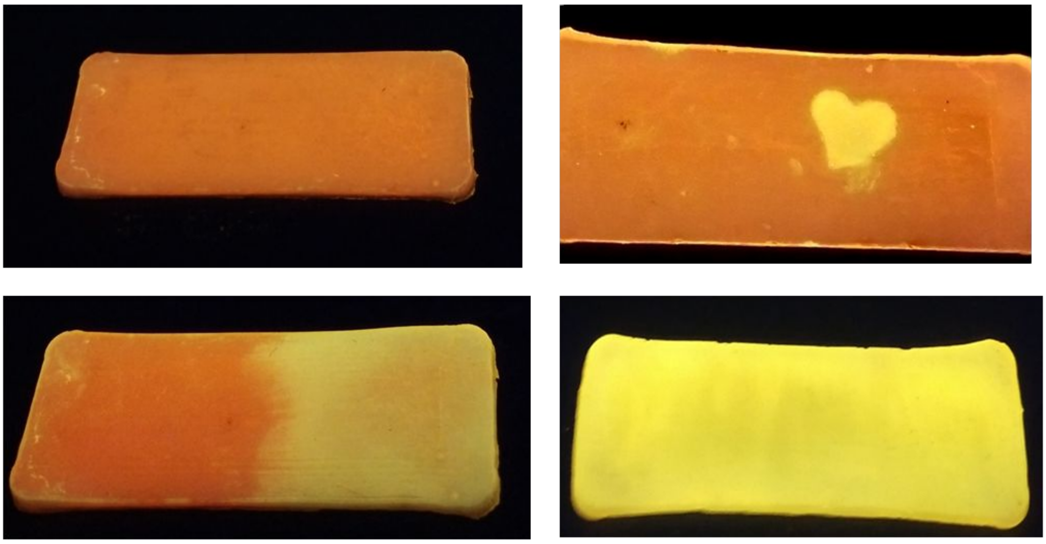


Figure S16. Photos of hydrogel before friction and after friction

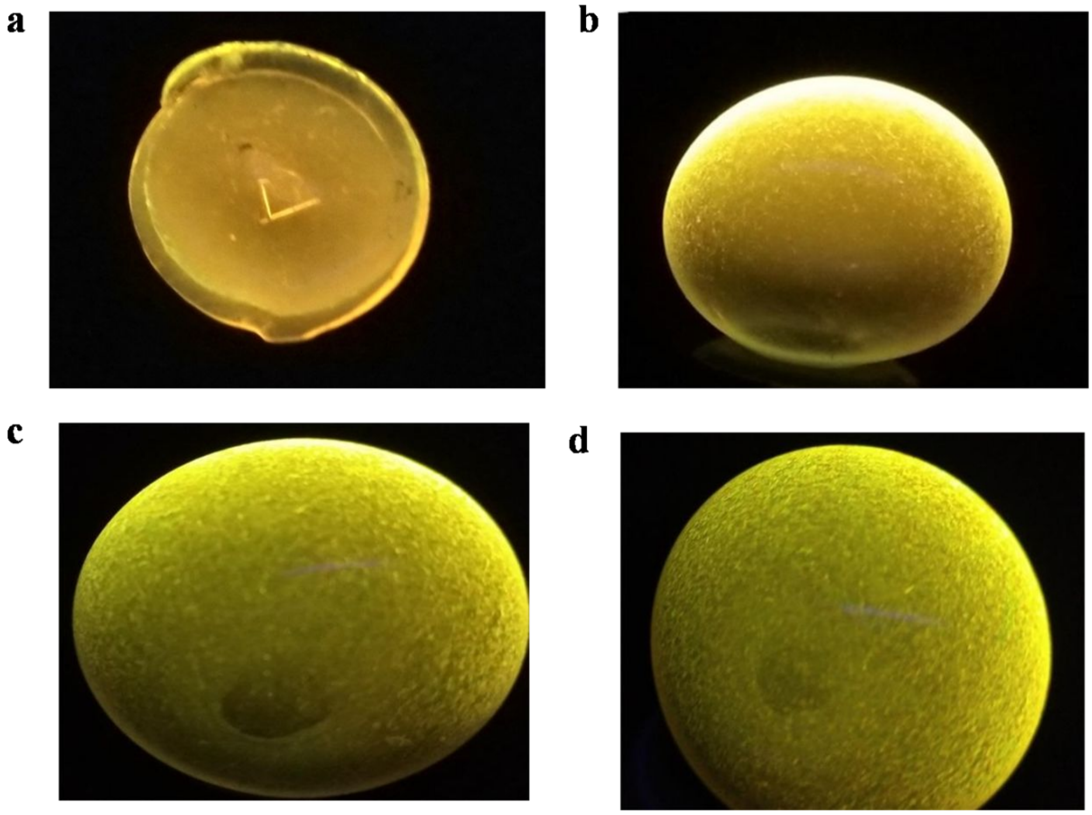


Figure S17. Photographs of the mechanochromic hydrogel under different bulging pneumatic pressures

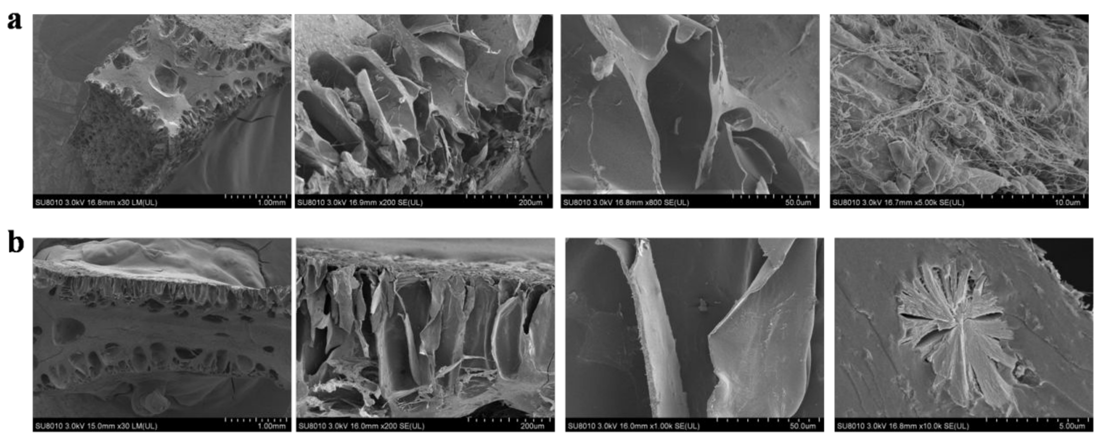


Figure S18. (a) SEM image of Eu-hydrogel (b) SEM image of Tb-hydrogel