

## Supporting Information

# Di(2-picolyl)amine-functionalized poly(ethylene glycol) hydrogels with tailororable metal-ligand coordination crosslinking

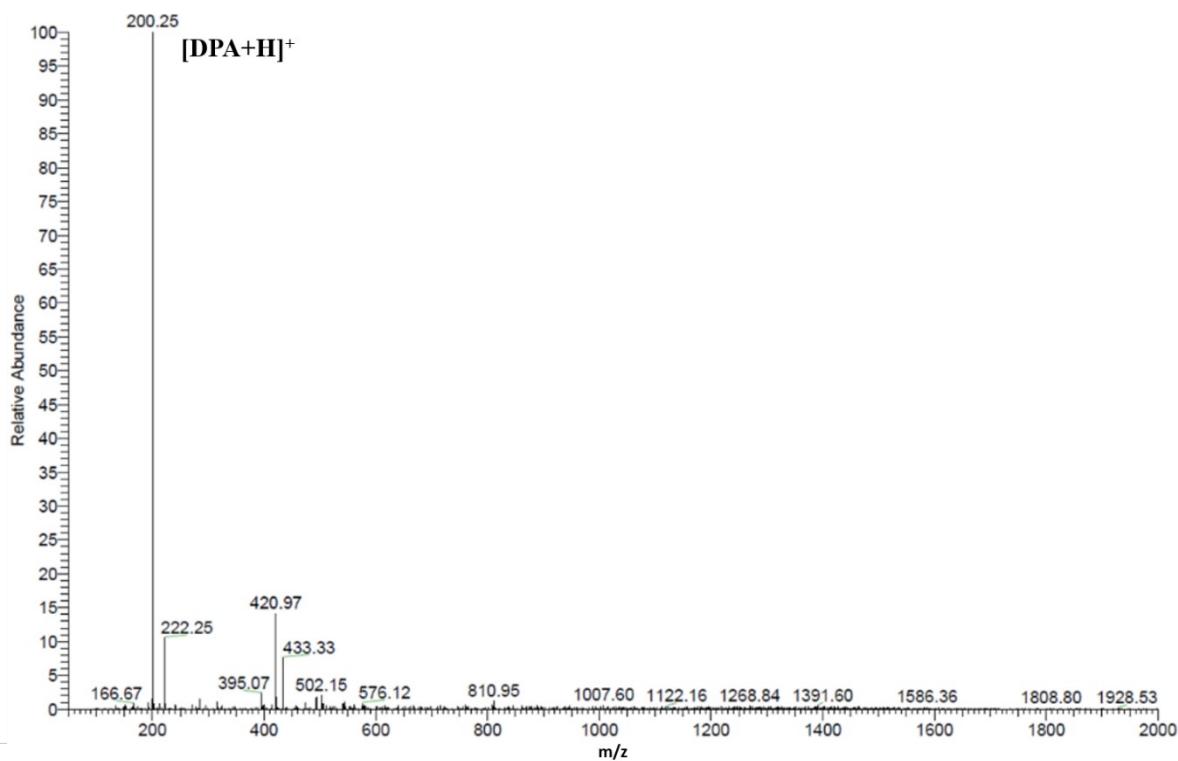
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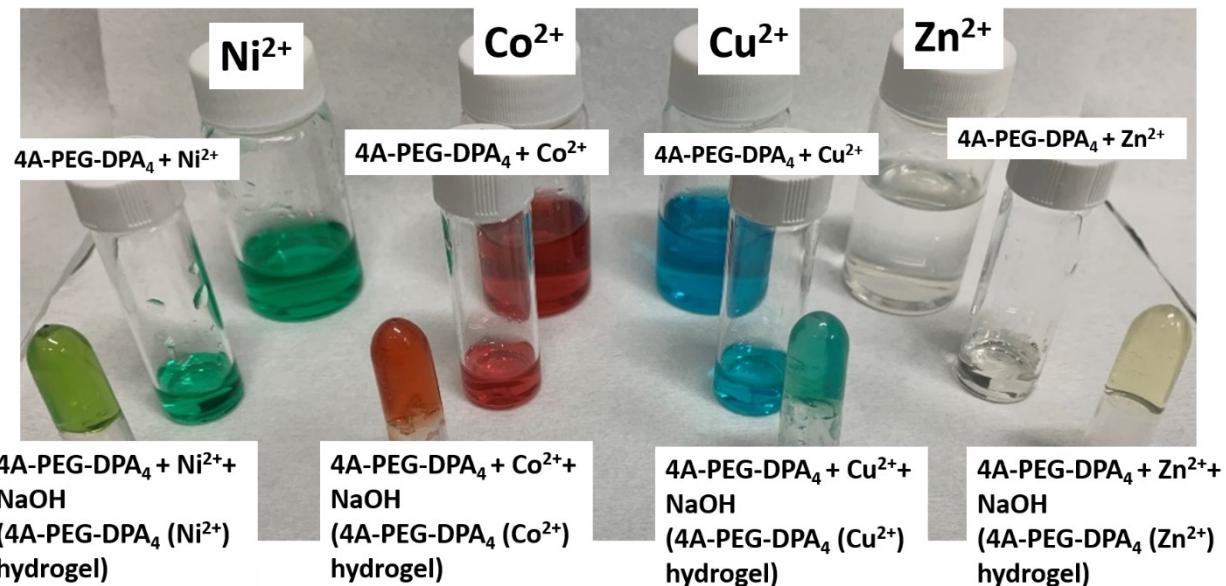
E-mail: [yicheun@ntu.edu.tw](mailto:yicheun@ntu.edu.tw)

\*Corresponding Author:

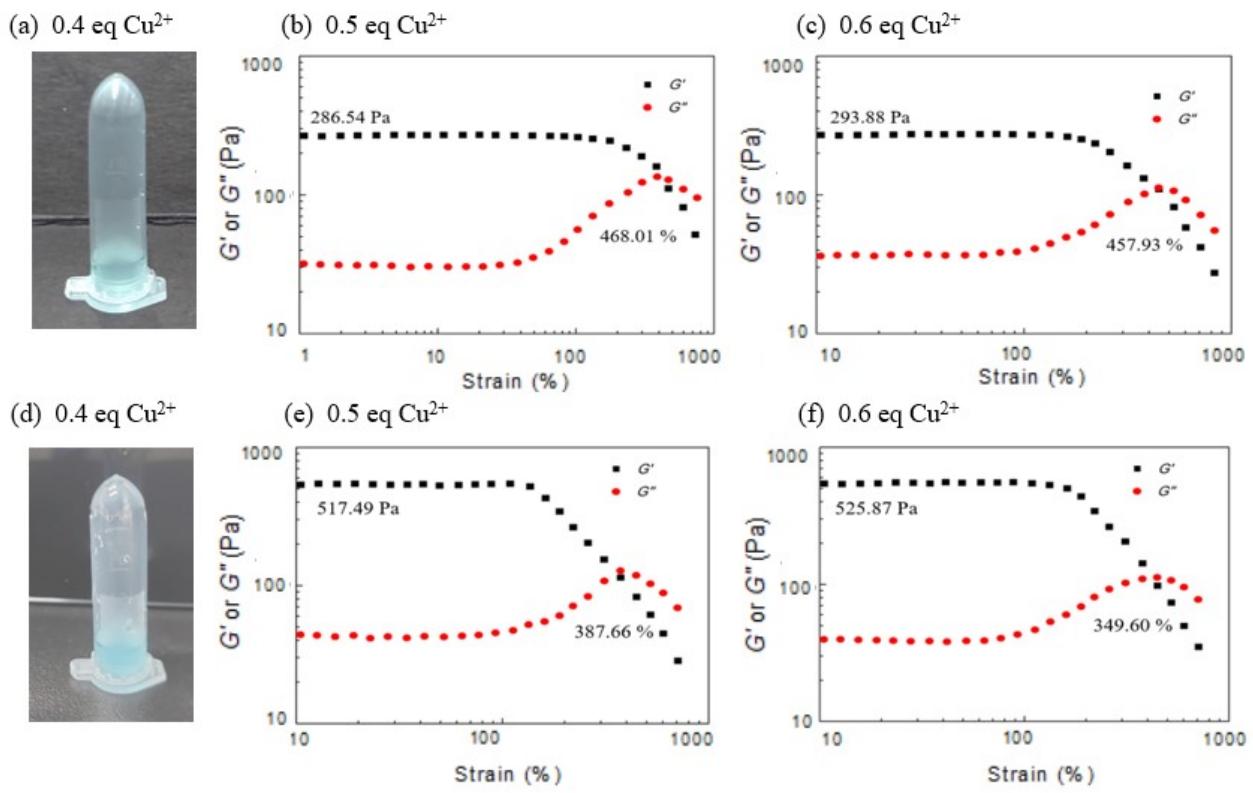
E-mail: [yicheun@ntu.edu.tw](mailto:yicheun@ntu.edu.tw). (Yi-Cheun Yeh)



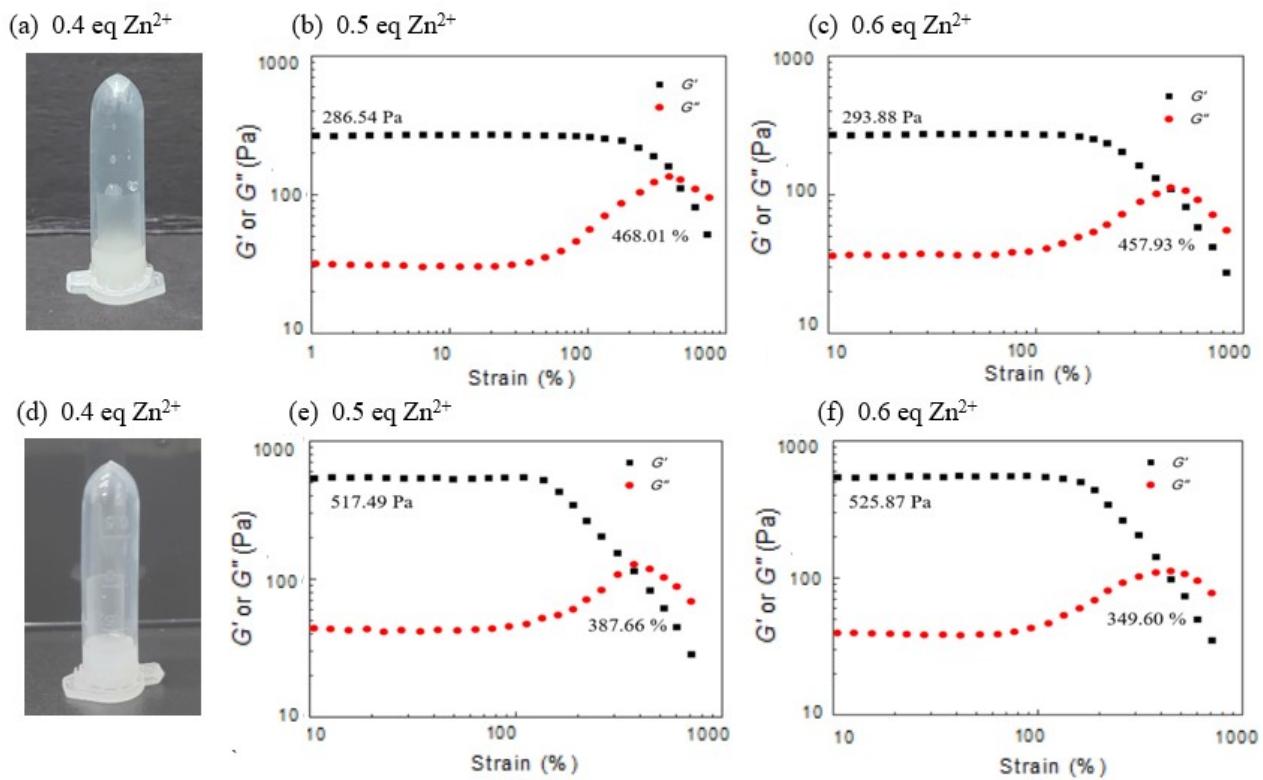
**Fig. S1** ESI spectrum of DPA molecule. The molecular weight of DPA is 199.11, thus the m/z signal of 200.25 is the  $[DPA+H]^+$  ion.



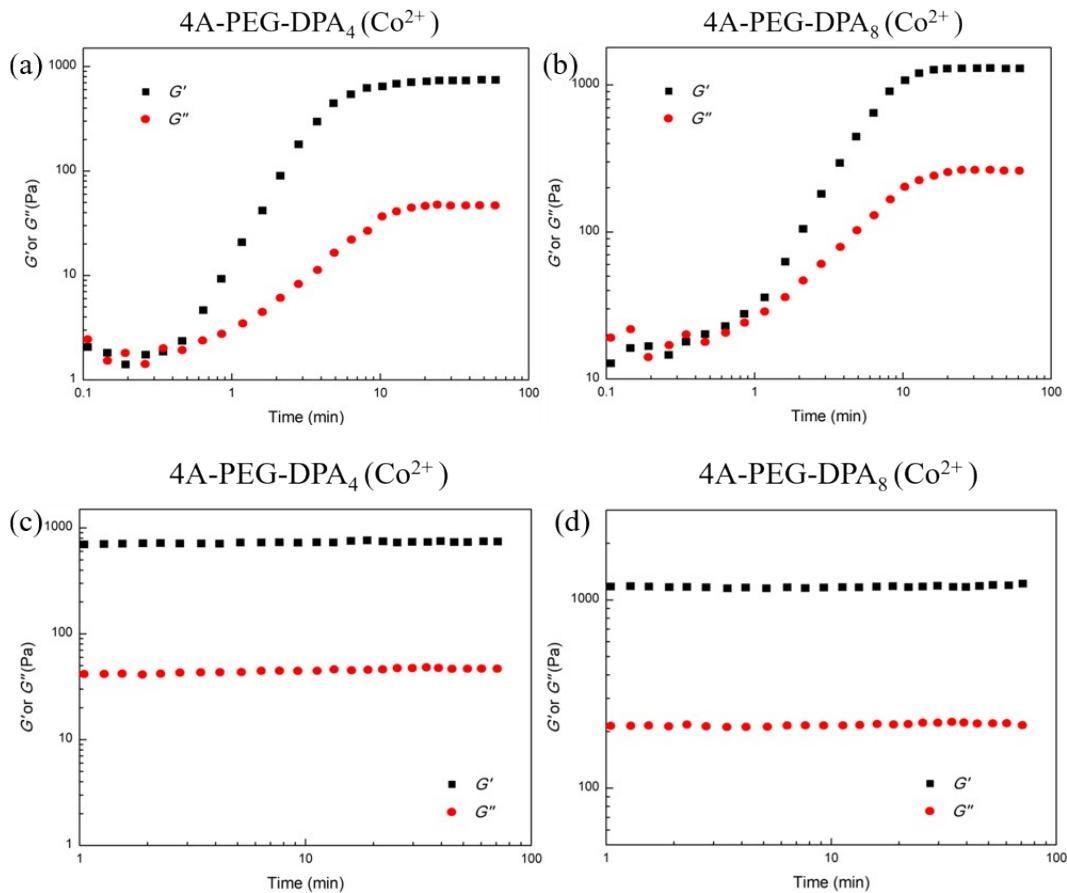
**Fig. S2** Images of solutions and hydrogels.



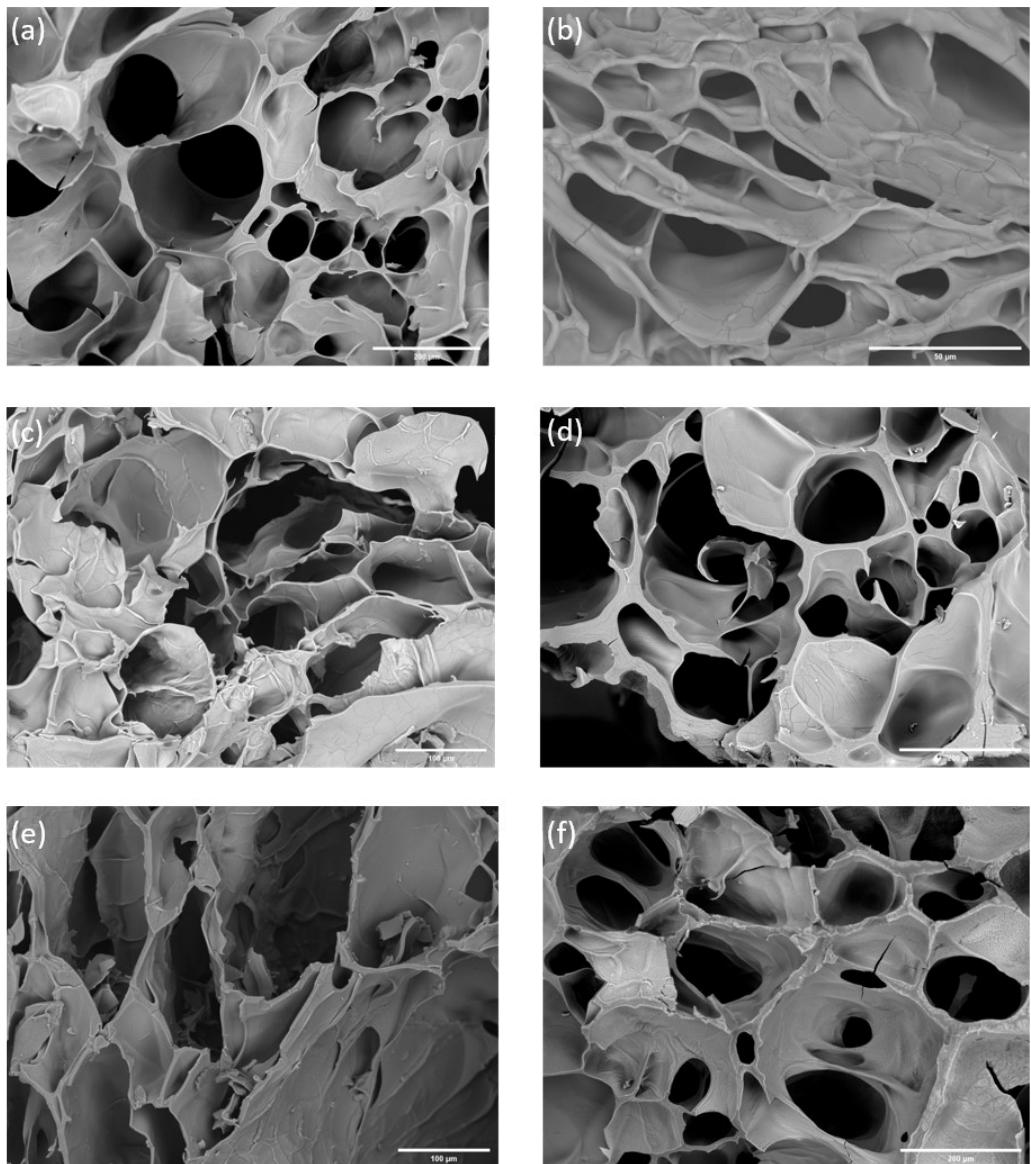
**Fig. S3** (a) Images of 4A-PEG-DPA<sub>4</sub>(Cu<sup>2+</sup>) hydrogels prepared with 0.4 eq of Cu<sup>2+</sup> ions. Oscillation strain sweeps of 4A-PEG-DPA<sub>4</sub>(Cu<sup>2+</sup>) hydrogel with (b) 0.5 eq and (c) 0.6 eq of Cu<sup>2+</sup> ions. (d) Images of 4A-PEG-DPA<sub>8</sub>(Cu<sup>2+</sup>) hydrogels prepared with 0.4 eq of Cu<sup>2+</sup> ions. Oscillation strain sweeps of 4A-PEG-DPA<sub>8</sub>(Cu<sup>2+</sup>) hydrogel with (e) 0.5 eq and (f) 0.6 eq of Cu<sup>2+</sup> ions.



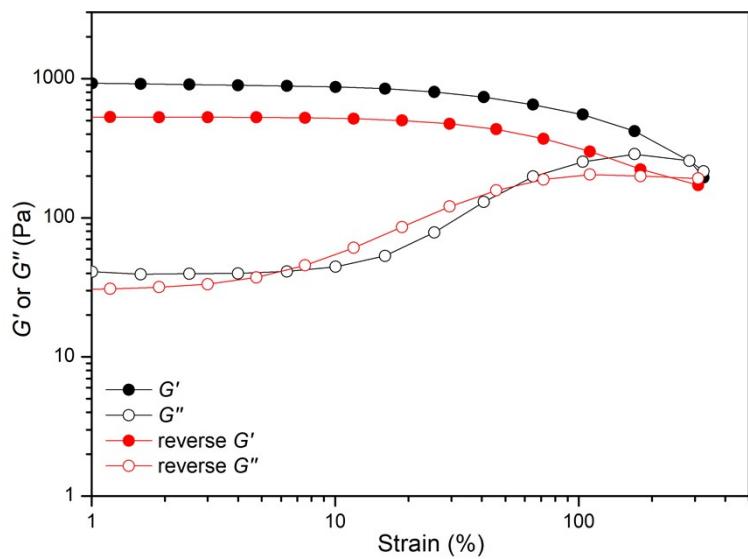
**Fig. S4** (a) Images of 4A-PEG-DPA<sub>4</sub>(Zn<sup>2+</sup>) hydrogels prepared with 0.4 eq of Zn<sup>2+</sup> ions. Oscillation strain sweeps of 4A-PEG-DPA<sub>4</sub>(Zn<sup>2+</sup>) hydrogel with (b) 0.5 eq and (c) 0.6 eq of Zn<sup>2+</sup> ions. (d) Images of 4A-PEG-DPA<sub>8</sub>(Zn<sup>2+</sup>) hydrogels prepared with 0.4 eq of Zn<sup>2+</sup> ions. Oscillation strain sweeps of 4A-PEG-DPA<sub>8</sub>(Zn<sup>2+</sup>) hydrogel with (e) 0.5 eq and (f) 0.6 eq of Zn<sup>2+</sup> ions.



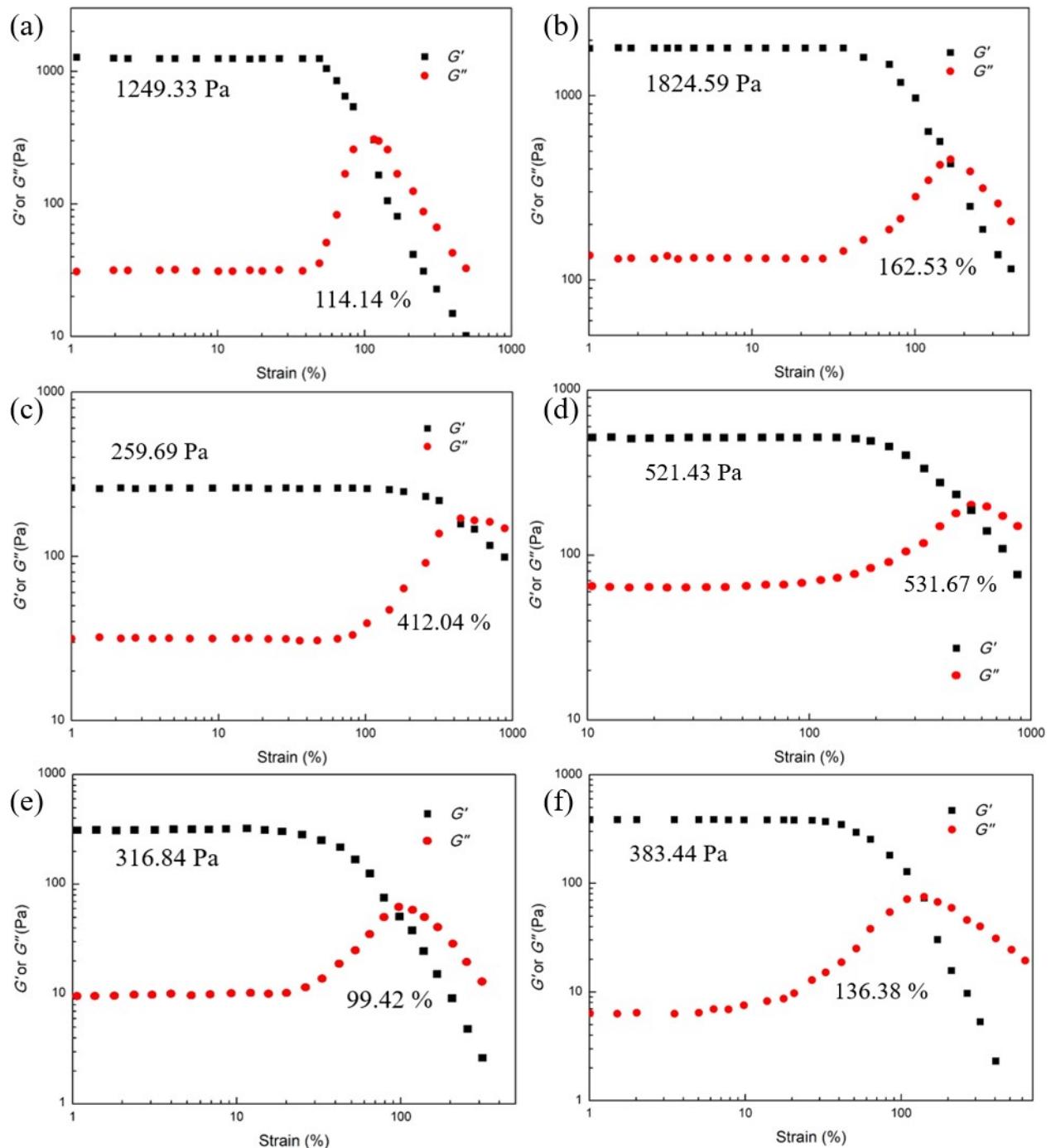
**Fig. S5** Oscillation time sweeps of (b) 4A-PEG-DPA<sub>4</sub> (Co<sup>2+</sup>) and (c) 4A-PEG-DPA<sub>8</sub> (Co<sup>2+</sup>) hydrogels, where the materials were quickly mixed and transferred on the plate of the rheometer. Oscillation time sweeps of (c) 4A-PEG-DPA<sub>4</sub>(Co<sup>2+</sup>) and (d) 4A-PEG-DPA<sub>8</sub> (Co<sup>2+</sup>) hydrogels, where the hydrogel was prepared and waited for 30 mins before loading on the plate of the rheometer.



**Fig. S6** Representative SEM images of (a) 4A-PEG-DPA<sub>4</sub> ( $\text{Ni}^{2+}$ ), (b) 4A-PEG-DPA<sub>8</sub> ( $\text{Ni}^{2+}$ ), (c) 4A-PEG-DPA<sub>4</sub> ( $\text{Cu}^{2+}$ ), (d) 4A-PEG-DPA<sub>8</sub> ( $\text{Cu}^{2+}$ ), (e) 4A-PEG-DPA<sub>4</sub> ( $\text{Zn}^{2+}$ ), and (f) 4A-PEG-DPA<sub>8</sub> ( $\text{Zn}^{2+}$ ) hydrogels.



**Fig. S7** The reverse strain sweep of 4A-PEG-DPA<sub>8</sub> (Co<sup>2+</sup>) hydrogel.



**Fig. S8** Representative oscillation strain sweeps of (a) 4A-PEG-DPA<sub>4</sub> ( $\text{Ni}^{2+}$ ), (b) 4A-PEG-DPA<sub>8</sub> ( $\text{Ni}^{2+}$ ), (c) 4A-PEG-DPA<sub>4</sub> ( $\text{Cu}^{2+}$ ), (d) 4A-PEG-DPA<sub>8</sub> ( $\text{Cu}^{2+}$ ), (e) 4A-PEG-DPA<sub>4</sub> ( $\text{Zn}^{2+}$ ), and (f) 4A-PEG-DPA<sub>8</sub> ( $\text{Zn}^{2+}$ ) hydrogels.