

## Supporting Information

### Photothermal and Robust Supramolecular soft material Cross-Linked by Dinuclear Heterodentate Coordination

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#### Supplementary Figures

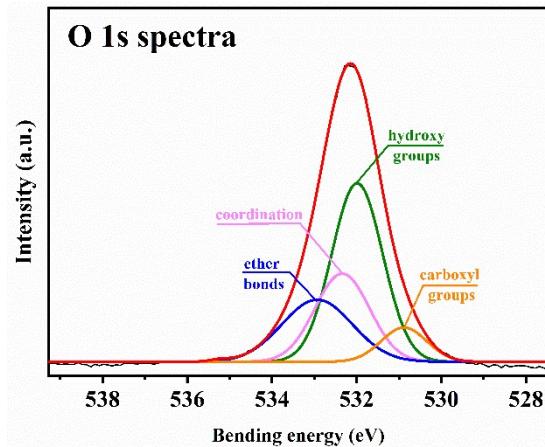


Fig. S1 High-resolution O 1s XPS spectra of XNBR/22.5OCTS-27/5Fe<sup>3+</sup>/4Cu<sup>2+</sup>. The red line was the global envelope used to fit the XPS spectra.

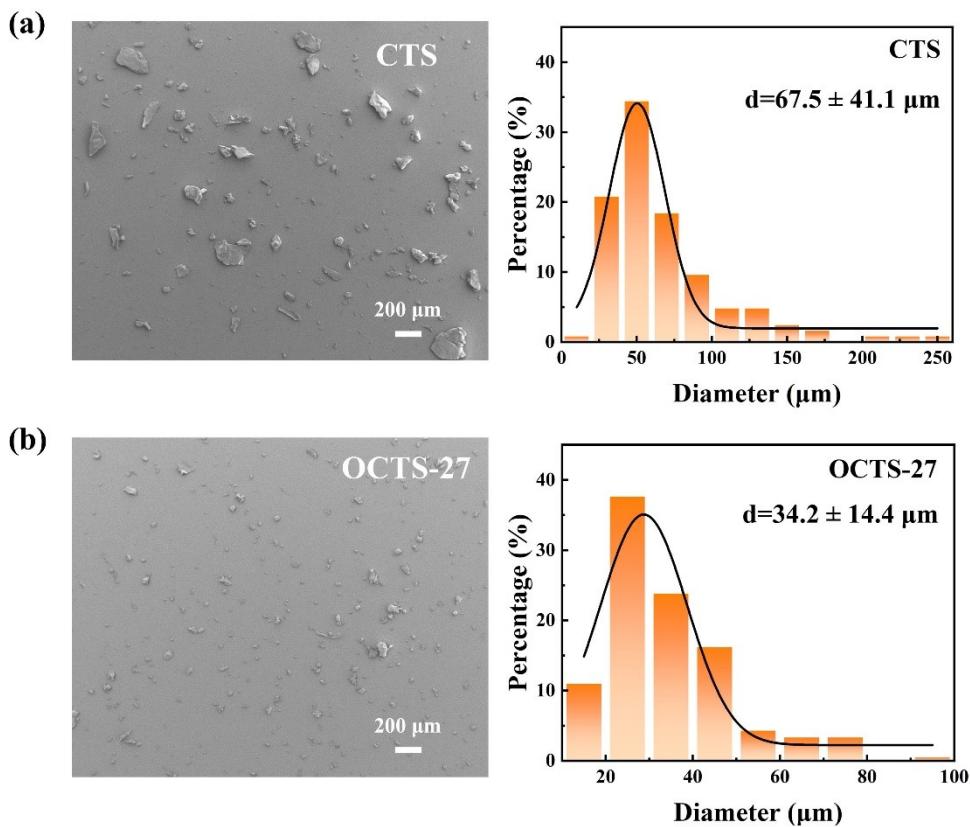
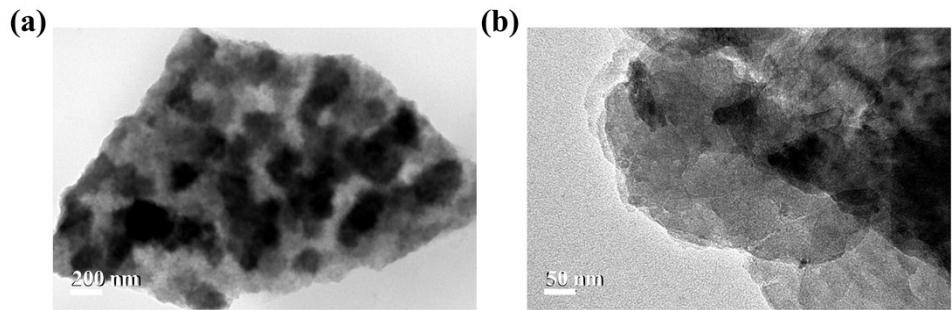
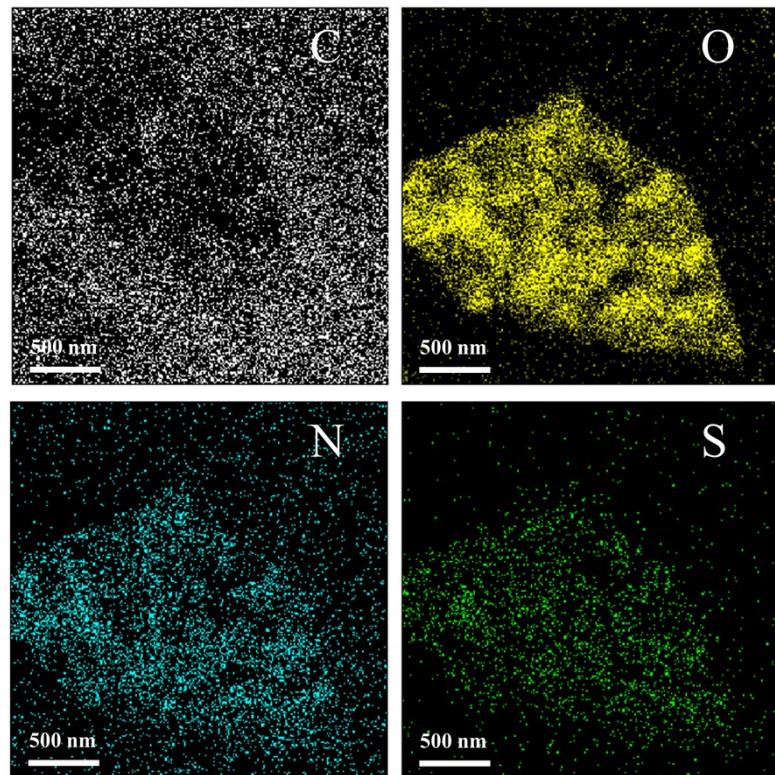


Fig. S2 Particle size and its distribution of a) CTS and b) OCTS-27.



**Fig. S3** a) TEM image and b) HRTEM image of XNBR/OCTS/Fe<sup>3+</sup>/Cu<sup>2+</sup>.



**Fig. S4** Element mapping images of XNBR/22.5OCTS-27/5Fe<sup>3+</sup>/4Cu<sup>2+</sup>: C, O, N, and S.

**Table S1** Comparison of mechanical properties between our work and those reported in literature

Material system	Tensile strength (MPa)	Elongation at break (%)	Ref.
NR/ZDMA/CNC	4.13	430	1
S-MXenes/S-ENR	4.55	388	2
Ti <sub>3</sub> C <sub>2</sub> MXene/ENR	4.93	1042	3
XNBR-API/Zn <sup>2+</sup>	6.8	2435	4
PMVS-COOH/PDMS-OH/TAPS	1.4	152	5
ENR/D/Fe <sup>3+</sup>	7.4	930	6
XNBR/TEA/Dy <sup>3+</sup>	4.5	2375	7
XNBR/OCTS/Fe <sup>3+</sup> /Cu <sup>2+</sup>	12.7	955	This work

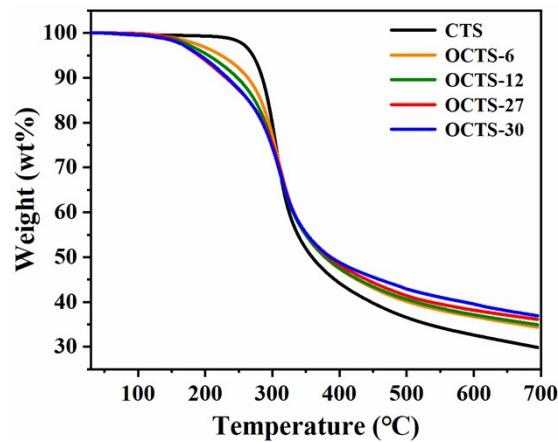


Fig. S5 TG curves of CTS and OCTS.

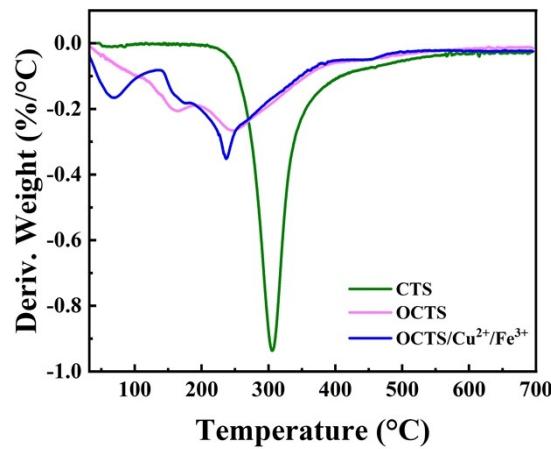


Fig. S6 DTG curves of CTS, OCTS, and OCTS/Fe<sup>3+</sup>/Cu<sup>2+</sup>.

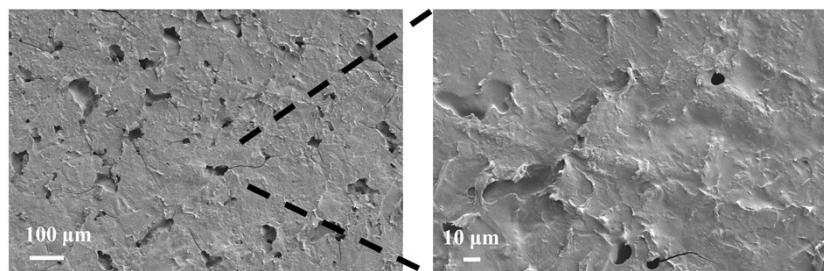


Fig. S7 SEM images of OCTS after carbonization.

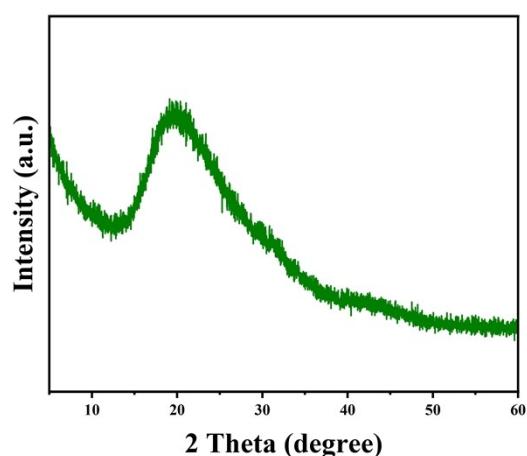
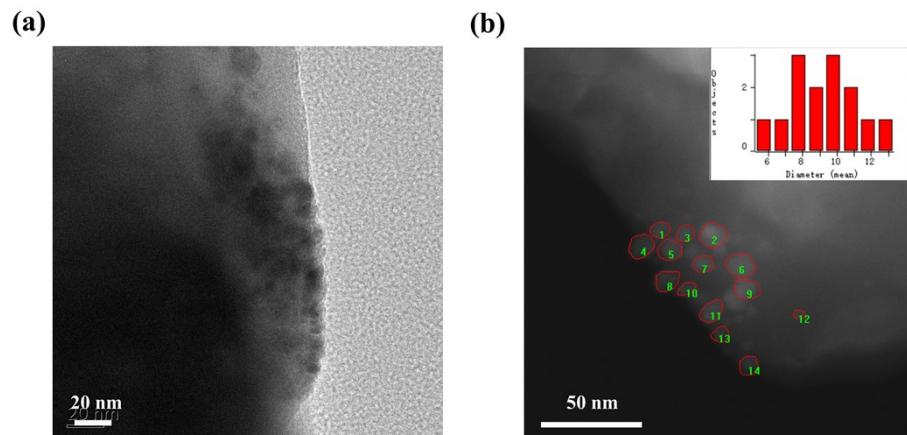
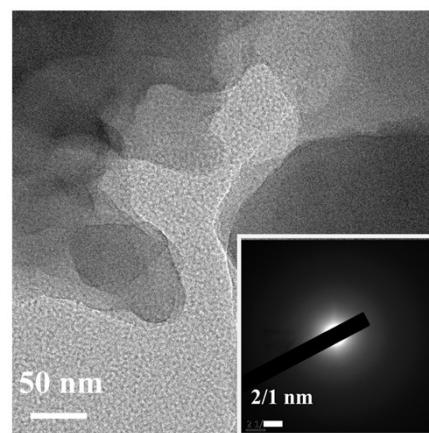


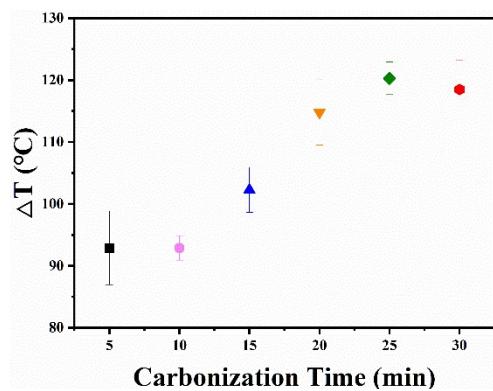
Fig. S8 XRD spectra of XNBR/OCTS/Fe<sup>3+</sup>/Cu<sup>2+</sup>.



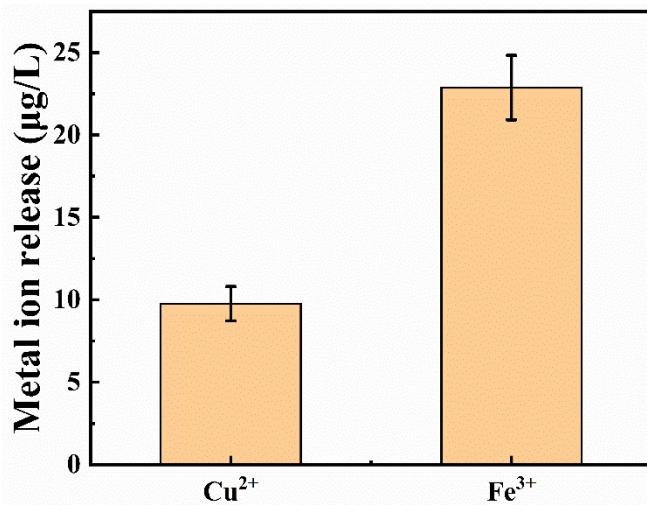
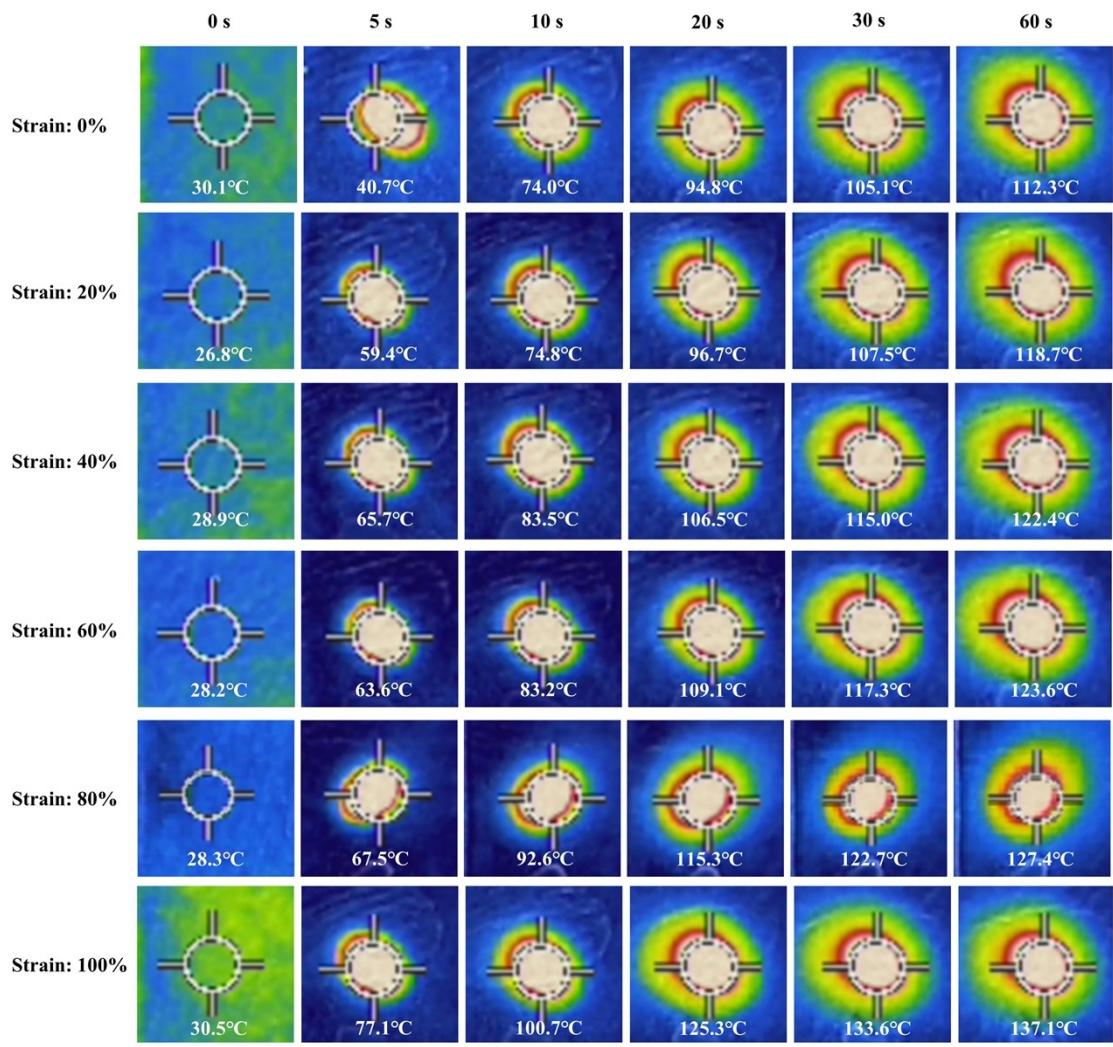
**Fig. S9** a) TEM image and b) STEM-HAADF image of OCTS/Fe<sup>3+</sup>/Cu<sup>2+</sup>.



**Fig. S10** TEM image of OCTS, where the white box is SAED pattern.



**Fig. S11** One-minute irradiation warming corresponding to OCTS at different carbonization temperatures.



**Fig. S12** IR imaging photographs of XNBR/22.5OCTS-27/5 $\text{Fe}^{3+}$ /4 $\text{Cu}^{2+}$  at 0–100% strain irradiated with near-infrared light.

**Fig. S13** ICP-MS results of Cu and Zn ion release from XNBR/22.5OCTS-27/5 $\text{Fe}^{3+}$ /4 $\text{Cu}^{2+}$  incubated in phosphate-buffered saline solution (PBS, pH 7.4) at 37 °C during 24 h.

## Reference

- 1 M. Wu, L. Yang, Z. Zheng, F. Wan, X. Teng and C. Xu, *Int. J. Biol. Macromol.*, 2022, **222**, 587–598.
- 2 Q. Guo, X. Zhang, F. Zhao, Q. Song, G. Su, Y. Tan, Q. Tao, T. Zhou, Y. Yu, Z. Zhou and C. Lu, *ACS Nano*, 2020, **14**, 2788–2797.

- 3 Q. Gan, L. Song, Y. Wang, Q. Yuan, W. Huang, Y. Zhu, Y. Huang and Y. Song, *Nano Energy*, 2024, **120**, 109141.
- 4 M. Das, A. Baran Bhattacharya, A. Rahman Parathodika and K. Naskar, *Eur. Polym. J.*, 2022, **174**, 111341.
- 5 H. Sun, X. Liu, B. Yu, Z. Feng, N. Ning, G.-H. Hu, M. Tian and L. Zhang, *Polym. Chem.*, 2019, **10**, 633-645.
- 6 K. Buaksuntar, K. Panmanee, K. Wongphul, P. Lim-arun, S. Jansinak, D. U. Shah and W. Smitthipong, *Polymer*, 2024, **291**, 126626.
- 7 X. Huang, A. Zhang, Q. Tan, K. Gou, Y. Chen, Y. Nie and G. Weng, *Macromolecules*, 2024, **57**, 963-975.