Electronic Supplementary Material (ESI) for Materials Advances.

High thermal conductive thermoplastic polyurethane/boron nitride/liquid metal composites: role of liquid bridge at the filler/filler interface

Zemin Ji,[a] Wenyi Liu,[a] Chenguang Ouyang[a] and Yanbao Li *[a]

[a] College of Material Science and Engineering, Nanjing Tech University, Nanjing 211816, PR China.
Email: ybli@njtech.edu.cn, lyanbao@163.com

Experimental

TPU/BN composites The thermoplastic polyurethane (TPU) solution (25 vol%) was prepared by dissolving TPU particles in N, N-dimethylformamide (DMF). Then boron nirides platelets (BN, 40 vol%) were added to the TPU solution. After stirring for 1 h, the TPU/BN suspension was poured into a mould (6 cm×6 cm× 4 mm). TPU/BN composites were obtained after dried in a vacuum oven at 60 °C for 30 h.

TPU/BN/LM composites prepared by hand grinding BN platelets and liquid metal (LM, a volume ratio of 3:1) were mixed by hand grinding for 30 min. BN/LM hybrid fillers were obtained. The TPU solution (25 vol%) was prepared by dissolving TPU particles in DMF. Then BN/LM hybrid fillers were added to the TPU solution. After stirring for 1 h, the TPU/BN/LM suspension was poured into a mould (6 cm×6 cm× 4 mm). TPU/BN/LM composites prepared by hand grinding were obtained after dried in a vacuum oven at 60 °C for 30 h.

TPU/BN/LM composites prepared by sonication mixing LM were sonicationed in DMF for 30 minutes. Then TPU particles and BN platelets were added to the LM/DMF suspension. After stirring for 1 h, the TPU/BN/LM suspension was poured into a mould (6 cm×6 cm× 4 mm). TPU/BN/LM composites prepared by sonication were obtained after dried in a vacuum oven at 60 °C for 30 h.

Contact angle tests Contact angle between LM droplets and BN film were tested through a JC2000CS contact-angle meter (Shanghai Zhongchen digital technic apparatus co., Ltd) equipped with a charge coupled device (CCD) camera. The BN film was prepared by cold-pressing BN plateltes under 10MPa. The contact angle was measured through shape analysis of images recorded by the CCD camera. LM droplets were dropped onto the BN film and rubbed on the BN film, respectively.

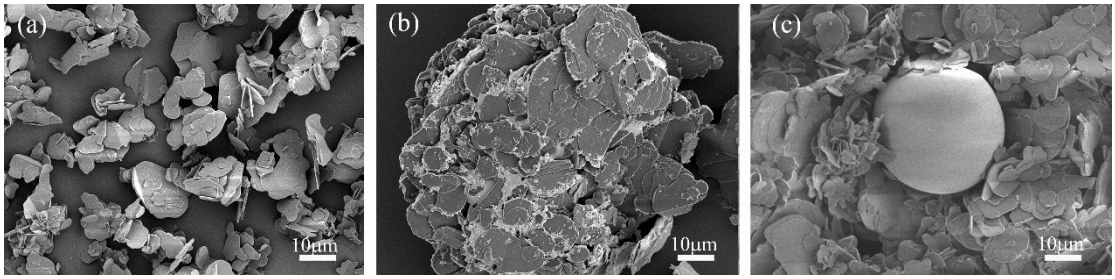

Fig. S1 SEM images of (a) BN platelets, and BN/LM hybrid fillers with a BN/LM ratio of 30/10 prepared by (b) hand grinding and (c) sonication mixing, respectively.

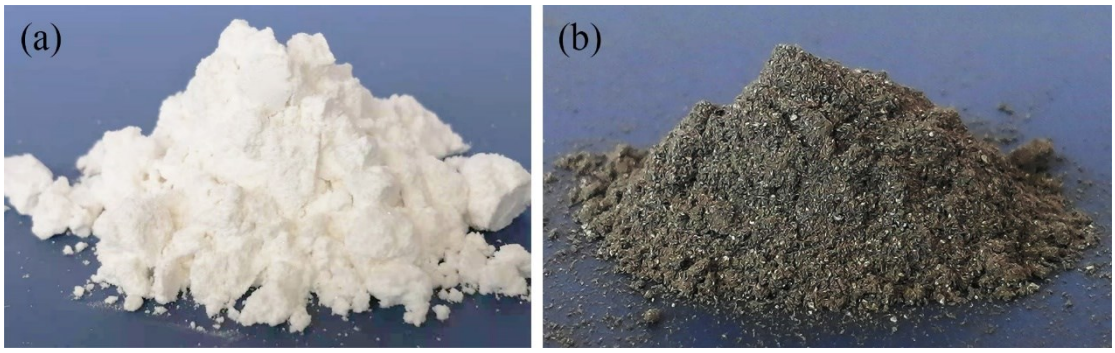

Fig. S2 Digital images of (a) BN powders and (b) BN/LM hybrid filler with the BN/LM ratio of

3:1.

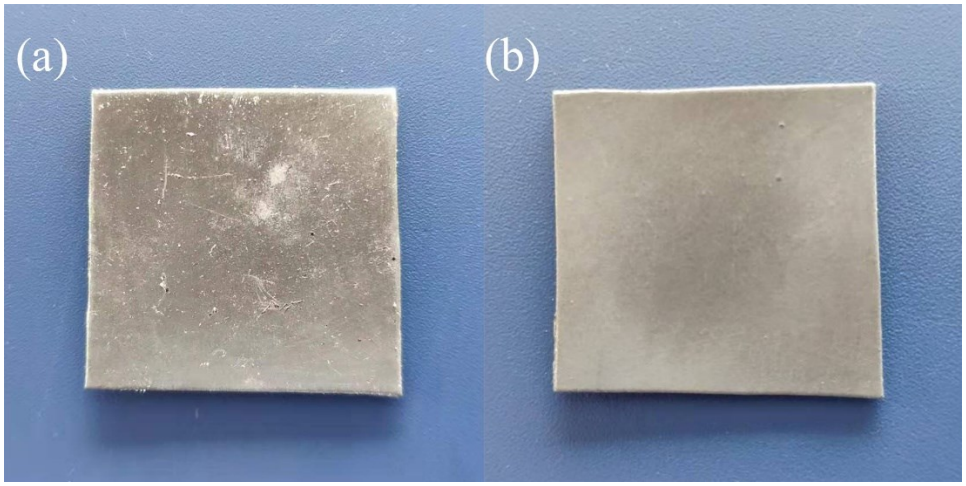

Fig. S3 Digital images of (a) TPU/LM composites and (b) TPU/BN/LM composites.

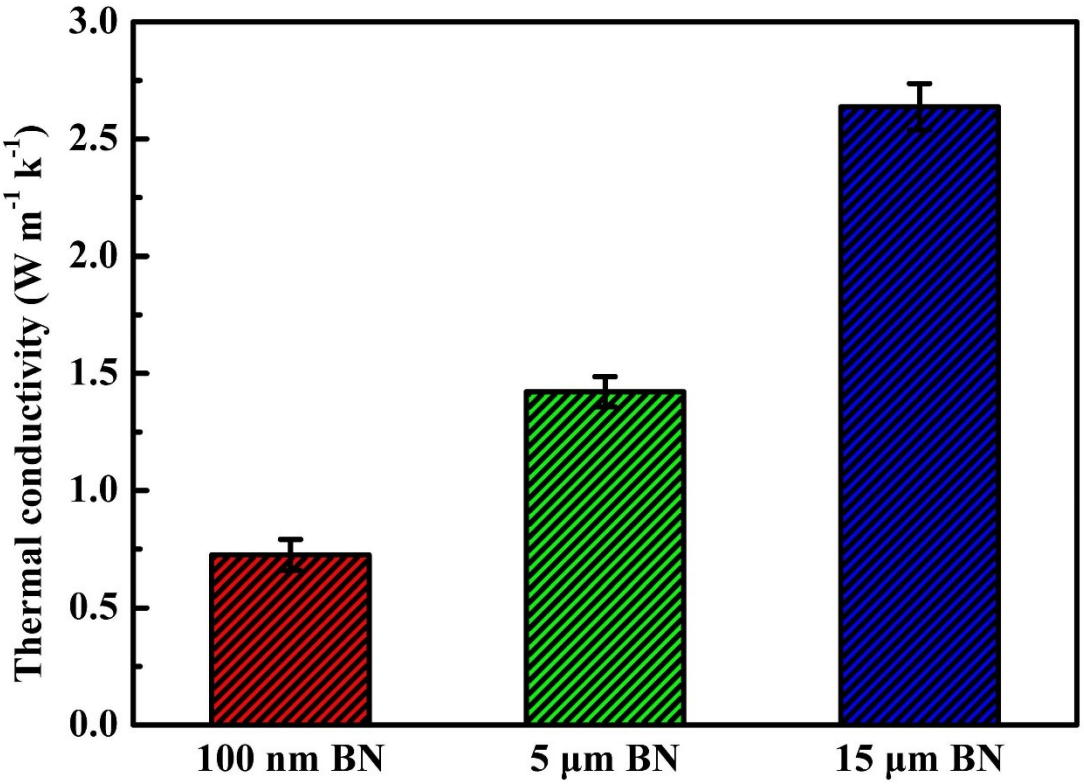

Fig.S4 Thermal conductivity (TC) of TPU/BN/LM composites containing different BN sizes with 40 vol% BN/LM at a BN/LM ratio of 30/10.

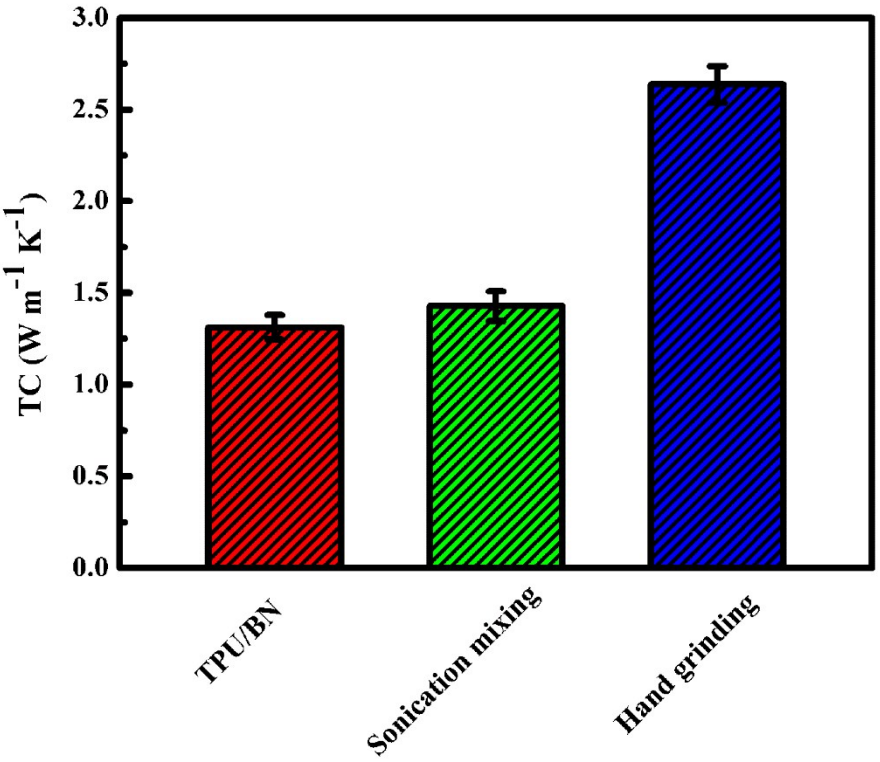

Fig. S5 Thermal conductivity of TPU/BN, TPU/BN/LM composites containing 40 vol% heat filler with a BN/LM ratio of 30/10 prepared by sonication mixing, TPU/BN/LM composites prepared by hand grinding.

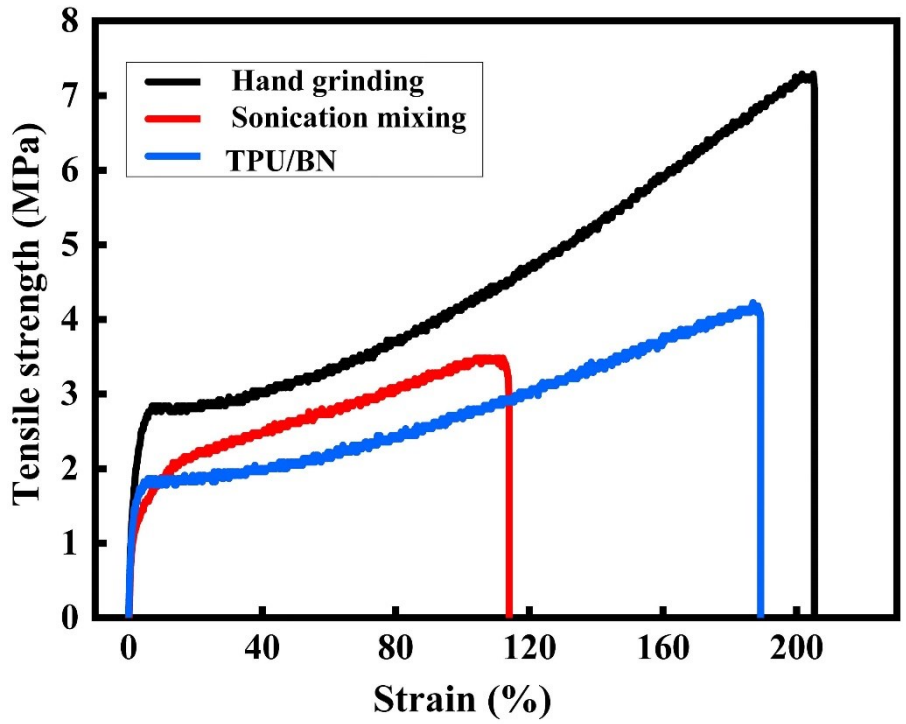

Fig. S6 Stress-strain curves of the TPU/BN composite containing 40 vol% BN and TPU/BN/LM composites containing 40 vol% hybrid filler with a BN/LM ratio of 30/10 prepared by different methods.

Fig. S7 Contact angle of LM and BN films. (a) LM was dropped on the BN film directly. (b) LM was rubbed on the BN film.

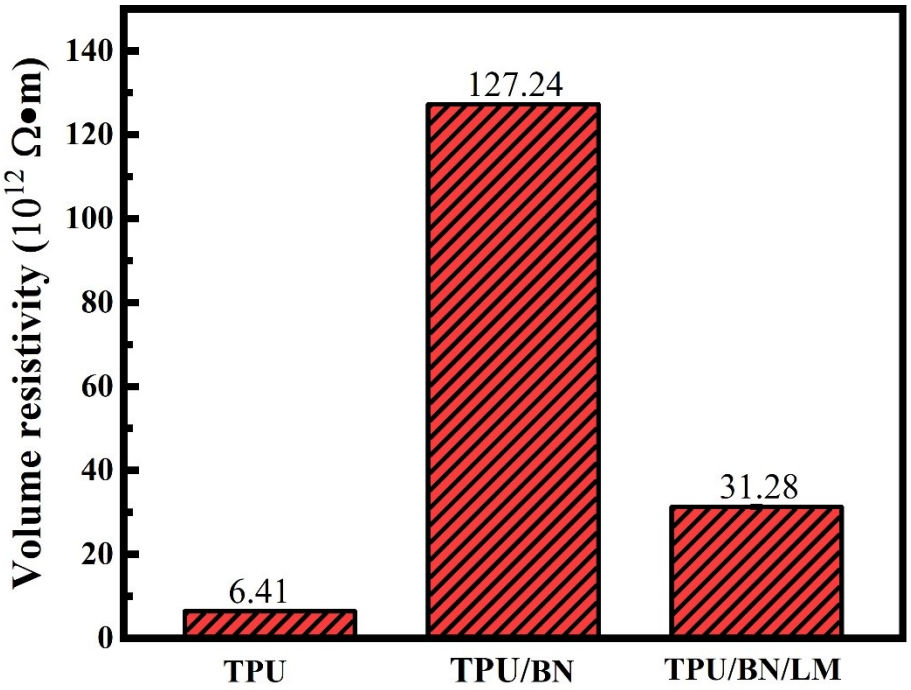

Fig. S8 Volume resistivity of TPU, TPU/BN containing 40 vol% BN and TPU/BN/LM containing 40 vol BN/LM with a BN/LM ratio of 30:10

Table S1 TC of the reported BN based polymer composites

Matrix	Filler	Filler content	TC ($Wm^{-1}K^{-1}$)	Method	Morphology of the filler	Reference
PA6	BN	40 vol%	$5.70^{//}$	Injection	Orientation	S1
PA6	BN/AlN	50 vol%	1.04	Hot-pressing	-	S2
Epoxy	BN	40 vol%	1.98	Impregnation	3D Network	S3
PC	BN	40 wt%	1.35	Hot-pressing	Random	S4
PPS	BN	40 vol%	4.15	Hot-pressing	3D segregation	S5
PP	BN	40 vol%	2.1	Hot-pressing	-	S6
PPS	BN	60 wt%%	2.64	Hot-pressing	-	S7
SiR	BN	40 vol%	1.48	-	Random	S8
PU	BN	40 wt%	$10^{//}$	Hot-pressing	3D Network	S9
PU	BN	40 wt%	$5.2^{//}$	Hot-pressing	High orientation	S10
PU	BN	50 wt%	3.06	Hot-pressing	Orientation	S11
TPU	BN	40 wt%	$2.56^{//}$, $0.91^{\perp}$	3D print	Orientation	S12
PU	BN	45 wt%	2.06	Casting	-	S13
PU	BN	35 wt%	1.78	-	-	S14
PU	BN	30 wt%	0.72	Hot-pressing	Orientation	S15
TPU	BN/LM	40 vol%	2.64	Casting	Random	Our work

Note: PA6: polyamide 6; PC: polycarbonate; PPS: Polyphenylene sulfide; PP: polypropylene; PU: polyurethane; TPU: thermoplastic polyurethane; BN: boron nitride; AlN: aluminium nitride; LM: liquid metal; SiR: Silicone Rubber; $^{//}$ and $^{\perp}$ dnote the in-plane and out-of-plane thermal conductivity of the composites, respectively.

S1. L. Wang, L. C. Zhang, A. Fischer, Y. H. Zhong, D. Drummer and W. Wu, *J. Polym. Eng.*, 2018, **38**, 767.

S2. G. Yildiz and M. Akkoyun, *J. Appl, Polym. Sci.*, 2021, **138**.

S3. X. Y. Leng, C. Xiao, L. Chen, Z. Su, K. Zheng, X. Zhang and X. Y. Tian, *High Perform. Polym.*, 2019, **31**, 350.

S4. S. T. Zhou, Y. Shi, Y. Bai, M. Liang and H. W. Zou, *Polym. Compos.*, 2020, **41**, 5418-5427.

S5. Y. Jiang, Y. J. Liu, P. Min and G. X. Sui, *Compos. Sci. Technol.*, 2017, **144**, 63.

S6. S. Takahashi, Y. Imai, A. Kan, Y. Hotta and H. Ogawa, *J. Alloys Compd.*, 2014, **615**, 141.

S7. J. W. Gu, Y. Q. Guo, X. T. Yang, C. B. Liang, W. C. Geng, L. Tang, N. Li and Q. Y. Zhang, *Compos. Part A Appl. Sci. Manuf.*, 2017, **95**, 267.

S8. W. Y. Zhou, S. H. Qi, H. Z. Zhao and N. L. Liu, *Polym. Compos.*, 2007, 28, 23.

S9. H. J. Hong, S. M. Kwan, D. S. Lee, S. M. Kim, Y. H. Kim, J. S. Lim, J. Y. Hwang and H. S. Jeong, *Compos. Sci. Technol.*, 2017, **152**, 94.

S10. C. P. Yu, W. B. Gong, W. Tian, Q. C. Zhang, Y. C. Xu, Z. Y. Lin, M. Hu, X. D. Fan and Y. G. Yao, *Compos. Sci. Technol.*, 2018, **160**, 199.

S11. T. Fei, Y. B. Li, B. C. Liu and C. B. Xia, *High Perform. Polym.*, 2020, **32**, 324.

S12. J. Liu, W. Li, Y. Guo, H. Zhang and Z. Zhang, *Compos. Part A Appl. Sci. Manuf.*, 2019, **120**, 140.

S13. A. Bashir, M. Maqbool, R. C. Lv, A. Usman, H. C. Guo, W. Aftab, H. Y. Niu, M. J. Liu and S. L. Bai, *Compos. Part B Eng.*, 2021, **218**, 108871.
S14. G. Christensen, D. Lou, H. P. Hong and G. P. Peterson, *Thermochim. Acta*, 2021, **700**, 178927.
S15. S. Ryu, H. Oh and J. Kim, *Mater. Chem. Phys.*, 2019, **223**, 607.