

Electronic Supporting Information (ESI)

A small amount of delaminated Ti_3C_2 flakes to greatly enhance the thermal conductivity of boron nitride papers by assembling the well-designed interface

*Xianwu Huang^a and Peiyi Wu^{*a,b}*

^aState Key Laboratory of Molecular Engineering of Polymers, Department of Macromolecular Science, Fudan University, Shanghai 200433, P. R. China

^bState Key Laboratory for Modification of Chemical Fibers and Polymer Materials, College of Chemistry, Chemical Engineering and Biotechnology, Center for Advanced Low-Dimension Materials, Donghua University, Shanghai 201620, China

E-mail: peiyiwu@fudan.edu.cn

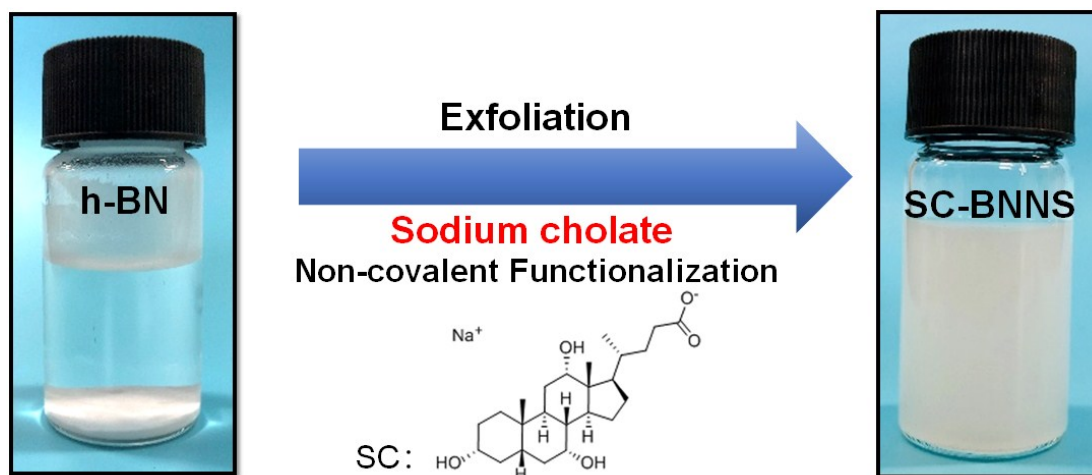


Fig. S1. The illustration of the exfoliation and SC non-covalent functionalization process of BNNS.

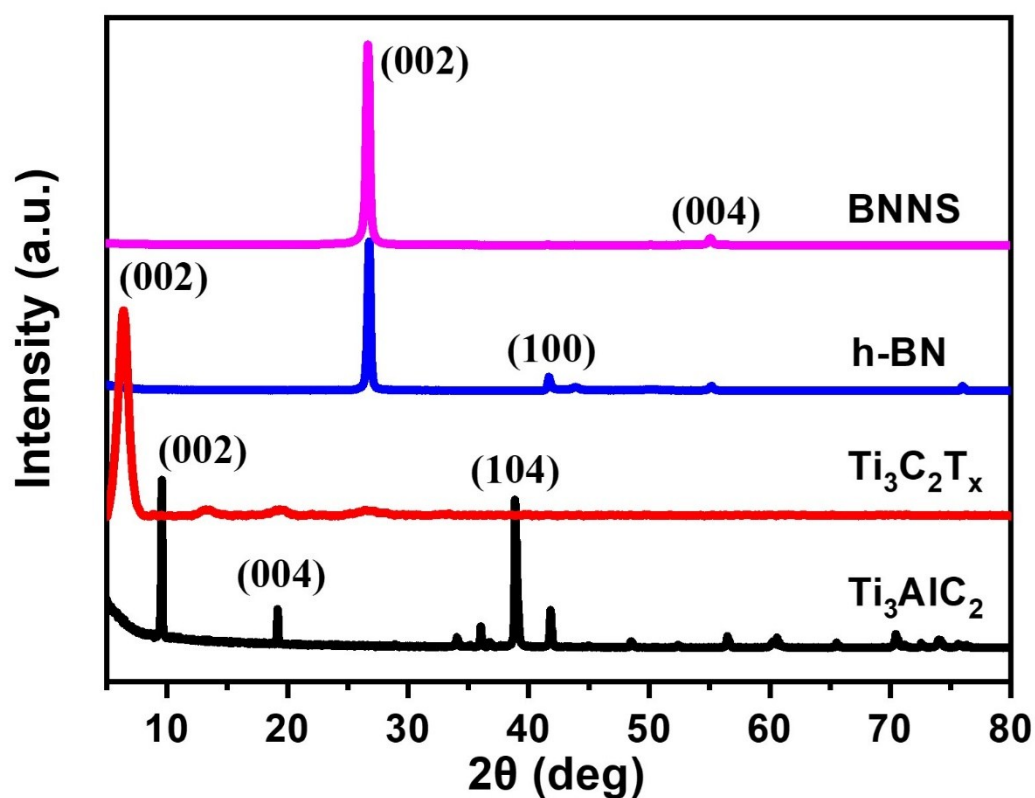


Fig. S2. XRD patterns of BNNS, h-BN, $\text{Ti}_3\text{C}_2\text{T}_x$, and Ti_3AlC_2 .

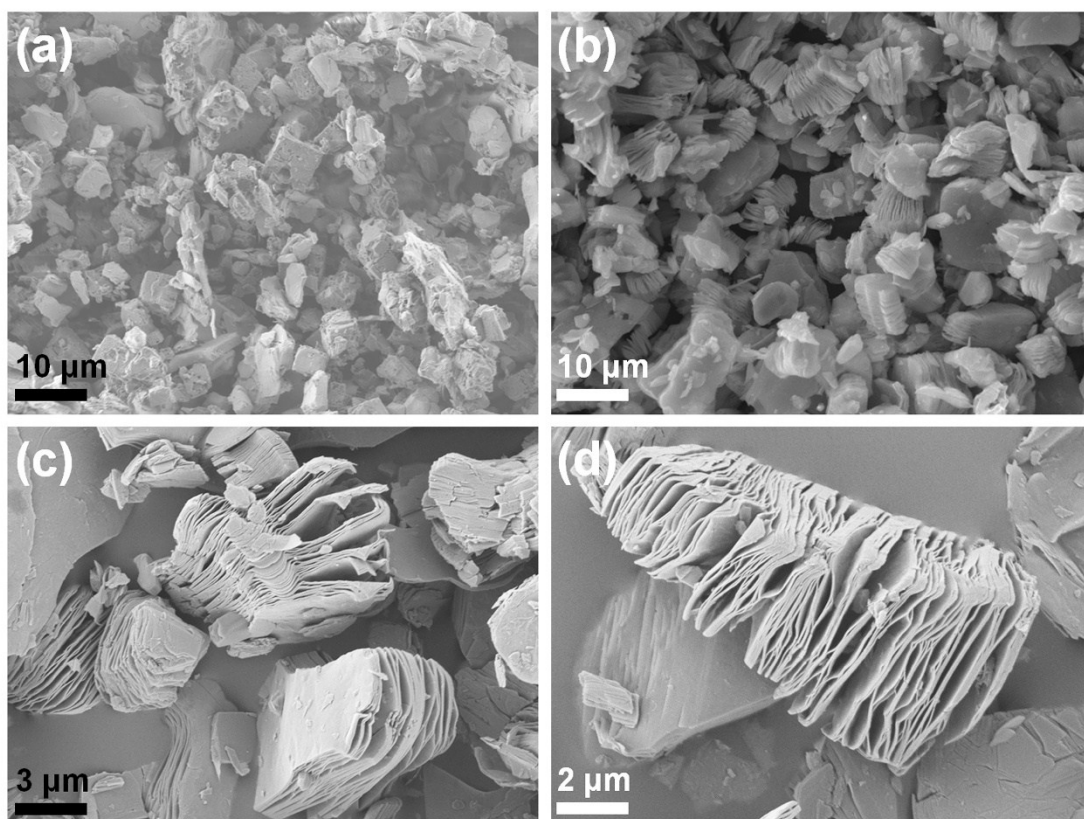


Fig. S3. SEM of MAX (a)before and (b, c, and d) after etching Al atom layers.

Table S1. ζ -potential of exfoliated BNNSs and MXene in DI water.

	BNNSs	MXene
ζ -potential/mV	-20.1	-39.0

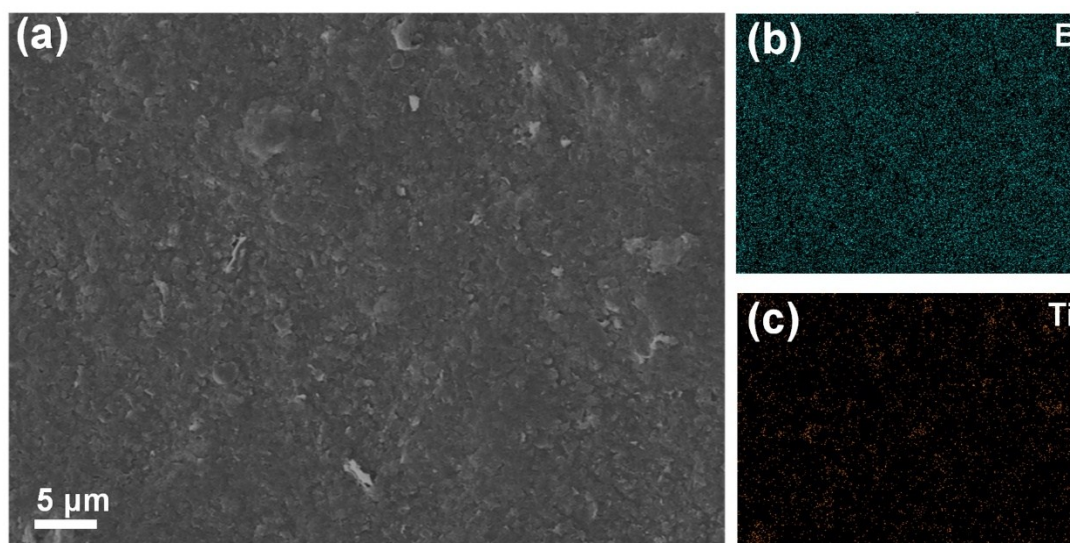


Fig. S4. (a) SEM of the surface of BM-5 film (b, c) and corresponding elemental mappings of B and Ti.

Table S2. ρ , α , c , and calculated λ of BNNS/MXene composite films.

Samples	$\alpha/ \text{mm}^2 \text{s}^{-1}$	$\rho/ \text{g cm}^{-3}$	$c/ \text{J g}^{-1} \text{K}^{-1}$	$\lambda/ \text{W m}^{-1} \text{K}^{-1}$
In-plane				
BM-0	12.81 ± 0.46	1.950	1.08	26.98 ± 0.96
BM-1	15.42 ± 0.61	1.957	1.09	32.89 ± 1.30
BM-5	24.07 ± 0.59	1.963	1.11	52.45 ± 1.29
BM-13	18.96 ± 0.61	1.983	1.16	43.61 ± 1.41
BM-17	14.67 ± 0.33	1.994	1.20	35.10 ± 0.80
BM-25	13.66 ± 0.25	2.016	1.25	34.42 ± 0.62
BM-100	13.43 ± 0.20	2.214	1.09	32.41 ± 0.48
Out-of-plane				
BM-0	2.25 ± 0.1	1.950	1.08	4.74 ± 0.21
BM-5	1.8 ± 0.08	1.963	1.11	3.92 ± 0.17
BM-100	1.6 ± 0.09	2.214	1.09	3.86 ± 0.22

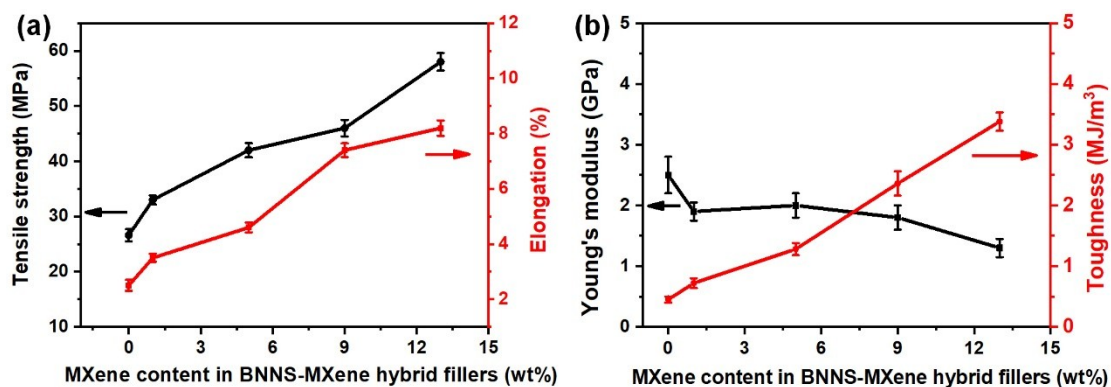


Fig. S5. (a) The tensile strength, elongation, (b) Young's modulus, and toughness of the BM film with different MXene content.

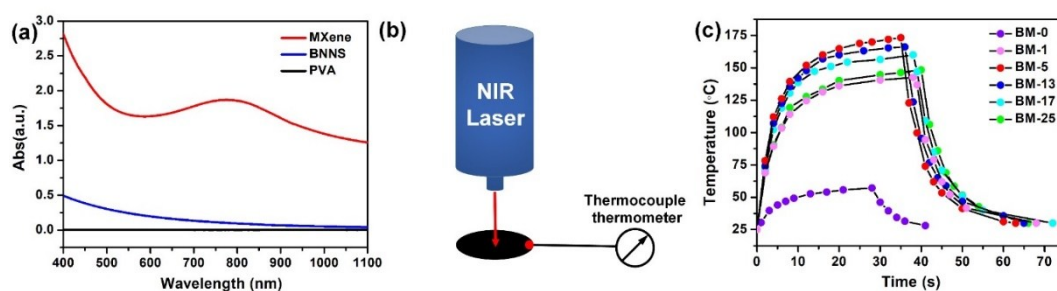


Fig. S6. (a) Ultra violet-visible (UV-Vis) spectrum of MXene, BNNS and PVA in DI water. (b) The facile experiment setup of NIR light-to-heat. (c) The curve of temperature versus time corresponding to NIR light-to-heat experiment.

Table S3 Comparisons between the TCs and the preparation methods of BNNS/MXene composite film and typical hBN or BNNS-based composites ^a

Matrix	Fillers	Fraction of filler	TC (W m ⁻¹ K ⁻¹)	Preparation methods	Year ^{Ref.}
BNNS	GO	5.0 wt%	29.8	Vacuum filtration	2016 ¹
PVA	BNNS	94 wt%	6.9	Vacuum filtration	2015 ²
PVA	BNNS/GO	98.4wt% (BNNS:GO=80:20)	11.9	Vacuum filtration	2017 ³
PVA	BNNS- Ag/SiCN W-Ag	95wt% (BNNS-Ag: SiCNW- Ag=85:10)	21.7	Vacuum filtration	2016 ⁴
PMMA	BNNS	80 wt%	11	Evaporation	2016 ⁵
EP	BNNS	44 vol%	9.0	Evaporation	2017 ⁶
EP	BNNS-Ag	25.1 vol%	12.6	Evaporation	2016 ⁷
CNF	BNNS	70 wt%	30.3	Vacuum filtration	2017 ⁸
PS	BNNS	13.4 vol%	8.0	Hot pressing	2017 ⁹
TPU	BNNS	95 wt%	50.3	Hot pressing	2018 ¹⁰
/	BNNS	100 wt%	51.1	Vacuum filtration	2017 ¹¹
PVA	BNNS- PDA	70 wt%	24.6	Vacuum filtration	2019 ¹²
BNNS	Graphene	5 wt%	63.5	Vacuum filtration	2019 ¹³
PVA	BNNS- MXene	80wt% (BNNS:MXene=95:5)	52.4	Vacuum filtration	This work

^a BNNS represents boron nitride nanosheets; BNNS-Ag means boron nitride nanosheets decorated with Ag nanoparticles; PVA represents poly (vinyl alcohol); SiCNW represents silicon carbide nanowires; PMMA represents polymethyl methacrylate; EP represents epoxy resin; CNF represents cellulose nanofiber; PS represents polystyrene; TPU represents thermoplastic polyurethane; PBI represents Polybenzimidazole; PDA represents polydopamine.

References

1. Y. Yao, X. Zeng, F. Wang, R. Sun, J.-b. Xu and C.-P. Wong, Significant Enhancement of Thermal Conductivity in Bioinspired Freestanding Boron Nitride Papers Filled with Graphene Oxide, *Chem. Mater.*, 2016, **28**, 1049-1057.
2. X. Zeng, L. Ye, S. Yu, H. Li, R. Sun, J. Xu and C. P. Wong, Artificial nacre-like papers based on noncovalent functionalized boron nitride nanosheets with excellent mechanical and thermally conductive properties, *Nanoscale*, 2015, **7**, 6774-6781.
3. P. Li, H. Shen, Z. Qian, X. Yang, N. Zhao, C. Zhu and J. Xu, Facile fabrication of flexible layered GO/BNNS composite films with high thermal conductivity, *J. Mater. Sci.*, 2017, **53**, 4189-4198.
4. Y. Yao, X. Zeng, R. Sun, J. B. Xu and C. P. Wong, Highly Thermally Conductive Composite Papers Prepared Based on the Thought of Bioinspired Engineering, *ACS Appl. Mater. Interfaces*, 2016, **8**, 15645-15653.
5. T. Morishita and H. Okamoto, Facile Exfoliation and Noncovalent Superacid Functionalization of Boron Nitride Nanosheets and Their Use for Highly Thermally Conductive and Electrically Insulating Polymer Nanocomposites, *ACS Appl. Mater. Interfaces*, 2016, **8**, 27064-27073.
6. C. Yu, J. Zhang, Z. Li, W. Tian, L. Wang, J. Luo, Q. Li, X. Fan and Y. Yao, Enhanced through-plane thermal conductivity of boron nitride/epoxy composites, *Composites Part A*, 2017, **98**, 25-31.
7. F. Wang, Y. Yao, X. Zeng, T. Huang, R. Sun, J. Xu and C.-P. Wong, Highly thermally conductive polymer nanocomposites based on boron nitride nanosheets decorated with silver nanoparticles, *RSC Adv.*, 2016, **6**, 41630-41636.
8. K. Wu, J. Fang, J. Ma, R. Huang, S. Chai, F. Chen and Q. Fu, Achieving a Collapsible, Strong, and Highly Thermally Conductive Film Based on Oriented Functionalized Boron Nitride Nanosheets and Cellulose Nanofiber, *ACS Appl. Mater. Interfaces*, 2017, **9**, 30035-30045.
9. X. Wang and P. Wu, Preparation of Highly Thermally Conductive Polymer Composite at Low Filler Content via a Self-Assembly Process between Polystyrene Microspheres and Boron Nitride Nanosheets, *ACS Appl Mater Interfaces*, 2017, **9**, 19934-19944.
10. C. Yu, W. Gong, W. Tian, Q. Zhang, Y. Xu, Z. Lin, M. Hu, X. Fan and Y. Yao, Hot-pressing induced alignment of boron nitride in polyurethane for composite films with thermal conductivity over 50 Wm⁻¹ K⁻¹, *Composites Science and Technology*, 2018, **160**, 199-207.

11. L. Fu, T. Wang, J. Yu, W. Dai, H. Sun, Z. Liu, R. Sun, N. Jiang, A. Yu and C.-T. Lin, An ultrathin high-performance heat spreader fabricated with hydroxylated boron nitride nanosheets, *2D Materials*, 2017, **4**, 025047.
12. Z.-G. Wang, M.-Z. Chen, Y.-H. Liu, H.-J. Duan, L. Xu, L. Zhou, J.-Z. Xu, J. Lei and Z.-M. Li, Nacre-like composite films with high thermal conductivity, flexibility, and solvent stability for thermal management applications, *J. Mater. Chem. C*, 2019, **7**, 9018-9024.
13. H.-R. Zhao, J.-H. Ding, Z.-Z. Shao, B.-Y. Xu, Q.-B. Zhou and H.-B. Yu, High-Quality Boron Nitride Nanosheets and Their Bioinspired Thermally Conductive Papers, *ACS Appl. Mater. Interfaces*, 2019, **11**, 37247-37255.