

Supplementary Information:

Branched-type nanocluster-junction fluorofluidic-gels for interface-adaptive thermal conduction and mechanical resistance

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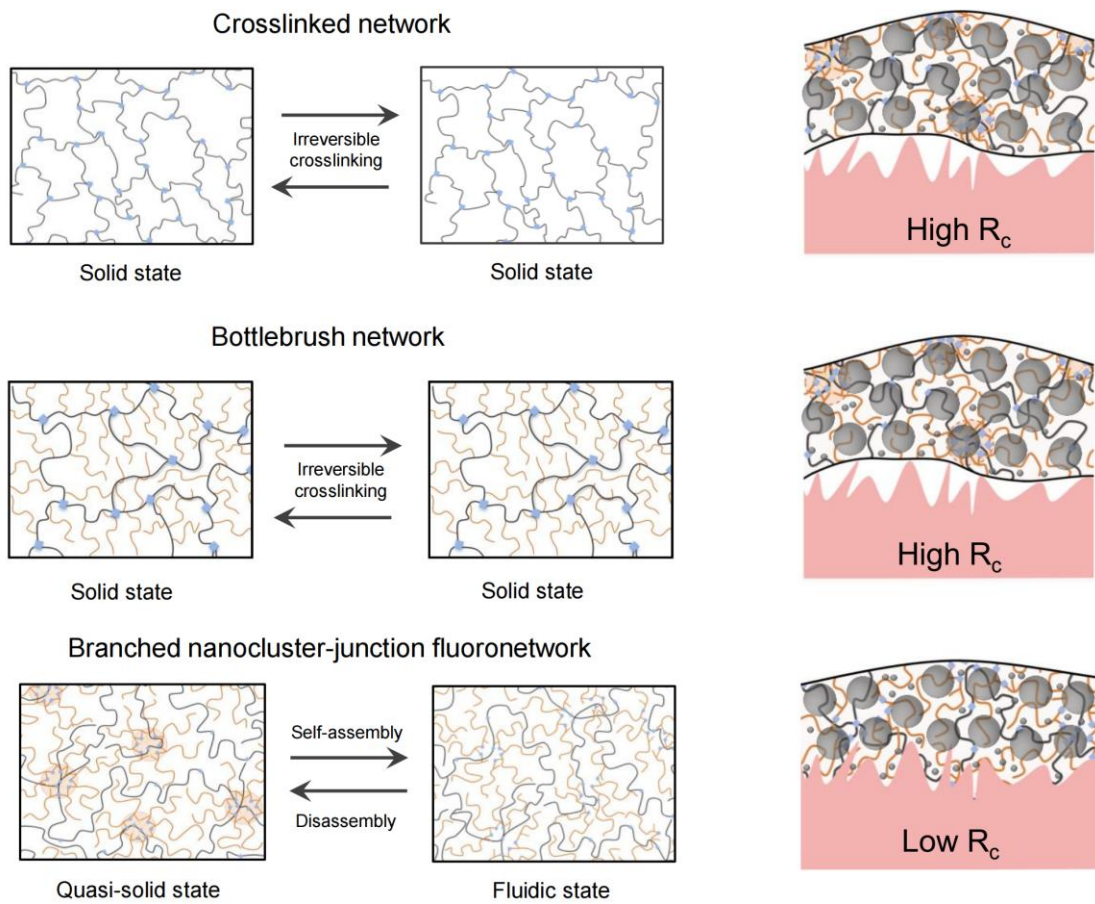
*Corresponding author. Email: zhaoziguang@ucas.ac.cn; stwang@mail.ipc.ac.cn;

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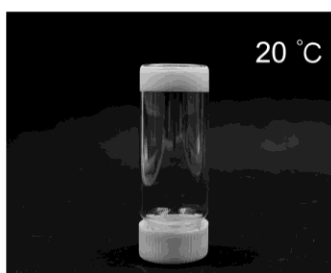
Supplementary Figs. 1-22

Supplementary Table 1

References



Supplementary Fig. 1 | Schematic illustration of diverse polymeric network designs and their corresponding thermal conduction performances.



Quasi-solid state



Fluidic State

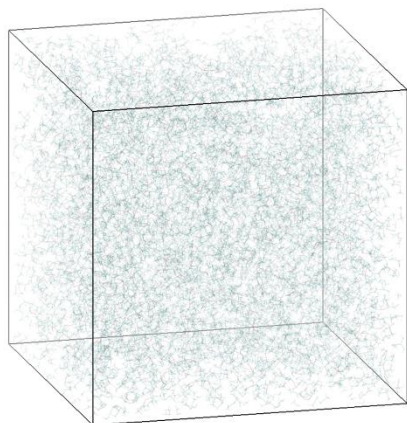
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32 **Supplementary Fig. 2** | TIF_{0.25} under different temperature conditions.

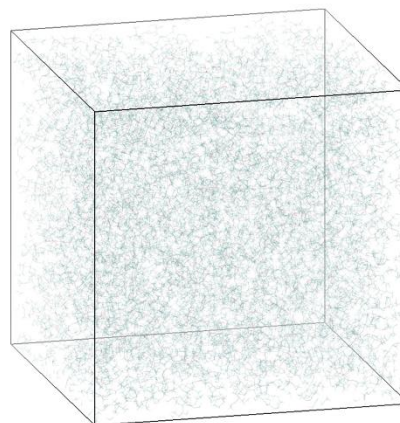
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Polyether network

20 °C

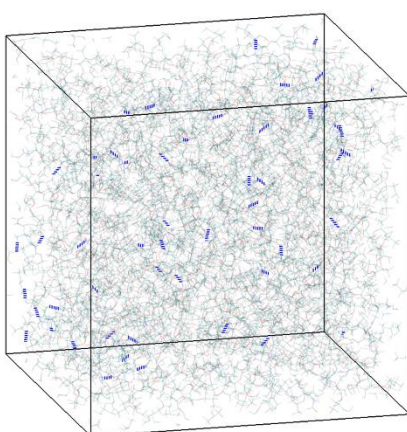


80 °C

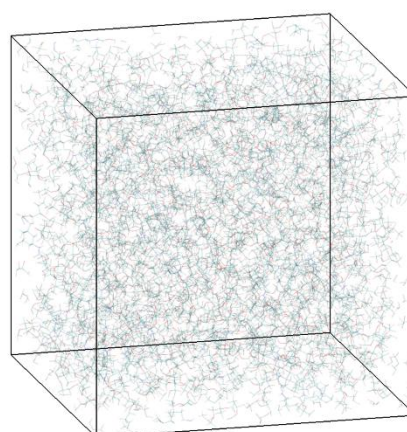


TIF_{0.25}

20 °C

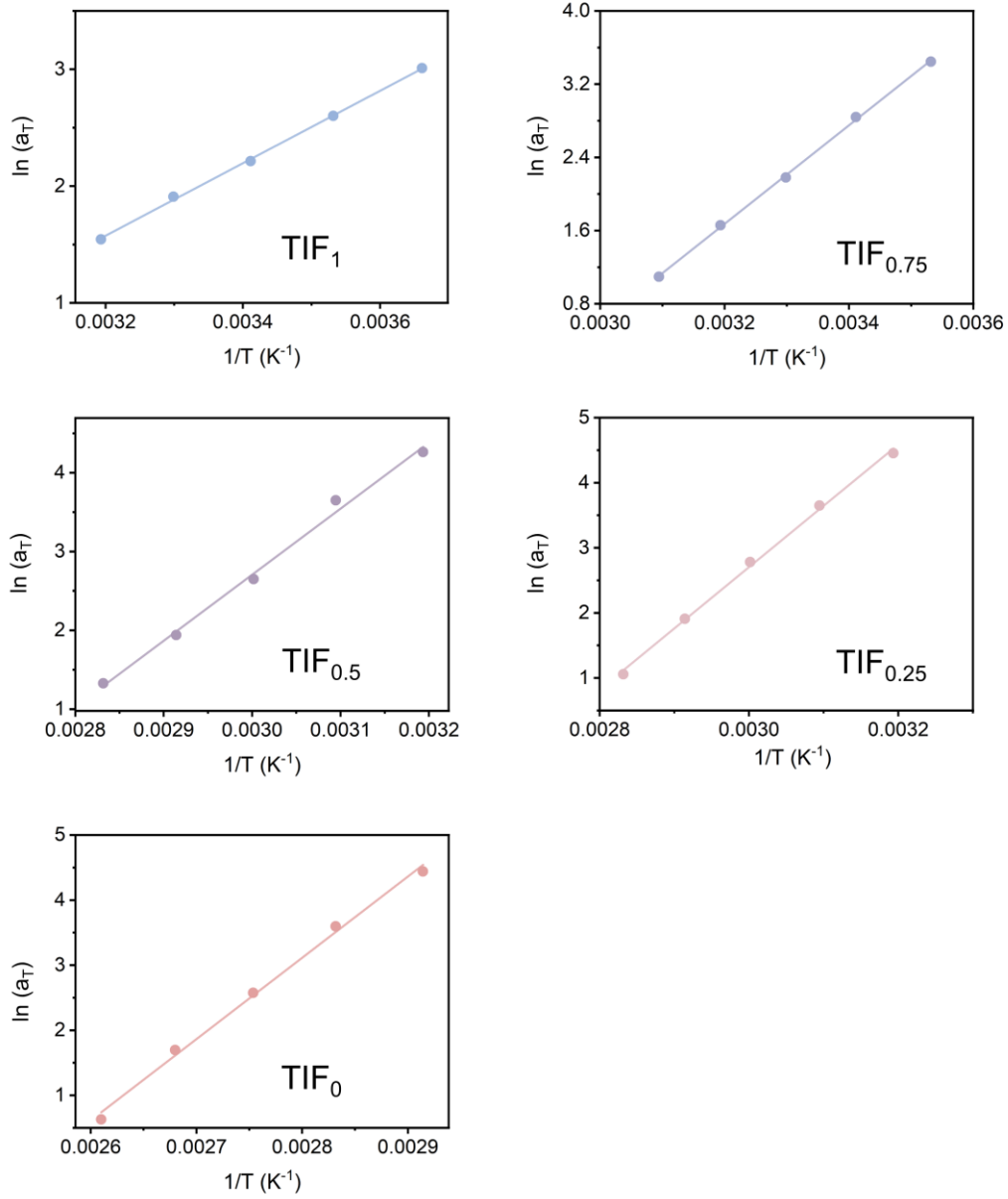


80 °C

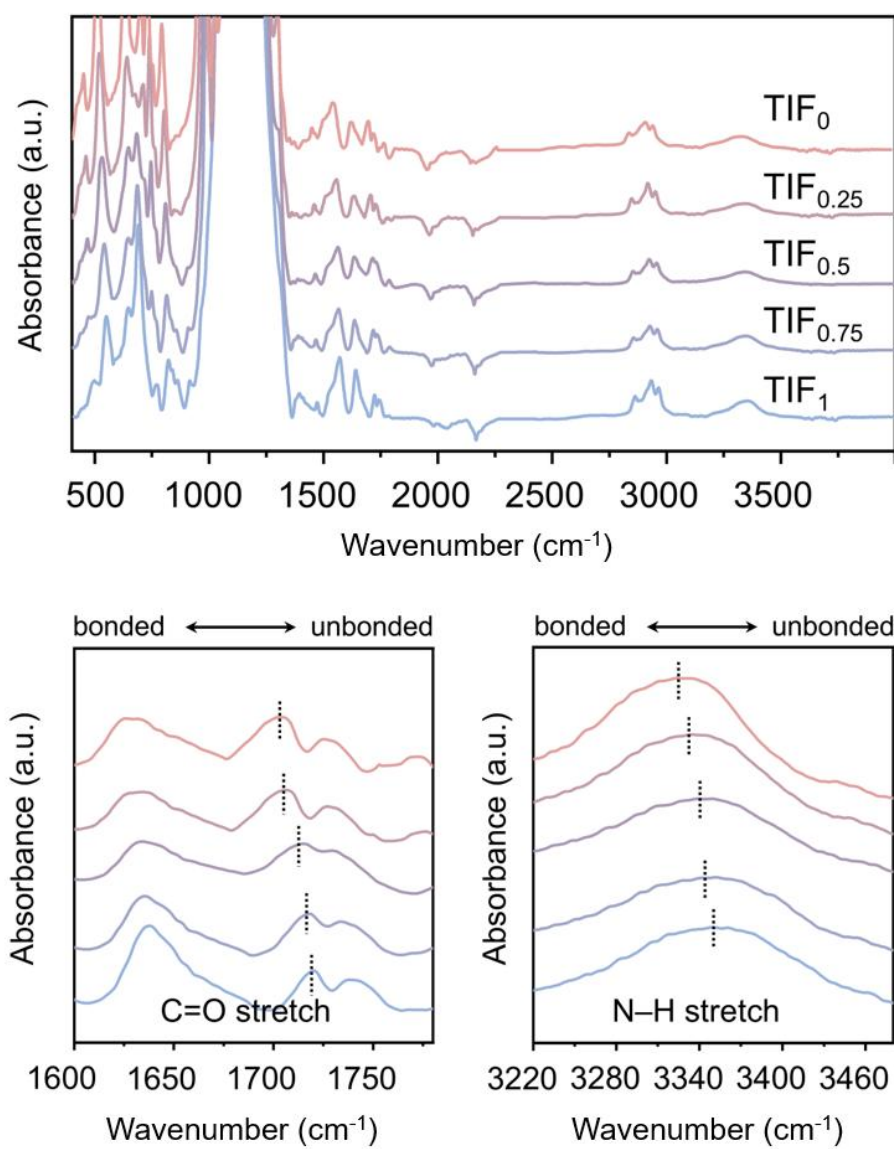


//// Hydrogen Bond

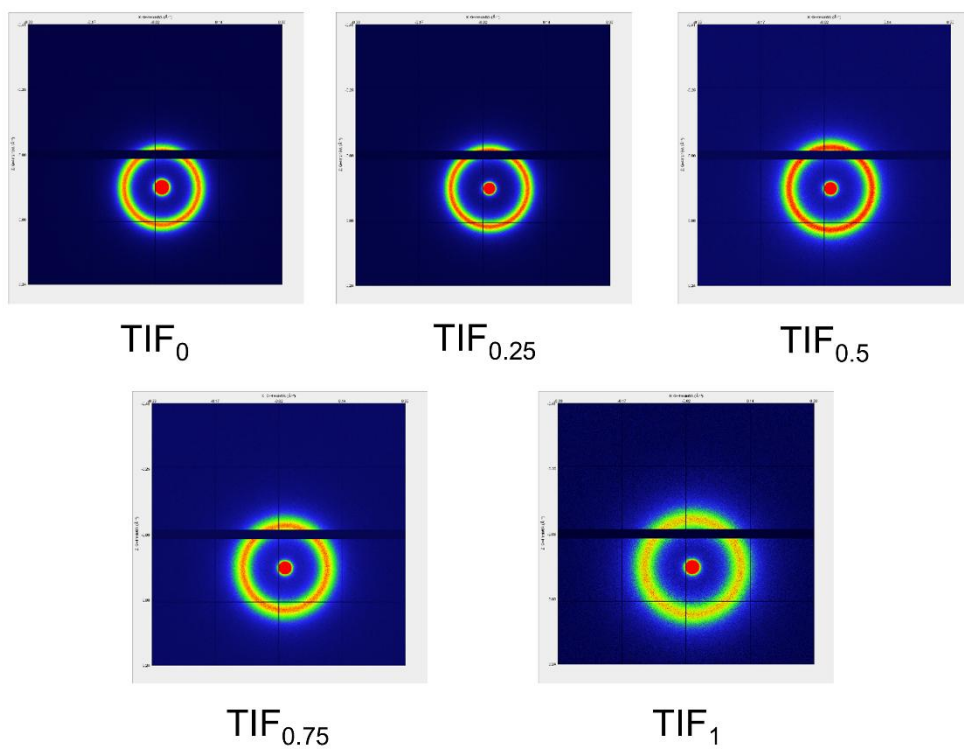
Supplementary Fig. 3 | Molecular dynamics simulation snapshots of the TIF and polyether network.



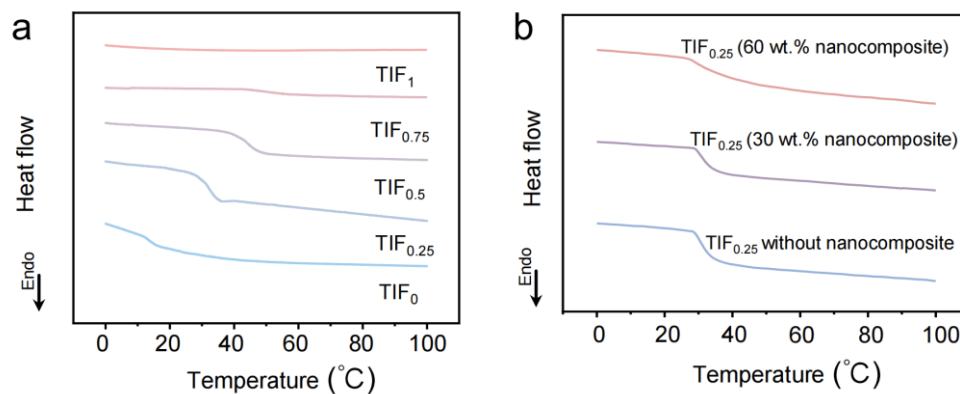
Supplementary Fig. 4 | Time-temperature superposition shift factors with fitted activation energies.



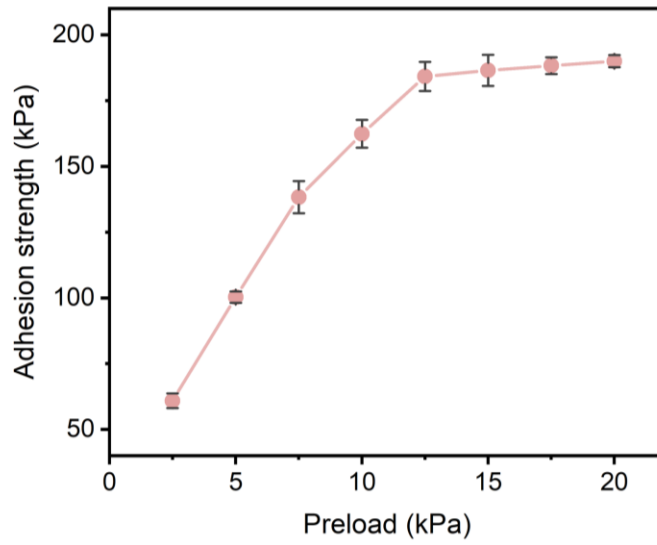
Supplementary Fig. 5 | FTIR spectra of different TIFs vertically offset for clarity.



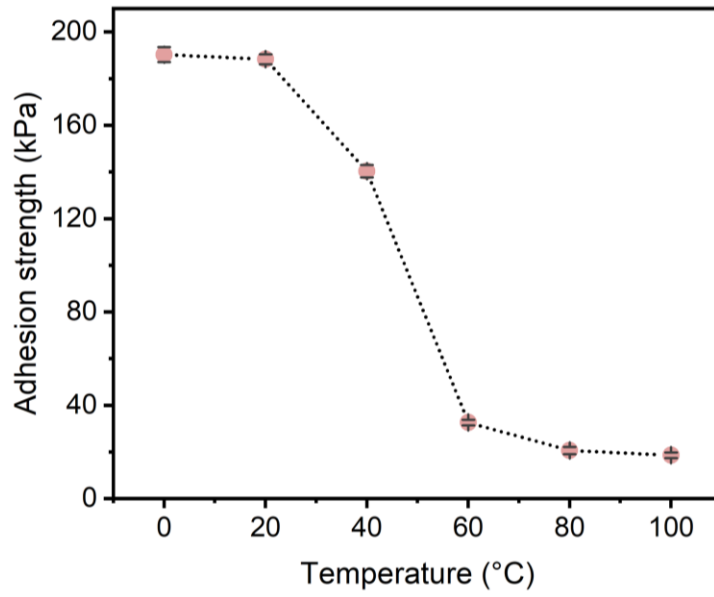
Supplementary Fig. 6 | SAXS two-dimensional maps for different TIFs.



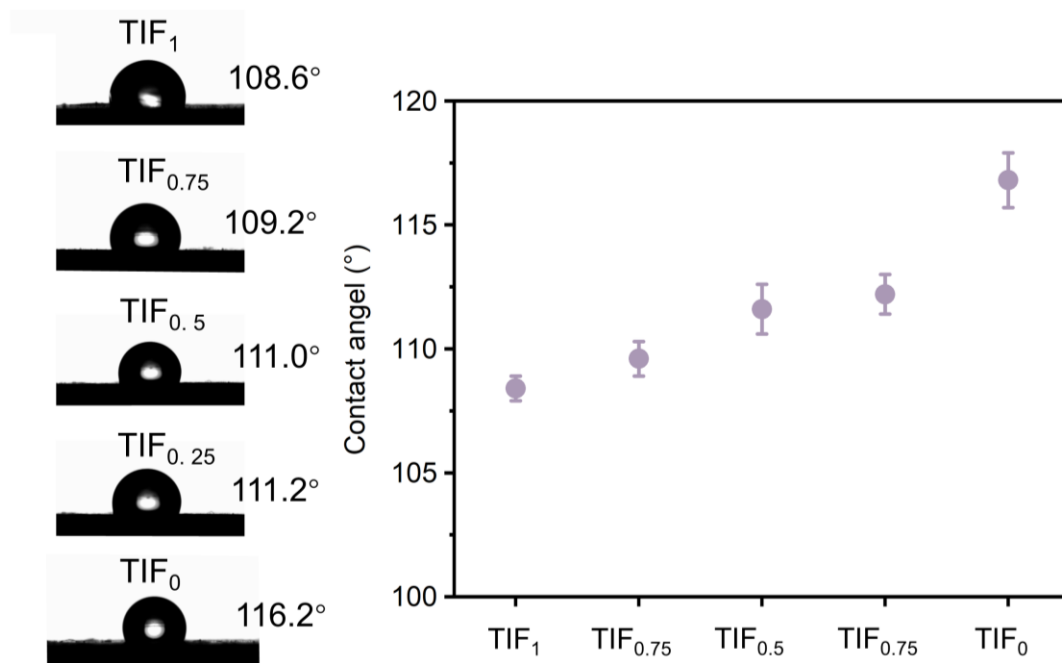
Supplementary Fig. 7 | (a) DSC curves for different TIFs vertically offset for clarity.
(b) DSC curves of TIFs with varied nanocomposite loadings.



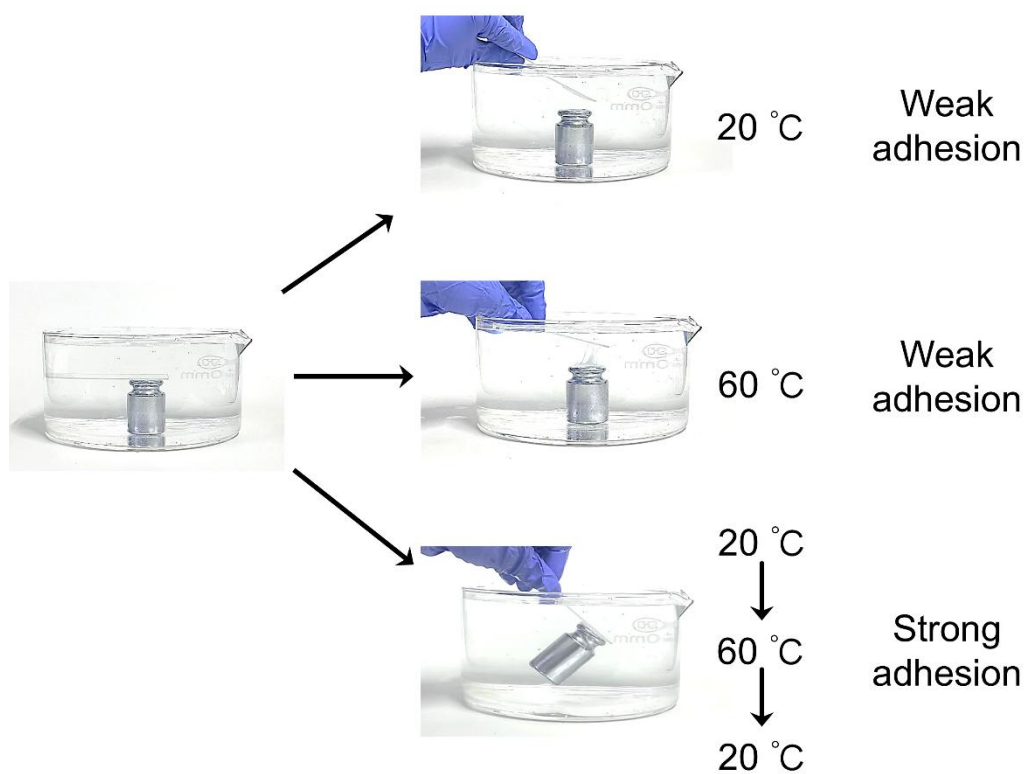
Supplementary Fig. 8 | The adhesion strength of the TIF_{0.25} with different preloads. The error bars on the *y* axis represent the standard deviation of the adhesion strength measured in the adhesion tests (n=3).



Supplementary Fig. 9 | The adhesion strength of the TIF_{0.25} with different temperatures. The error bars on the y axis represent the standard deviation of the adhesion strength measured in the adhesion tests (n=3) with different temperatures.



Supplementary Fig. 10 | Water contact angle of different TIFs. The error bars on the y axis represent the standard deviation of the contact angle measured in the tests (n=3).

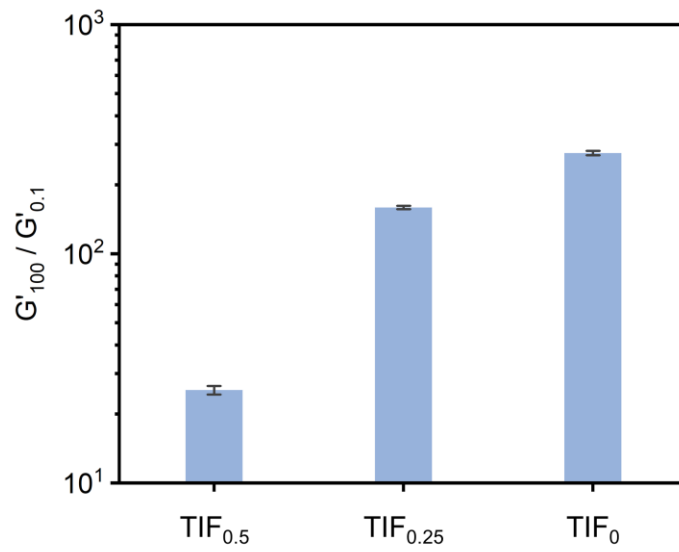


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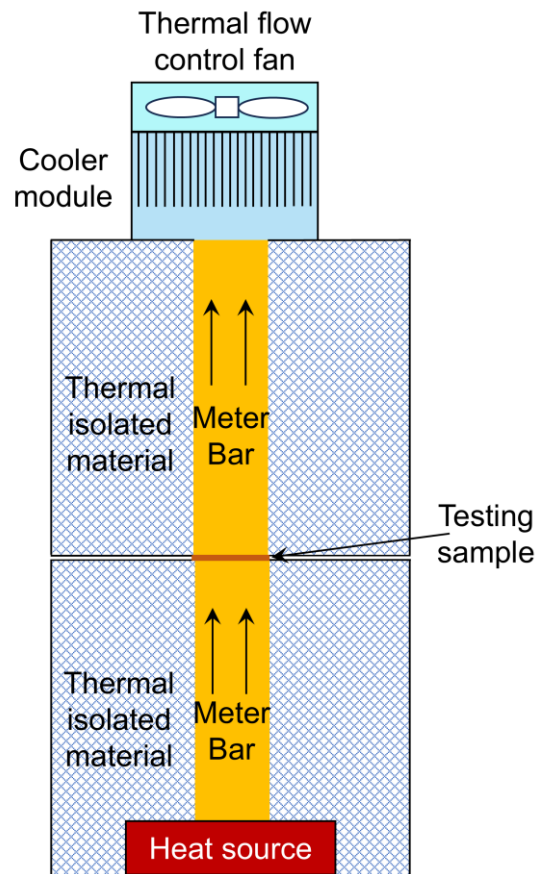
75 **Supplementary Fig. 11** | The underwater adhesion performance of the TIF_{0.25} with
 76 different temperatures.

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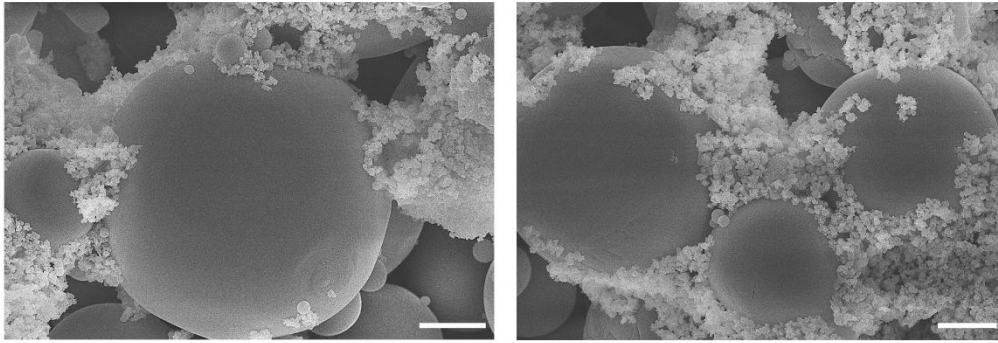
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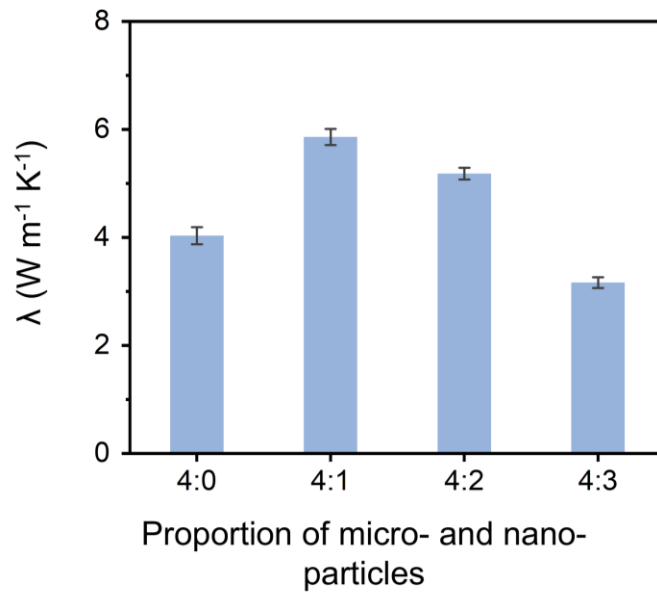
Supplementary Fig. 12 | Impact-stiffening response reflected by the ratio of storage modulus at 100 Hz to that at 0.1 Hz. The error bars on the y axis represent the standard deviation of the ratios of G' with 100 Hz and 0.1 Hz measured in the rheological tests ($n=3$).



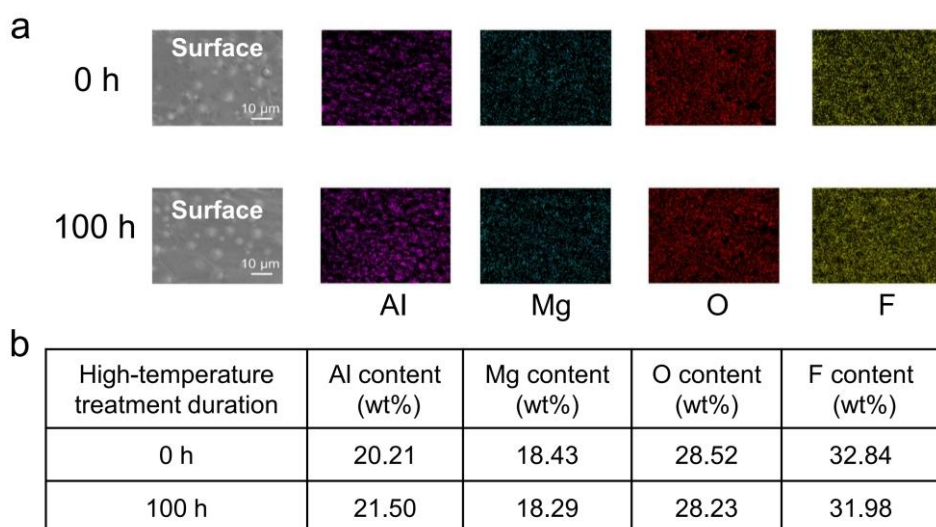
Supplementary Fig. 13 | Schematic of the heat resistance testing device.



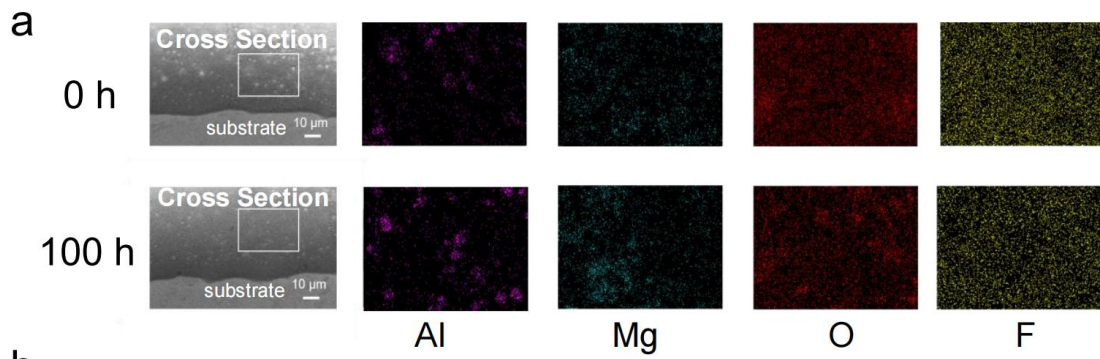
Supplementary Fig. 14 | The SEM images of the Al_2O_3 microparticles and MgO nanoparticles before incorporation into the TIF matrix. Scale bar, 2 μm .



Supplementary Fig. 15 | Out-of-plane thermal conductivity (λ) of the nanoparticle-filled $\text{TIF}_{0.25}$ nanocomposite (60% nanocomposite content) with varying proportions of Al_2O_3 microparticles and MgO nanoparticles. The error bars on the y axis represent the standard deviation of the thermal conductivity measured in the thermal property tests (n=3).



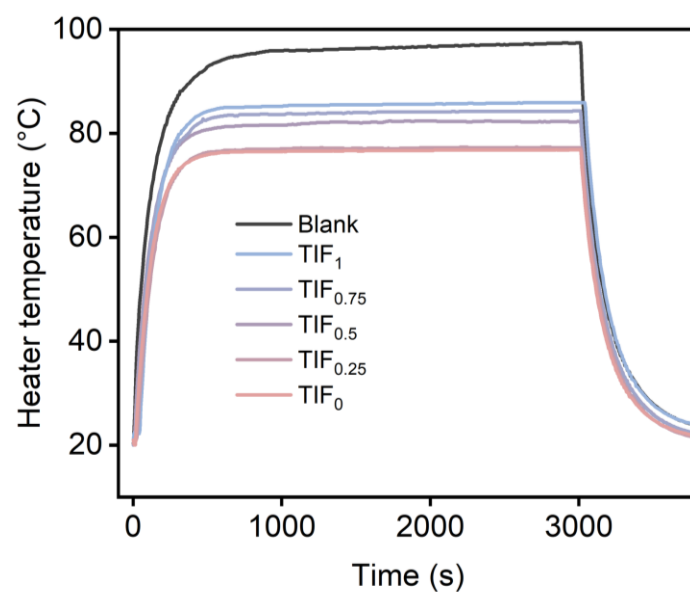
Supplementary Fig. 16 | (a) The SEM image of the surface view of $\text{TIF}_{0.25}$ after continuous heat treatment at 80 °C for different durations, and the corresponding EDS element distribution diagrams. (b) The mass contents of Al, Mg, O, and F calculated from the EDS elemental distribution map of the surface view of $\text{TIF}_{0.25}$ after long-term high-temperature treatment at 80 °C for different durations.



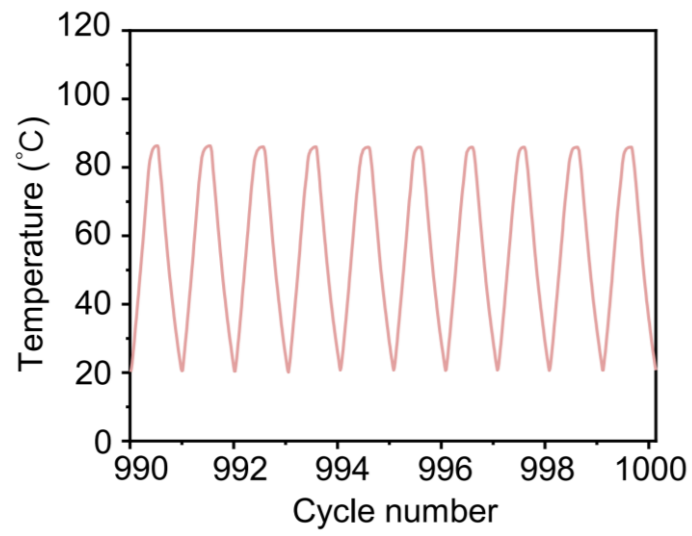
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High-temperature treatment duration	Al content (wt%)	Mg content (wt%)	O content (wt%)	F content (wt%)
0 h	14.33	15.39	32.46	37.82
100 h	15.85	16.34	32.00	35.81

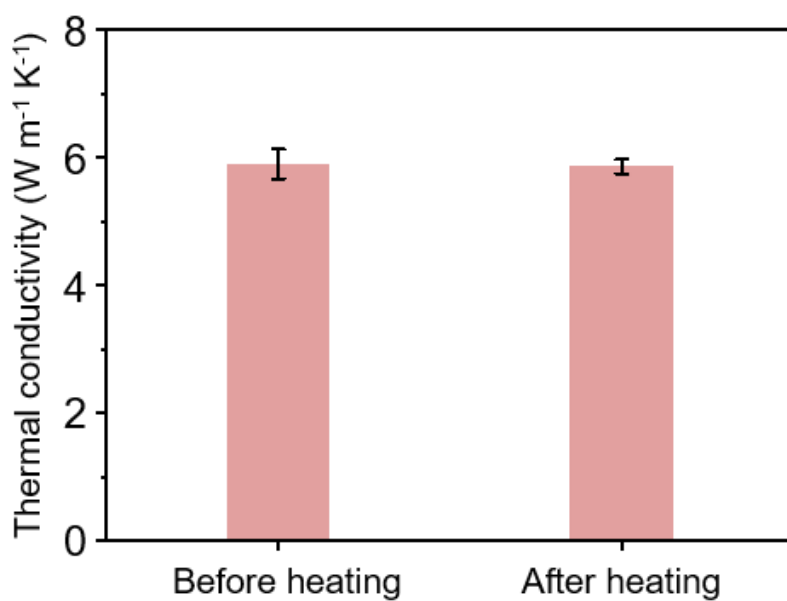
Supplementary Fig. 17 | (a) The SEM image of the cross section of TIF_{0.25} after continuous heat treatment at 80 °C for different durations, and the corresponding EDS element distribution diagrams. (b) The mass contents of Al, Mg, O, and F calculated from the EDS elemental distribution map of the cross section of TIF_{0.25} after long-term high-temperature treatment at 80 °C for different durations.



Supplementary Fig. 18 | Temperature evolution of different TIFs as a function of heating time at an applied heating power of 150 W.



Supplementary Fig. 19 | Temperature variation of TIF_{0.25} from the 990th to the 1000th heating cycles.



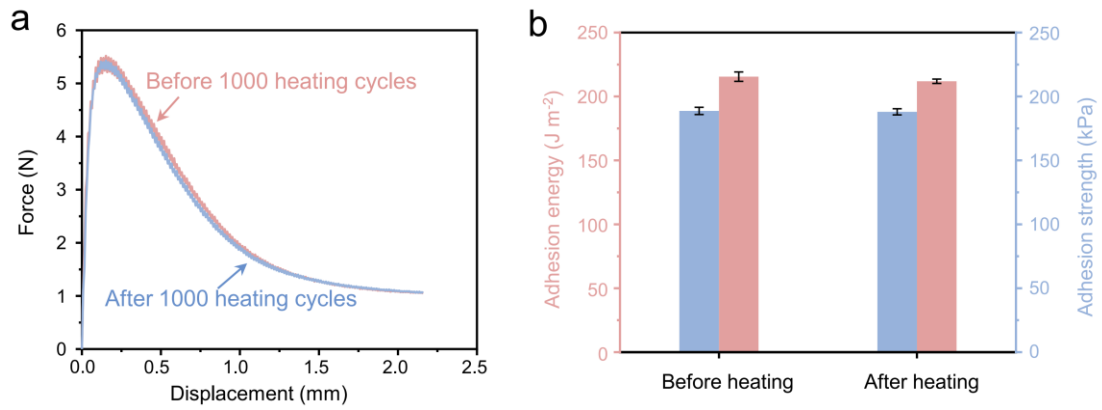
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131 **Supplementary Fig. 20** | Thermal conductivity of the TIF_{0.25} before and after 1000
132 heating cycles. The error bars on the *y* axis represent the standard deviation of the
133 thermal conductivity measured in the thermal property tests (n=3).

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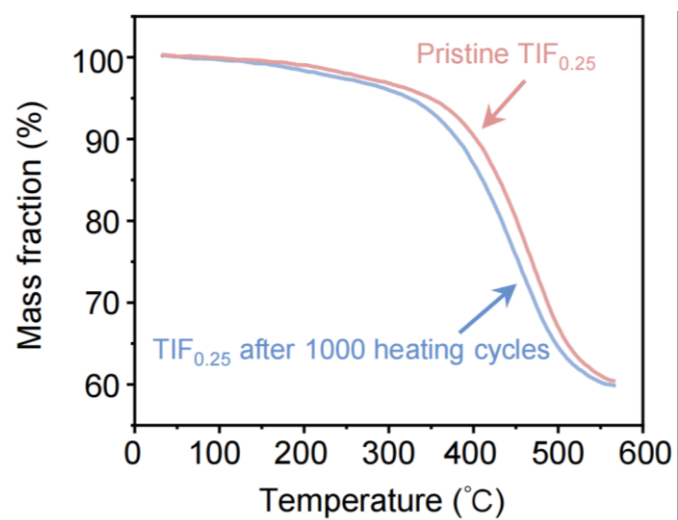
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Supplementary Fig. 21 | Force-displacement curves, adhesion strength and adhesion energy of the TIF_{0.25} before and after 1000 heating cycles. The error bars on the y axis represent the standard deviation of the adhesion energy and strength measured in the adhesion tests (n=3).



Supplementary Fig. 22 | The TGA analyses of pristine TIF_{0.25} and after 1000 heating cycles.

Supplementary Table 1. Thermal conductivity and contact thermal resistance of different insulating thermal interface materials

Type of insulating thermal interface materials	λ (W m ⁻¹ K ⁻¹)	R _c (cm ² K W ⁻¹)	Reference
Thermal Interface Grease	4.2	0.27	1
	4.7	0.18	1
	7.5	0.226	2
	2	0.379	2
	0.6	0.494	2
	4.38	0.243	3
	1.438	0.623	4
	4.4	0.52	5
Thermal Interface Pad	5	1.17	6
	5	0.63	7
	4.09	1.03	8
	7.04	1.43	8
	2.5	1.419	2
	3.5	2.065	2
Phase Transition TIM	2.5	2	9
	2.1	2.2	10
	1.23	0.5	11
	0.93	0.2	12
This work	5.89	0.15	\

References

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