Electronic Supplementary Material (ESI) for Materials Horizons.
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Supplementary Information for

Highly oriented BN-based TIMs with high through-plane thermal conductivity and low compression modulus

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Supplementary Figures

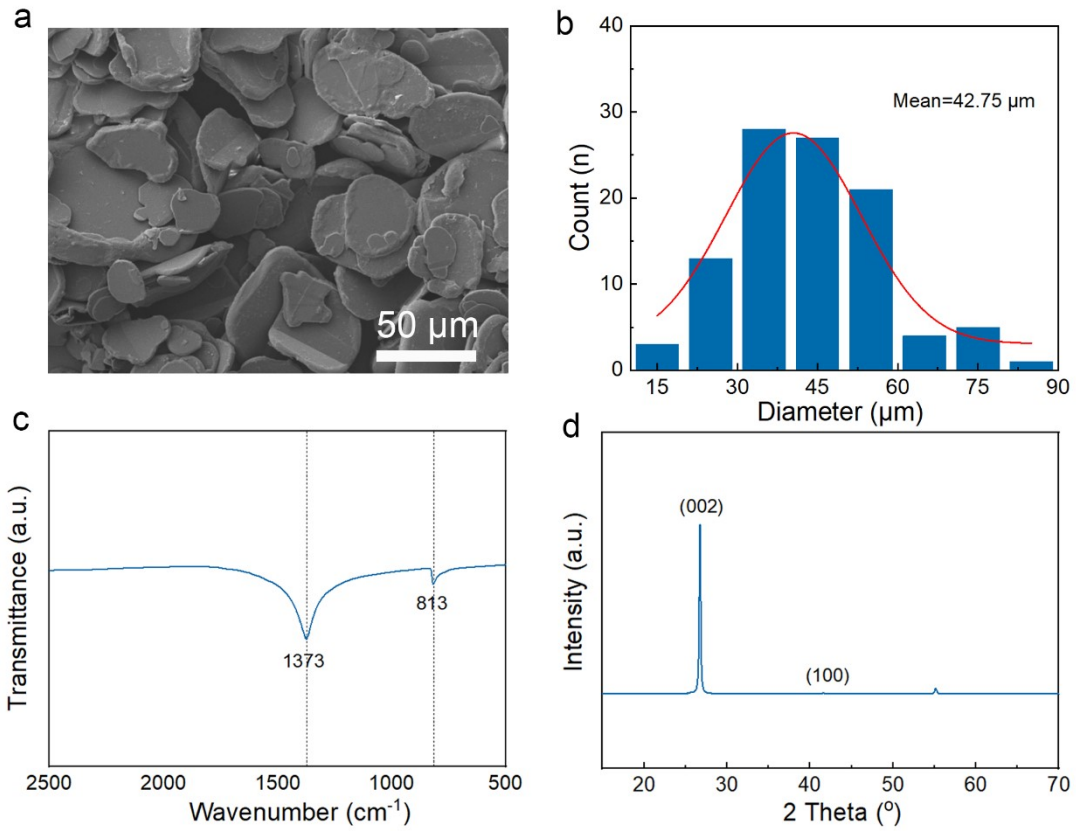

Fig. S1 (a) SEM image of h-BN (PT110). (b)Size distribution of h-BN (PT110). (c) FTIR spectra of h-BN (PT110). (d) XRD pattern of h-BN (PT110).

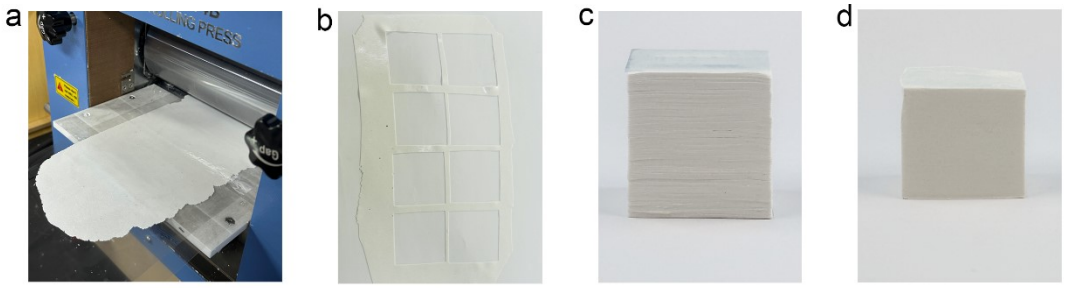

Fig. S2 (a) The process of horizontal orientation. (b) BN/PDMS film after cutting. (c) Stacked BN/PDMS film. (d) Densified BN/PDMS block.

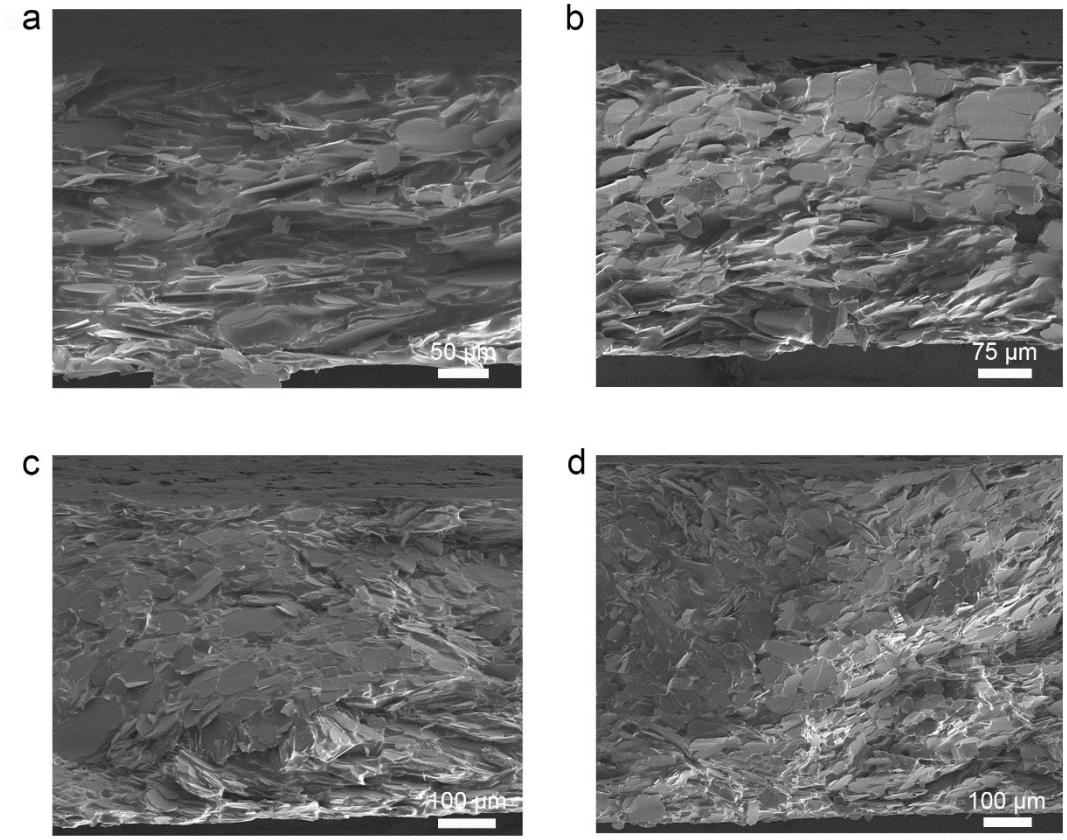

Fig. S3 (a) SEM images of BN/PDMS film with varying thicknesses (0.3, 0.4, 0.55 and 0.75 mm)

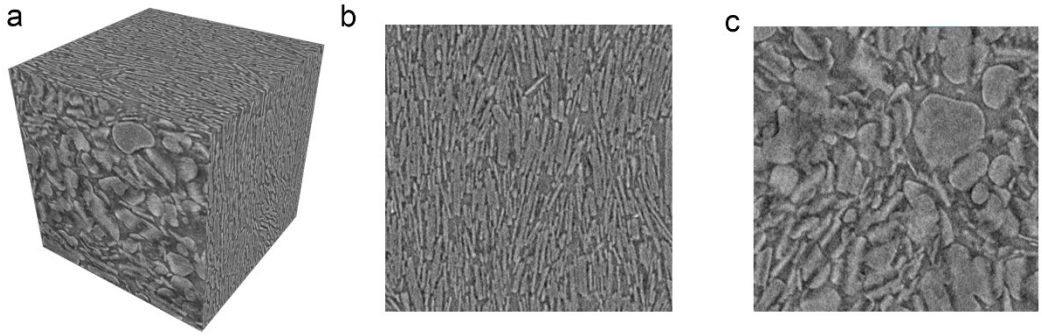

Fig. S4 (a) The original Micro-CT images of Pad-75-V. (b) Screenshot of XZ plane. (c) Screenshot of YZ plane.

Fig. S5 (a) The azimuthal angel (φ) plots of Pad-75-V. (b) The azimuthal angel (φ) plots of Pad-75-R.

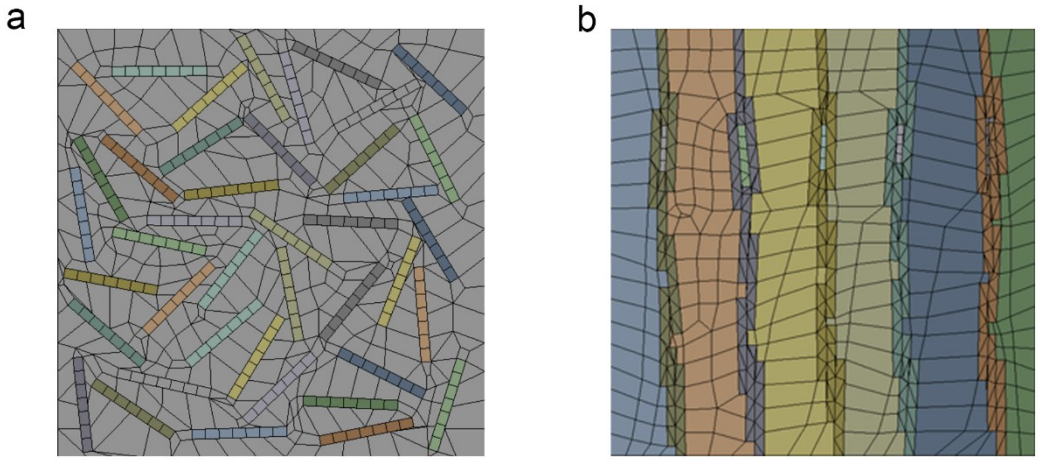

Fig. S6 The corresponding grid divisions of ANSYS models.

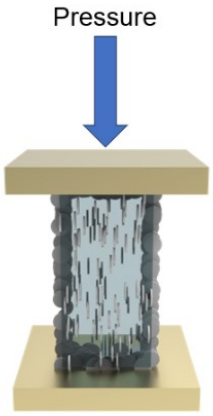

Fig. S7 Schematic diagram of compression direction.

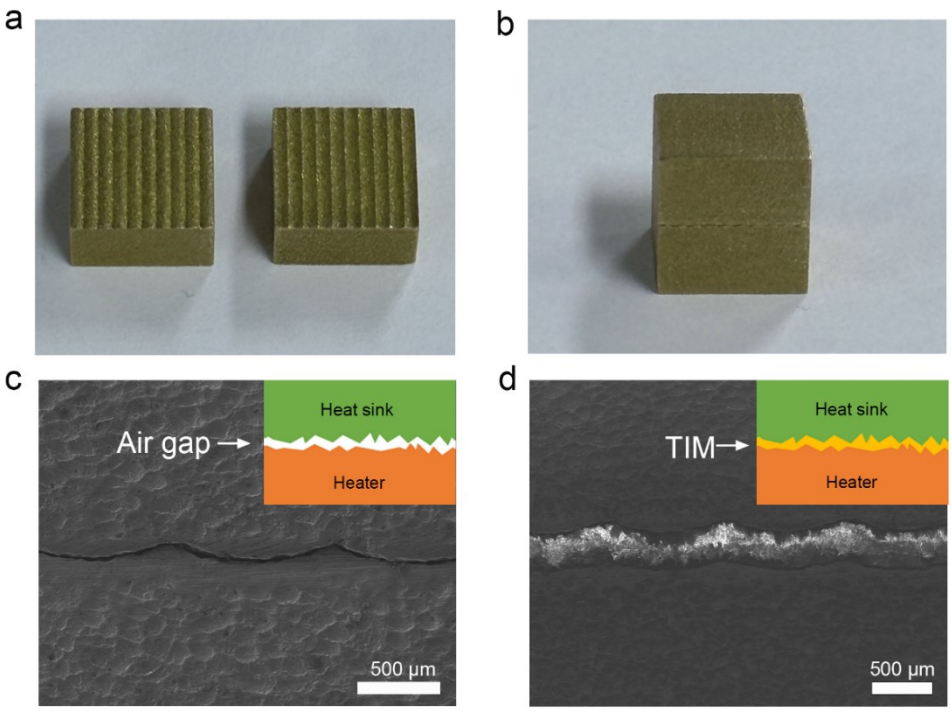

Fig. S8 (a), (b) The optical images of customized molds. (c), (d) Filling ability of Pad-75-V.

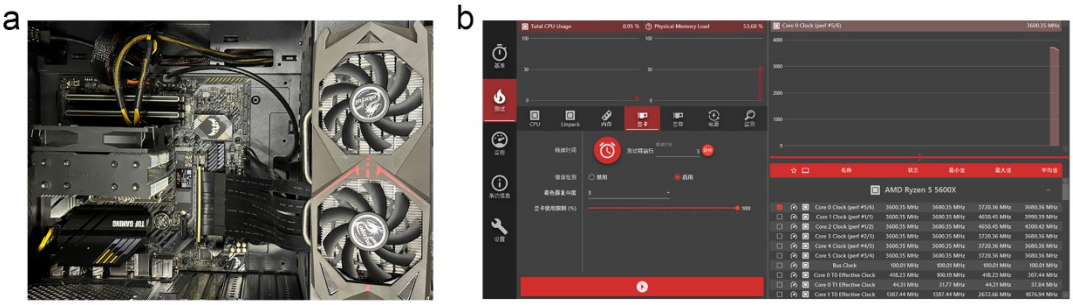

Fig. S8 (a) Device of TIM performance test in graphics card (GTX 1060ti).
(b) The running page of the OCCT software.

Table S1 Through-plane thermal conductivity of BN/BNNS based composites

Materials	Method	Loading	Through-plane TC ($W\ m^{-1}\ K^{-1}$)	References
BN/epoxy	Blending	10 wt%	0.57	[1]
BN/epoxy	Blending	20 wt%	0.64	[2]
BN/epoxy	Blending	30 wt%	1.17	[3]
BN/PI	Blending	40 wt%	0.74	[4]
BNNS/PI	Blending	40 wt%	0.67	[5]
BN/epoxy	Magnetic field	50 wt%	3.59	[6]
BN/PDMS	Micro-fluidic	9.85 vol%	6.40	[7]
BNNS/PDMS	Wet-spinning	27.05 vol%	5.13	[8]
BN/MVQ	Stacking-cutting	54 vol%	6.34	[9]
BN/PVDF	Pressing-cutting	30 wt %	3.5	[10]
BN/SR	Rolling-cutting	60 wt%	7.62	[11]
BN/SG	Micro-fluidic	60 wt%	5.65	[12]
BN/PDMS	Rolling-cutting	60 wt%	6.59	This work
BN/PDMS	Rolling-cutting	65 wt%	7.77	This work
BN/PDMS	Rolling-cutting	70 wt%	9.22	This work
BN/PDMS	Rolling-cutting	75 wt%	12.11	This work

Table S2 Measured density, thermal diffusivity, and heat capacity of BN/PDMS composites with various BN contents with vertically aligned structure at room temperature.

Samples	Density ($g\ cm^{-3}$)	Thermal diffusivity ($mm^2\ s^{-1}$)	Heat capacity ($J\ kg^{-1}\ K^{-1}$)
Pad-60-V	1.49	3.88	1.14
Pad-65-V	1.57	4.67	1.06
Pad-70-V	1.65	5.48	1.02
Pad-75-V	1.70	7.20	0.99

Table S3 Compression modulus of reported BN/BNNS composites.

Materials	Through-plane TC ($W\ m^{-1}\ K^{-1}$)	Compression modulus (kpa)	References
BN/PU	11.5	150	[13]
LM-BN/PDMS	4.3	193	[14]
GNP-BN/PDMS	1.16	1470	[15]
BN/PDMS	6.4	2200	[7]
BN/PDMS	5.65	2350	[12]
BNNS/Epoxy	6.07	4000	[16]
BN/PDMS	12.11	55	This work

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