

**Supplementary Information**

Polyurea interface structure with dual dynamic bonds endowing  
composites with synchronous self-healing and thermal conductivity

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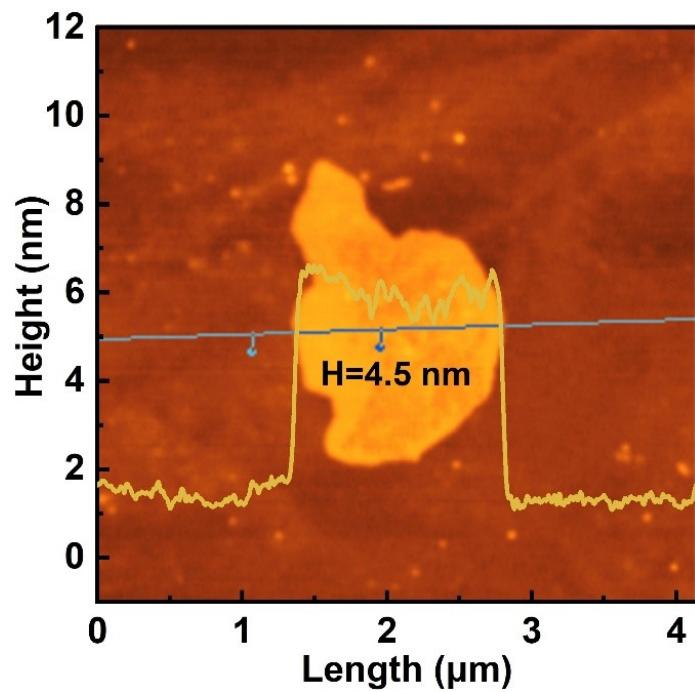


Fig. S1 AFM topographic image and curves of surface scale for GO.

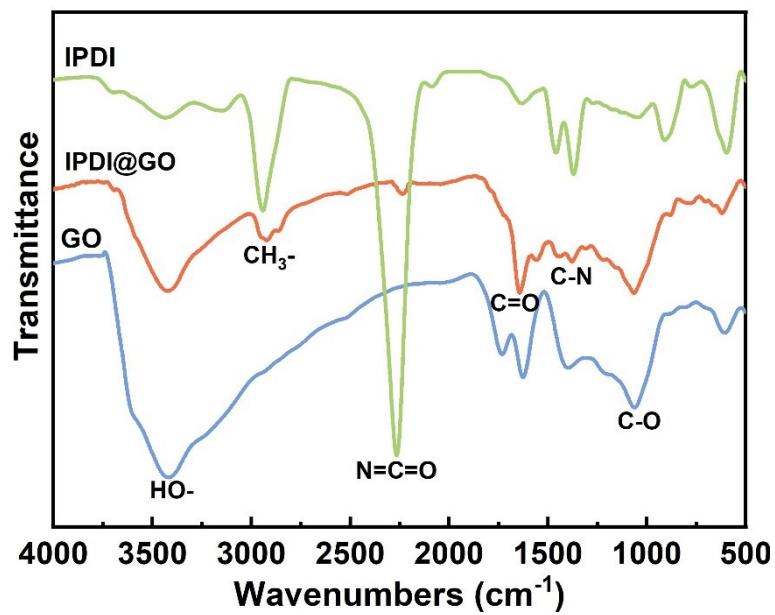


Fig. S2 FT-IR spectrum of GO, IPDI, and IPDI@GO.

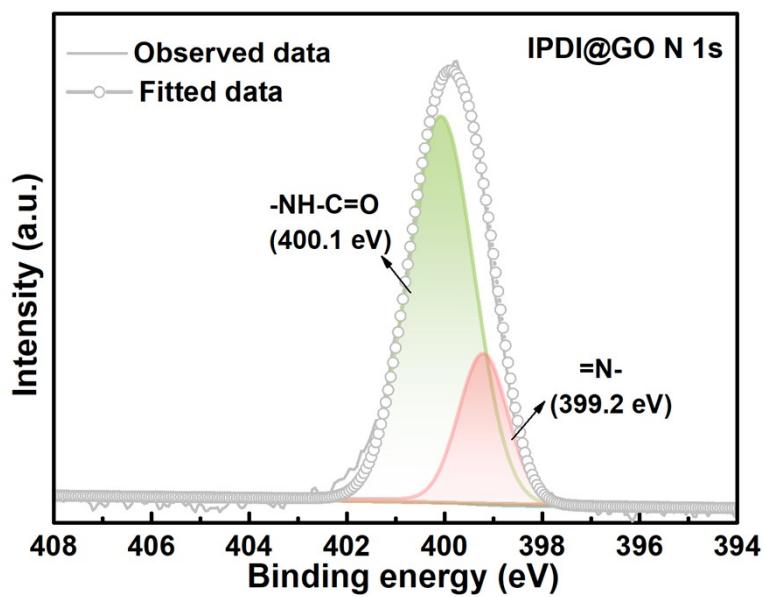


Fig. S3 XPS N1s spectra of IPDI@GO.

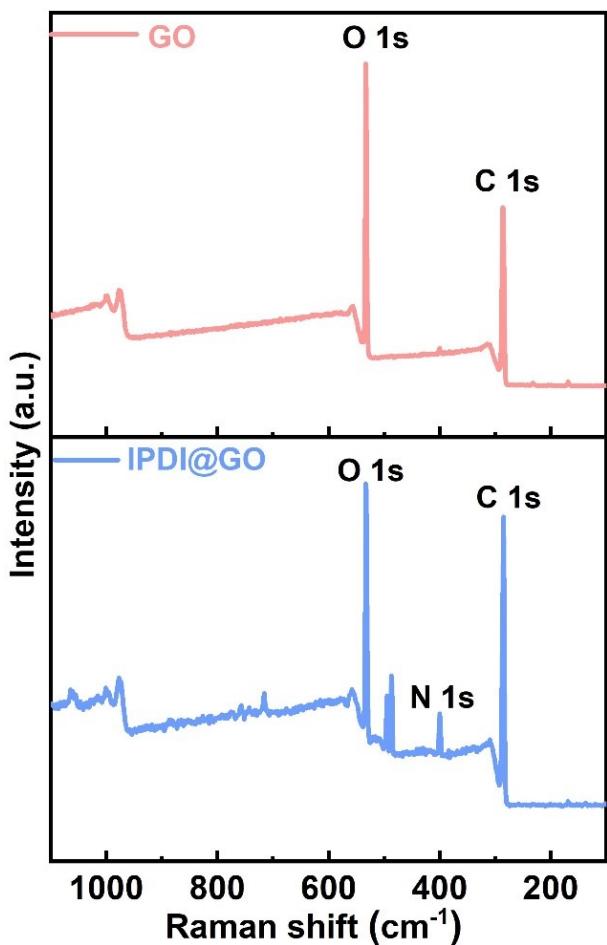


Fig. S4 XPS full spectra of GO and IPDI@GO.

**Table S1.** Comparison of atomic concentration of C and N in GO and IPDI@GO by the XPS full spectra.

Samples	Atomic concentration (%)		C/N atomic ration
	C 1s	N 1s	
GO	67.87	1.01	67.20
IPDI@GO	72.26	5.29	13.66

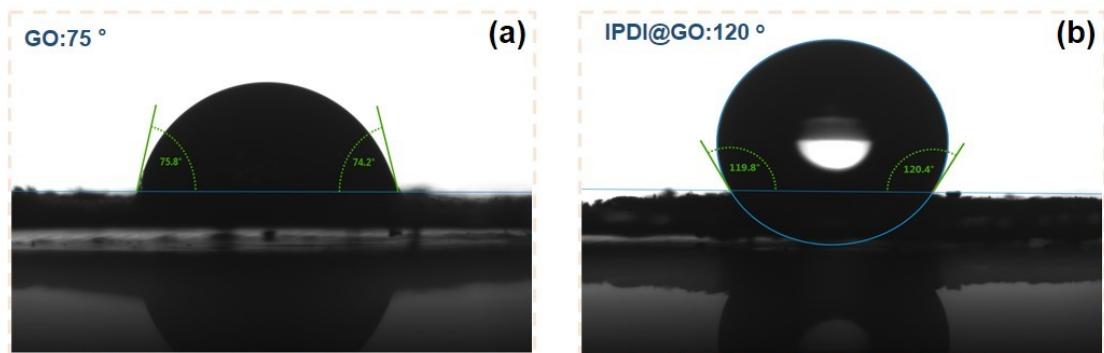


Fig. S5 Contact angles of  $\text{H}_2\text{O}$  on the surface of GO and IPDI@GO.

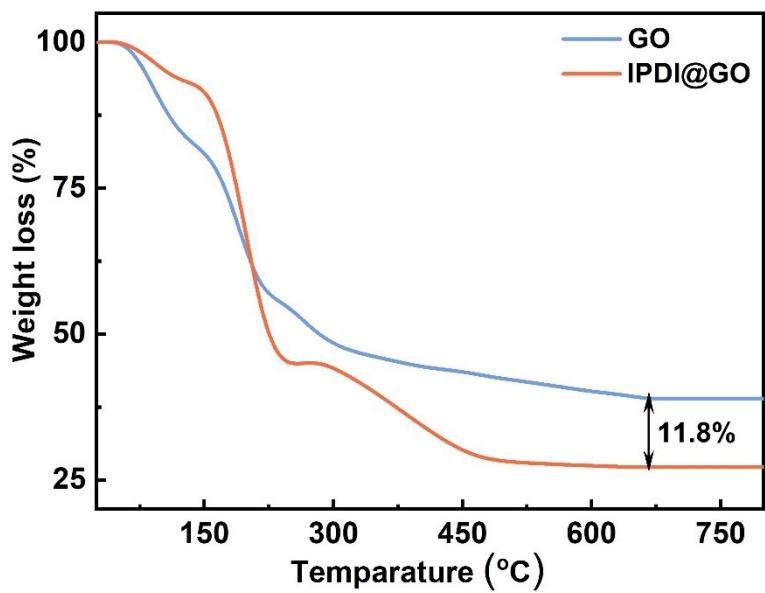


Fig. S6 TGA of GO and IPDI@GO.

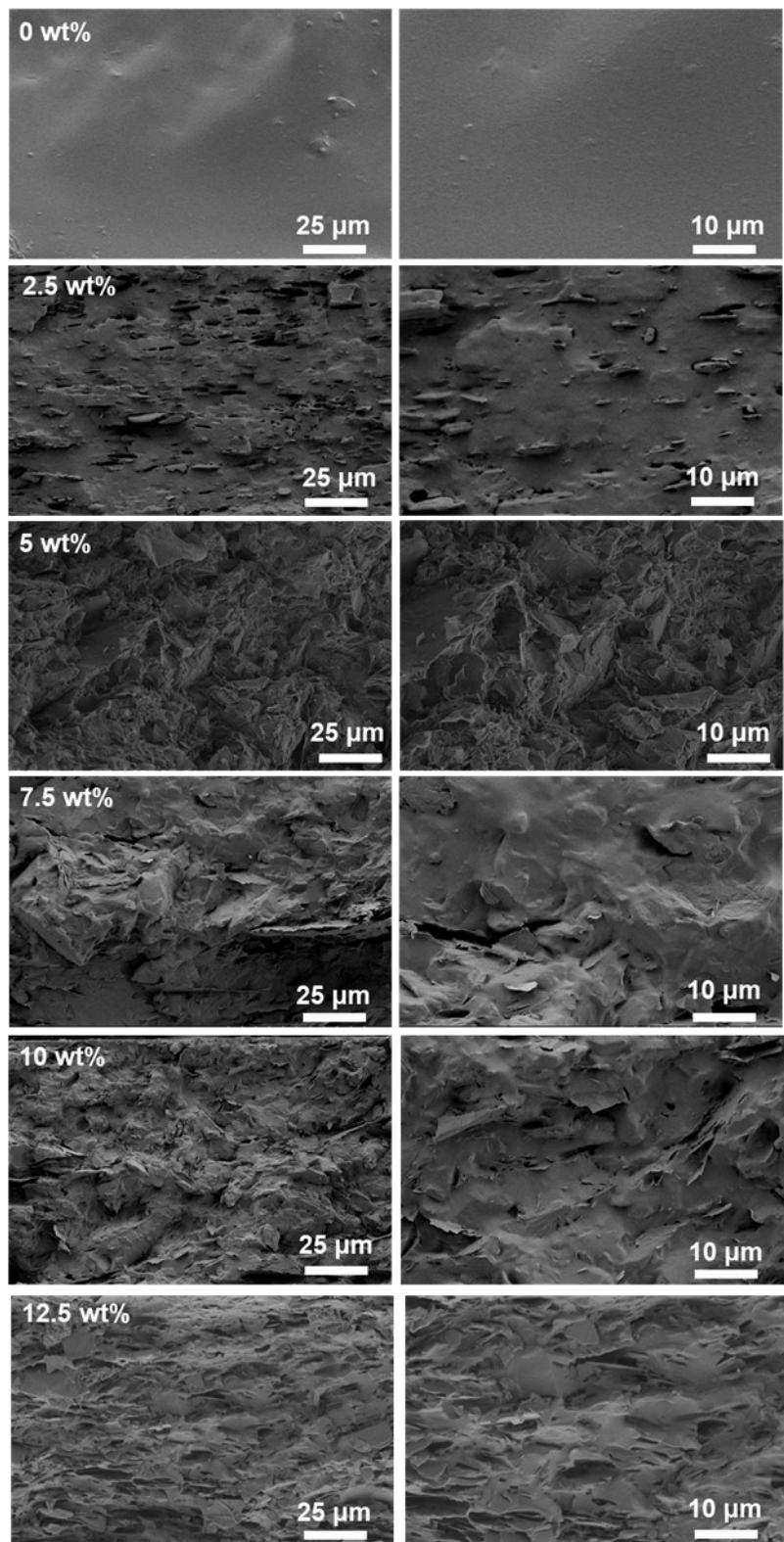


Fig.S7 Cross-sectional SEM images of IPDI@GO<sub>x</sub>/D-PUA (x refers to the mass percentage of the filler content).

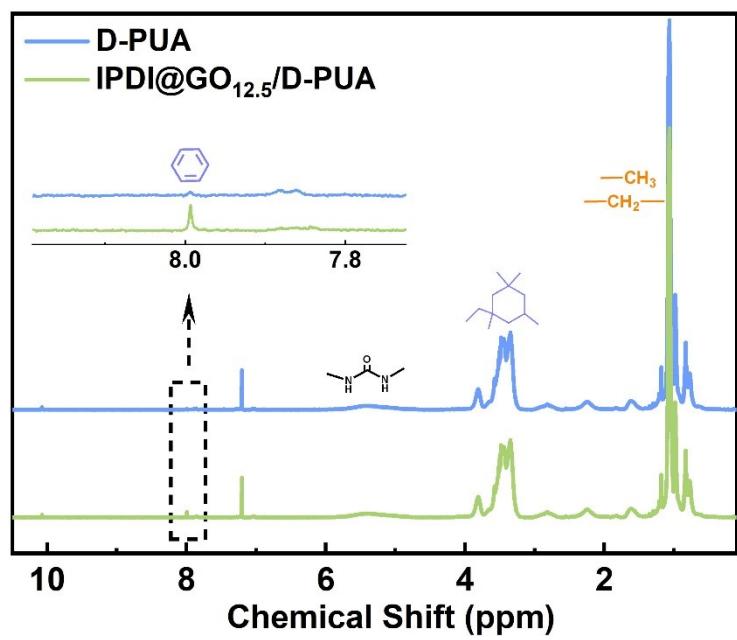


Fig. S8 <sup>1</sup>H NMR of D-PUA and IPDI@GO<sub>12.5</sub>/D-PUA.

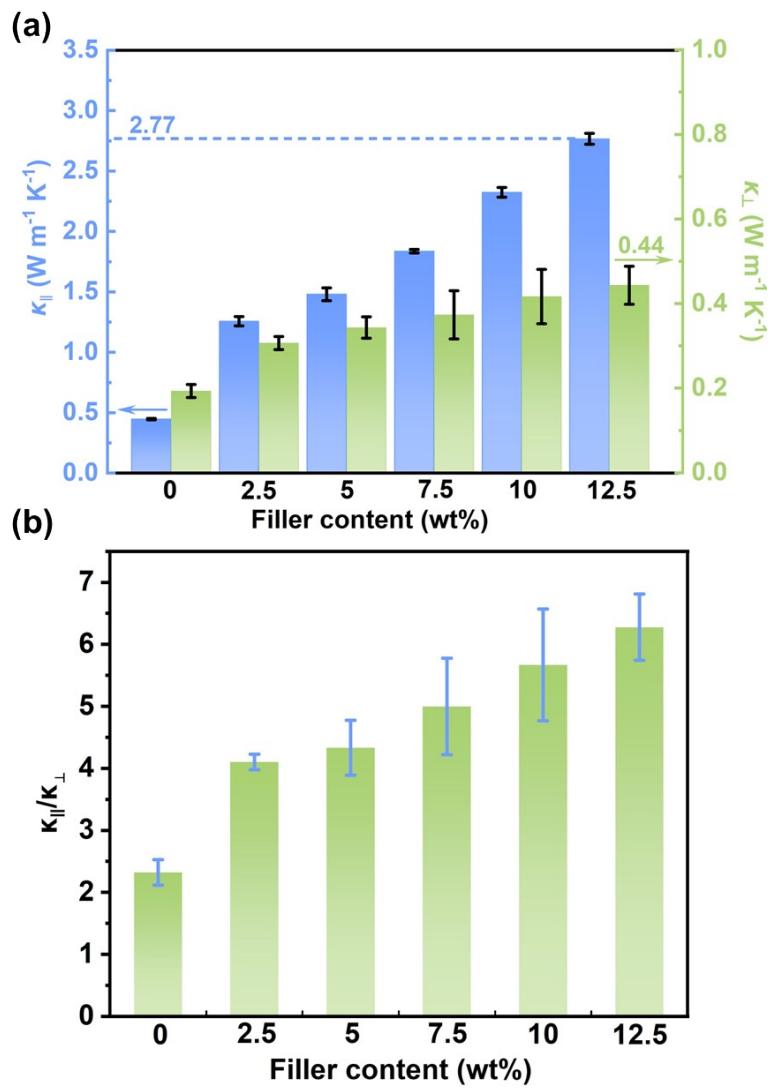


Fig.S9 (a)  $\kappa_{\parallel}$  and  $\kappa_{\perp}$ , (b) Ratio of  $\kappa_{\parallel}$  and  $\kappa_{\perp}$  for IPDI@GO<sub>x</sub>/D-PUA with different filler loading.

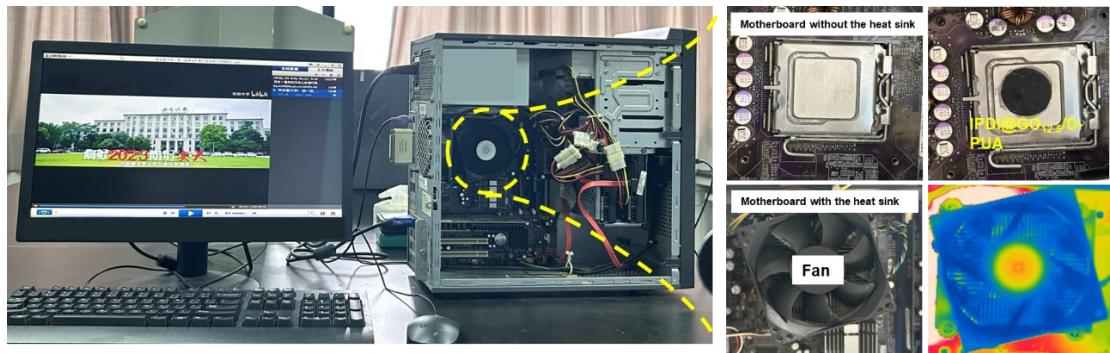


Fig. S10 Experimental setup for comparing the cooling efficiency between IPDI@GO<sub>12.5</sub>/D-PUA and BGS-GP1000 based on desktop computer systems.

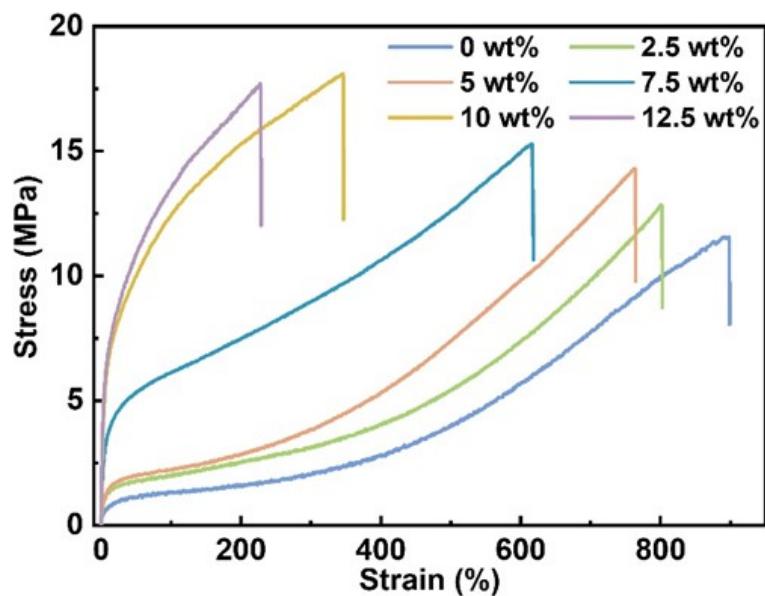


Fig. S11 Stress-strain curves of IPDI@GO<sub>x</sub>/D-PUA with different filler loading.

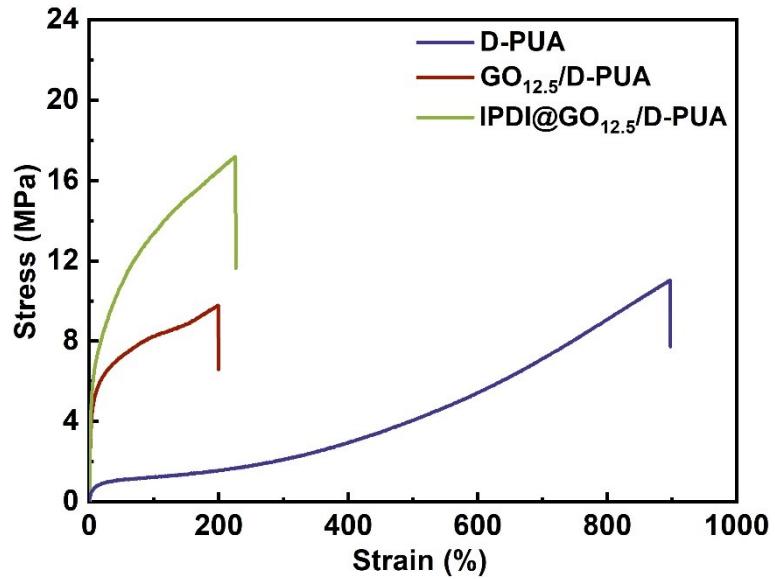


Fig. S12 Stress-strain curves of the D-PUA,  $\text{GO}_{12.5}/\text{D-PUA}$ , and  $\text{IPDI}@\text{GO}_{12.5}/\text{D-PUA}$ .

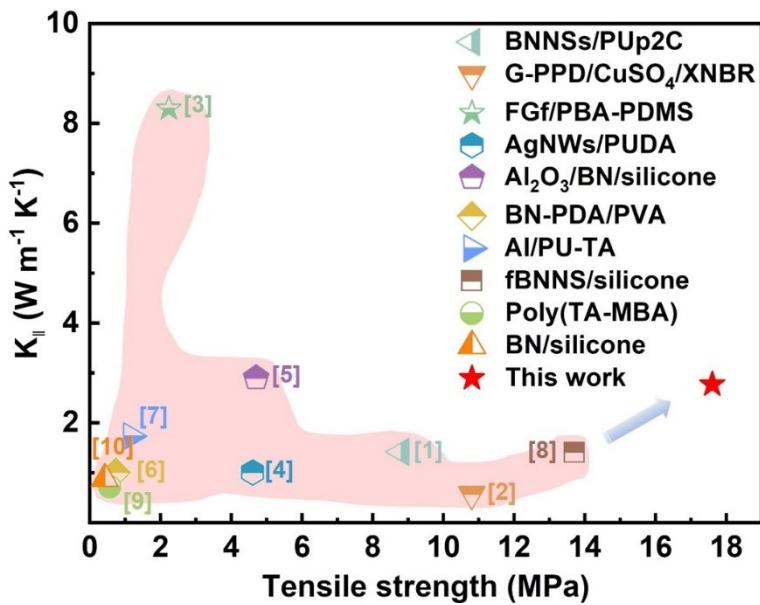


Fig. S13 Comparison of  $\kappa_{\parallel}$  and tensile strength for IPDI@GO<sub>12.5</sub>/D-PUA with previously reported composites [1-10].

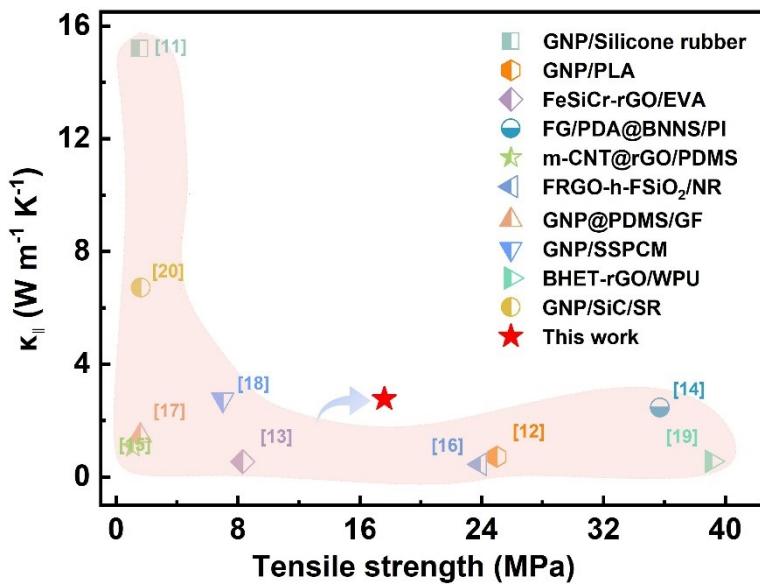


Fig. S14 Comparison of  $\kappa_{\parallel}$  and tensile strength for IPDI@GO<sub>12.5</sub>/D-PUA with previously reported graphene-based nanocomposites [11-20].



Fig. S15 Digital image of IPDI@GO<sub>12.5</sub>/D-PUA on petal.

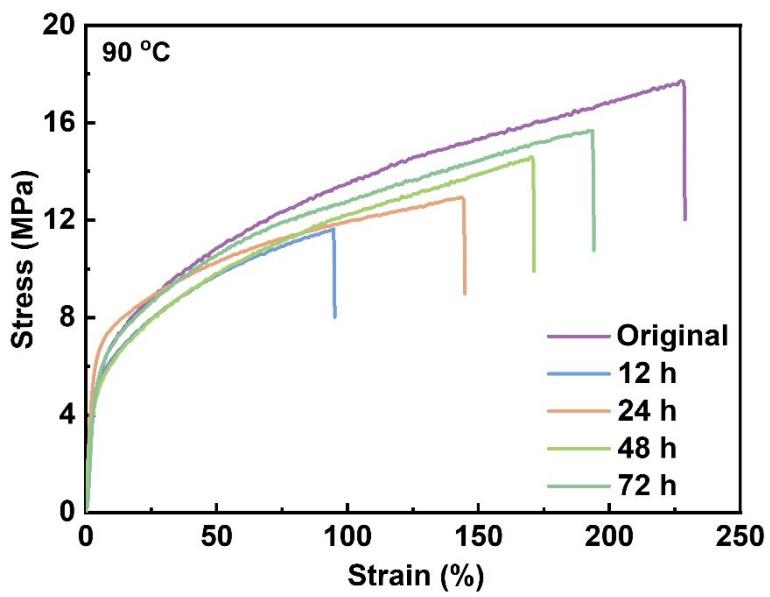


Fig.S16 Stress-strain curves of the IPDI@GO<sub>12.5</sub>/D-PUA at 90 °C for different healing durations (12 h, 24 h, 48 h, and 72 h).

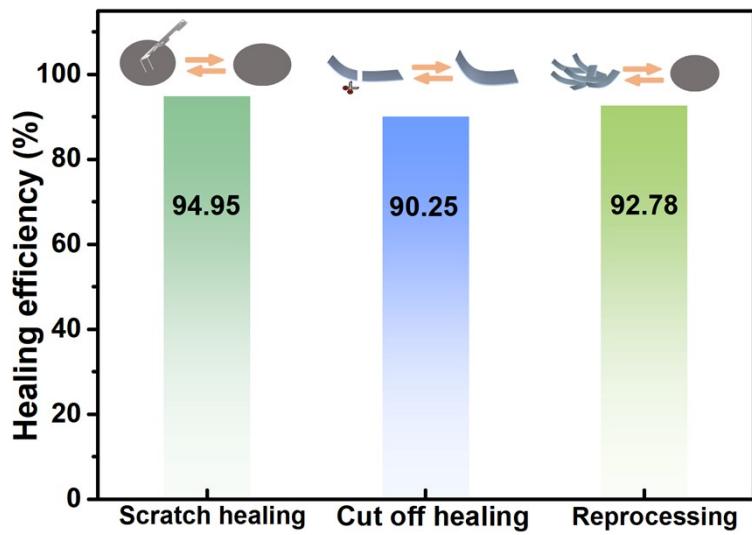


Fig.S17 Healing efficiency of  $\kappa_{\parallel}$  of the IPDI@GO<sub>12.5</sub>/D-PUA after different self-healing processes.

## References

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