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Supporting Information

A thermal interface material based on foam-templated three-dimensional hierarchical porous boron nitride

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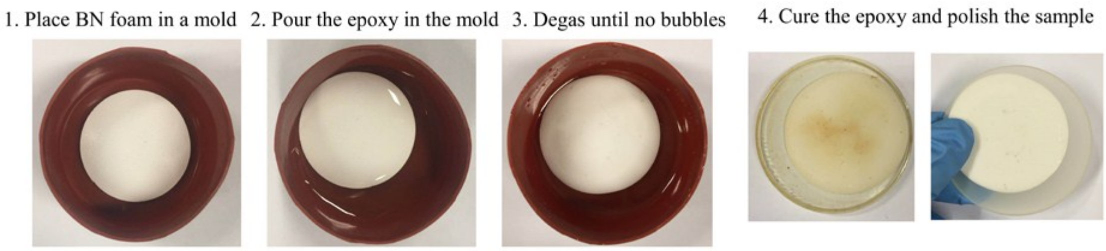

Figure S1 Preparation process of the BN/epoxy composites

The porous BN foam was placed in a mold. Then, the epoxy was poured into the mold, leaving BN foam floating on the liquid epoxy. We then used vacuum degassing to facilitate the infiltration of epoxy into the porous BN foam. (The time of degassing was optimized to ensure the epoxy can well fill the top part pores of the foam.) Then the mold was heated to 160 ^{o}C to cure the epoxy. After curing, we can obtain the BN/epoxy composites and the bottom part has more epoxy than the top part. Finally, we use an abrasive paper to polish the sample to remove the redundant epoxy at the bottom part until a uniform BN-epoxy composite is obtained.

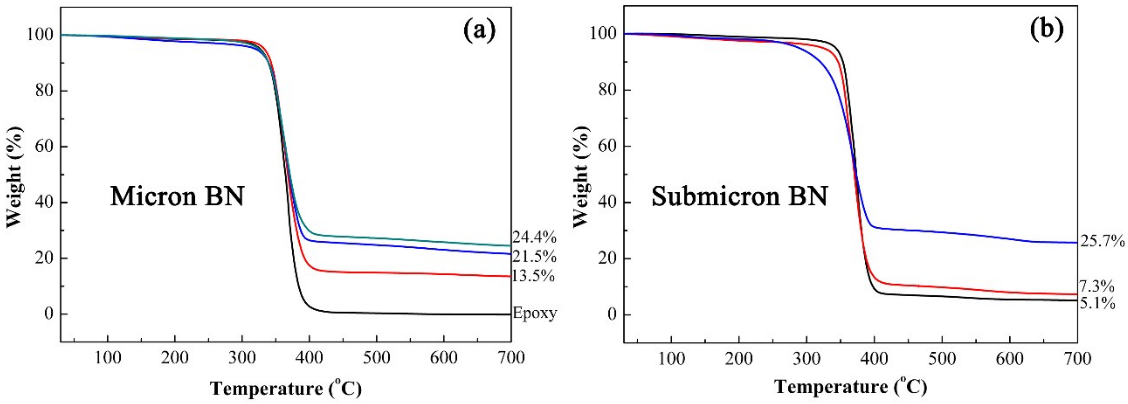

Figure S2. TGA curves of (a) micron and (b) submicron BN/epoxy composites

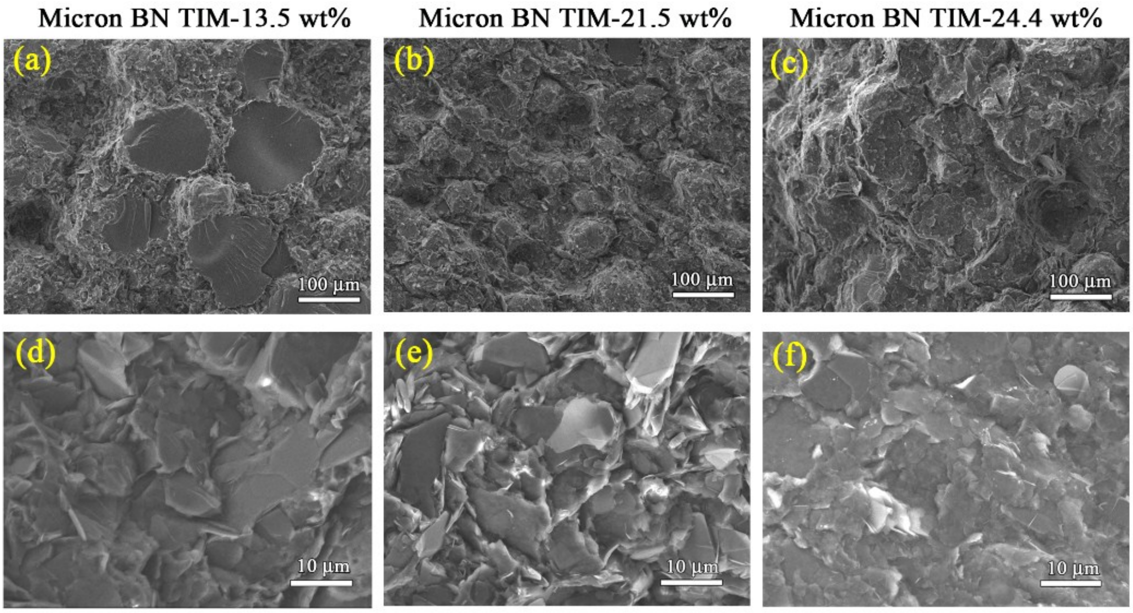

Figure S3. Microstructures of fracture surface of micron BN/epoxy composites with (a), (d) 13.5 wt%, (b), (e) 21.5 wt% and (c), (f) 24.4 wt% fillers, respectively

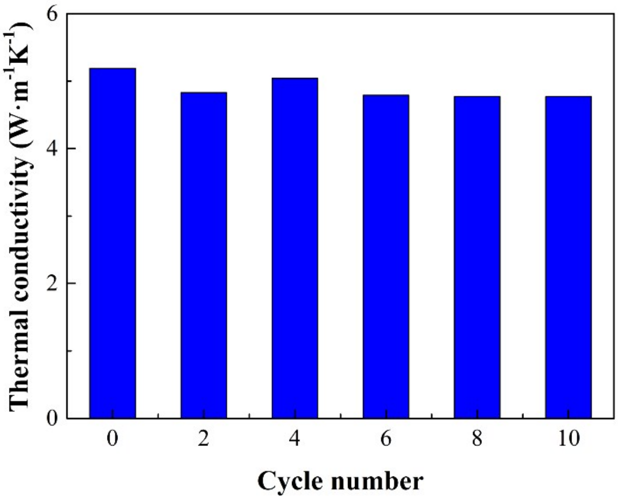

Figure S4. In-plane thermal conductivity of the 24.4 wt% micron BN/epoxy composite upon multiple heating and cooling cycles

We performed multiple heating and cooling cycles alternating between 25 and 100 °C to test the thermal duration and performance stability of the composite. In every circle, the composite was held at 100 °C for 5 minutes. The in-plane thermal conductivity of the 24.4 wt% micron BN/epoxy composite after every two cycles were recorded. It can be clearly seen that the composite exhibits a good stability, showing a thermal conductivity variation of less than 8% within 10 cycles.

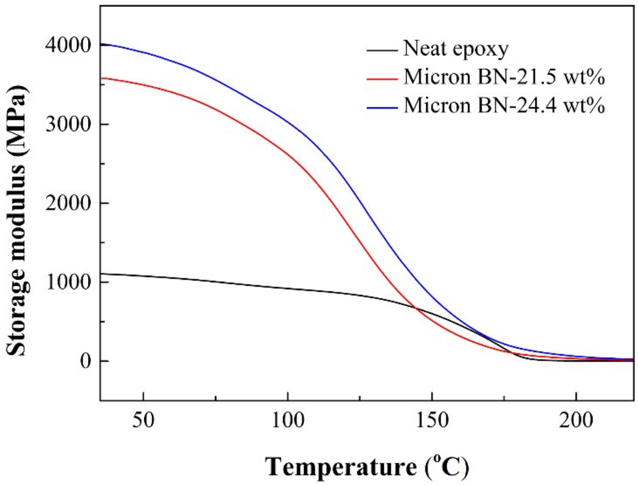

Figure S5. Storage modulus of BN/epoxy composites as a function of temperature

Typically, the deformability of the BN-epoxy composites decreases with the increased loading of BN fillers. High concentrations of BN make the composites rigid and brittle; accordingly, we measured the storage modulus of the 21.5 wt% and 24.4 wt% micron BN/epoxy composites to evaluate their stiffness. Their storage moduli are higher than that of the neat epoxy due to the high Young's modulus of BN. Table S1 compares the storage modulus and thermal conductivity of the 24.4 wt% micron BN/epoxy with other BN/epoxy composites. It can been seen that our material design can achieve high thermal conductivity at a reasonably low storage modulus.

Table S1. Comparison of thermal conductivity and storage modulus of our material with other BN/epoxy composites

Material	BN concentration (wt %)	Thermal conductivity ($Wm^{-1}K^{-1}$)	Storage modulus (GPa)	Reference
BN/epoxy	23.7	5.05	4.61	[S1]
BN/Epoxy	26.5	1.20	3.75	[S2]
BN/Epoxy	40.0	0.98	5.60	[S3]
BN/epoxy	**24.4**	**5.19**	**4.02**	**This work**

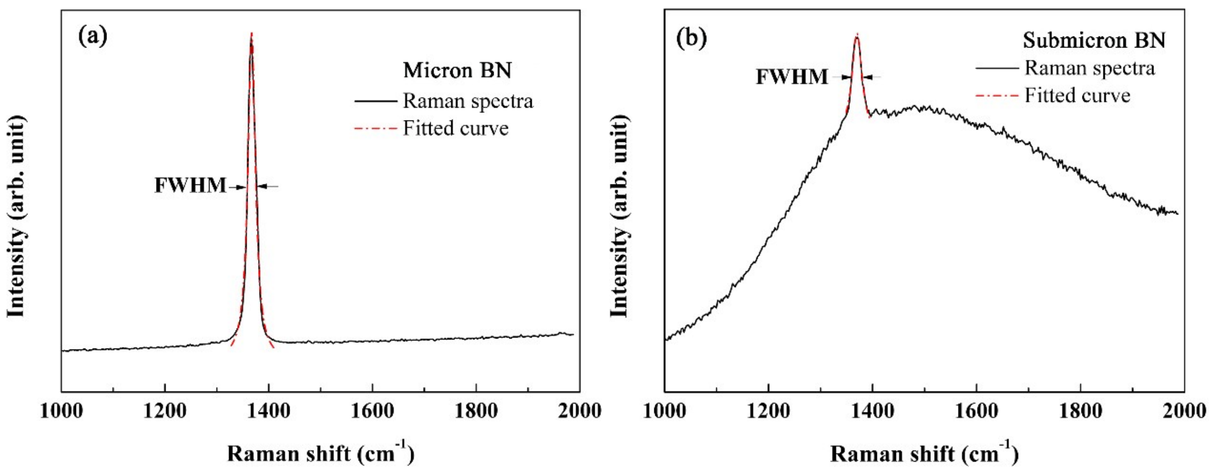

Figure S6. Raman spectra of (a) micron and (b) submicron BN

Reference

[S1] Y. Yao *et al*. Construction of 3D skeleton for polymer composites achieving a high thermal conductivity. *Small*, 2018, 14, 1704044.

[S2] Y. Jiang *et al*. Enhanced thermal conductivity and ideal dielectric properties of epoxy composites containing polymer modified hexagonal boron nitride. *Composites Part A: Applied Science and Manufacturing*, 2018, 107, 657.

[S3] I. Isarn *et al*. Thermoconductive thermosetting composites based on boron nitride fillers and thiol-epoxy matrices. *Plomers*, 2018, 10, 277.