Supporting Information

Improved thermal conductivity and excellent electrical insulation property of polysiloxane nanocomposites incorporated functional boron nitride sheets via in-situ polymerization

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Fig. S1. AFM image of the BNNS.

Fig. S2. Raman spectrum of the BNNS.

Table S1. Element content of BNNS and f-BNNS.

Fig. S3. TGA curves of BN and f-BNNS.

Fig. S4. The through-plane κ of f-BNNS/SCLCP nanocomposites in this work.

Fig. S5. The simulated thermal conductivity curves of f-BNNS/SCLCP nanocomposites system based on effective medium theory model and the experimental values.

Table S2. The detailed thermal conductivity data of the samples.

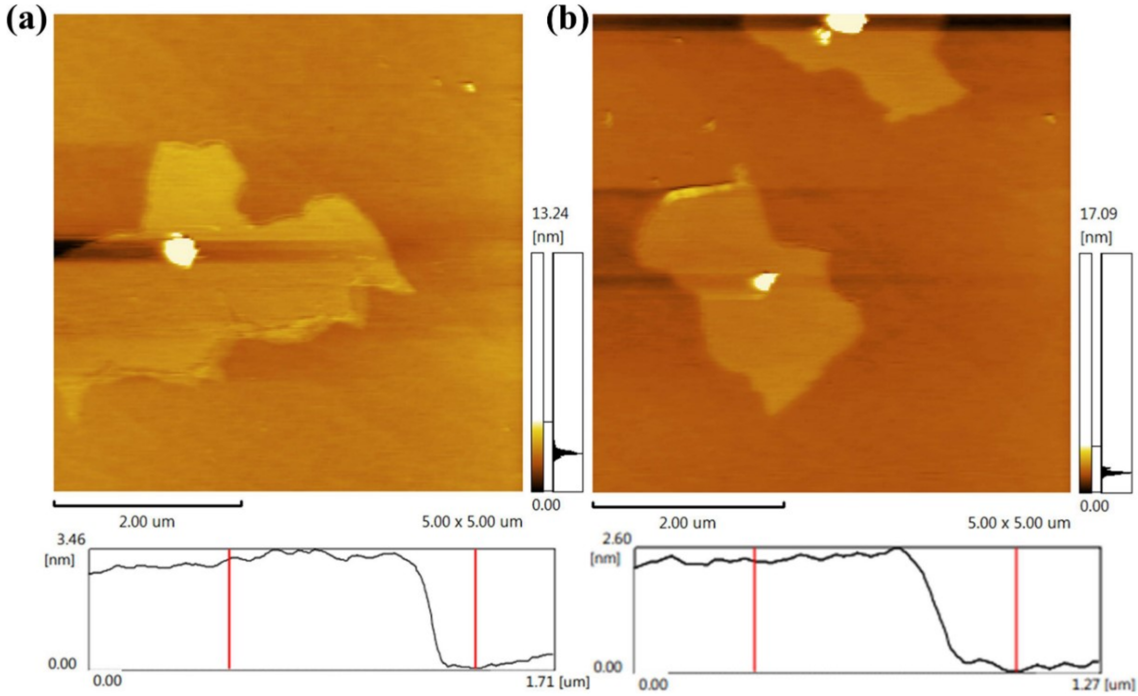

Fig. S1. AFM image of the BNNS.

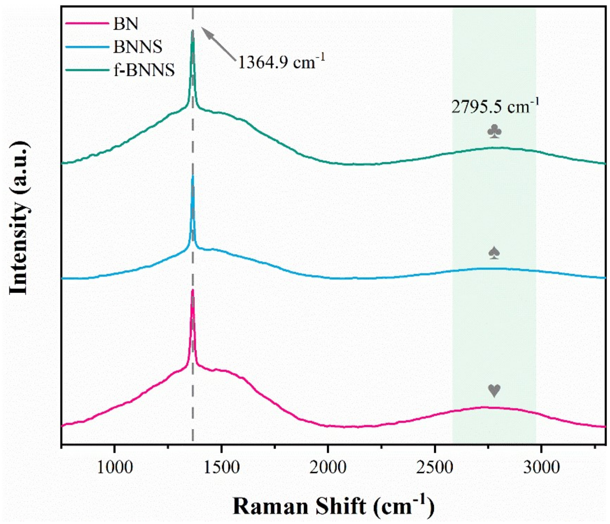

Fig. S2. Raman spectrum of the BNNS.

Table S1. Element content of BNNS and f-BNNS.

Samples	**Elemental content (Atomic%)**				
	B	C	N	O	Si
BNNS	48.57	8.47	39.86	2.64	/
f-BNNS	35.57	29.78	26.99	6.90	0.77

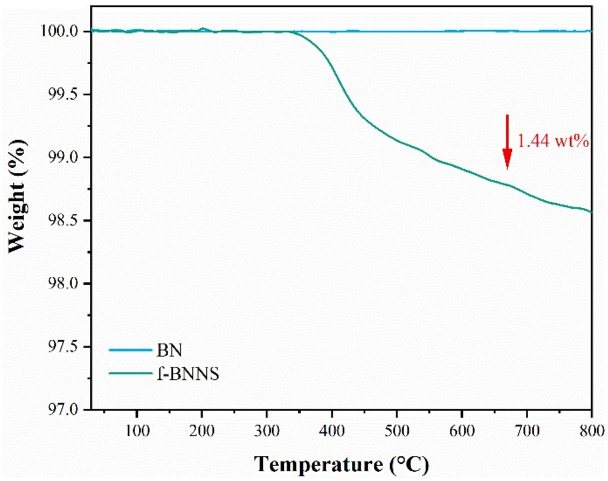

Fig. S3. TGA curves of BN and f-BNNS.

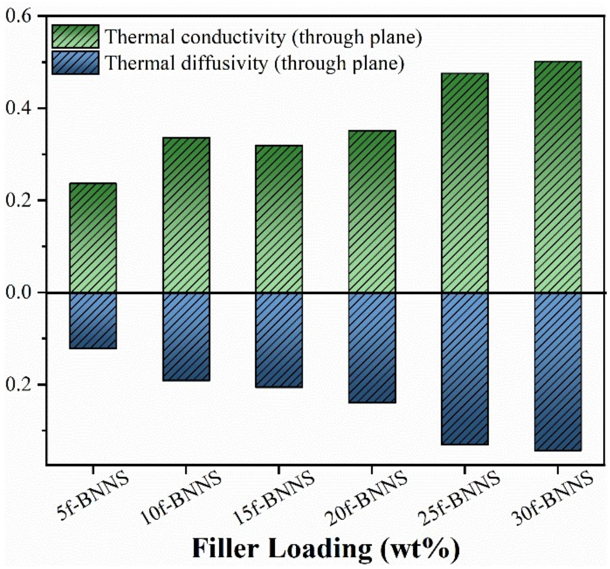

Fig. S4. The through-plane κ of f-BNNS/SCLCP nanocomposites in this work.

Table S2. The detailed thermal conductivity data of the samples.

Samples	ρ ($g\cdot cm^{-3}$)	C_p ($Jg^{-1}K^{-1}$)	α (mm^2S^{-1})	$\kappa_\perp$ ($Wm^{-1}K^{-1}$)
5f-BNNS/SCLCP	1.036	1.87	0.122	0.236
10f-BNNS/SCLCP	1.089	1.61	0.191	0.335
15f-BNNS/SCLCP	1.114	1.39	0.206	0.319
20f-BNNS/SCLCP	1.124	1.3	0.24	0.351
25f-BNNS/SCLCP	1.134	1.27	0.33	0.475
30f-BNNS/SCLCP	1.141	1.28	0.343	0.501

- **Section 1. Theoretical calculation of thermal resistance between the f-BNNS/SCLCP materials by Effective medium theory (EMT)**

To further explore the association between the thermal conductive phenomenon and the structure of the nanocomposites, the effective medium theory (EMT) model was employed to fit the experimental data and calculate the interface thermal resistance of filler-SCLCPs. According to the EMT model, [1-4] the thermal conductivity of the materials can be expressed as follows:

$$\kappa = \kappa_m \frac{3 + f(\beta_\perp + \beta_\parallel)}{3 - f\beta_\perp} \#E1$$

where

$$\beta_\perp = \frac{2\left[d(\kappa_f - \kappa_m) - 2R_{BD}\kappa_f\kappa_m\right]}{d(\kappa_f + \kappa_m) + 2R_{BD}\kappa_f\kappa_m} \#E2$$

$$\beta_\parallel = \frac{L(\kappa_f - \kappa_m) - 2R_{BD}\kappa_f\kappa_m}{L\kappa_m + 2R_{BD}\kappa_f\kappa_m} \#E3$$

and κ, κ_m and κ_f are the thermal conductivity of the composites, SCLCPs and filler, respectively, f is the volume fraction of the filler, d and L are the thickness and lateral size of the filler, and R_{BD} is the interface thermal resistance between filler and SCLCP. f can be calculated from the weight fration:

$$f = \frac{w\rho_c}{\rho_f} \#E4$$

where w is the weight fraction of f-BNNS nanoplatelets in the f-BNNS/SCLCP nanocomposite films, ρ_c is the density of the nanocomposites; ρ_f is the density of f-BNNS nanoplatelets. In this case, the d and L of f-BNNS are approximately 3 nm and 400 nm measured by AFM and TEM. The thermal conductivity of BNNS is ~600 W m^{-1} K^{-1}. Our measurements of pure SCLCP give κ_m=0.269 W m^{-1} K^{-1}. ρ_g is wildly accepted as 2.29 g cm^{-3}. By fitting the in-plane thermal conductivity of the composites (Fig. S5), the parameters $\beta_\perp$ and $\beta_{//}$ can be obtained to calculate the interface thermal resistance. In this case, the R_{BD} for f-BNNS/SCLCP interface and BNNS/SCLCP interface are 4.09×10^{-9} m^2 K W^{-1} and 5.0×10^{-9} m^2 K W^{-1}, which is consistent with the previously reported results. [5, 6]

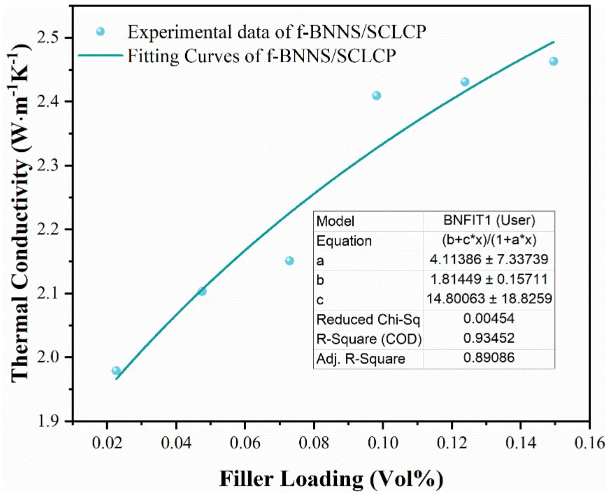

Fig. S5. The simulated thermal conductivity curves of f-BNNS/SCLCP nanocomposites based on EMT model and the experimental values.

Reference

1. C. W. Nan, X. P. Li and R. Birringer, *J. Am. Ceram. Soc.*, 2000, **83**, 848-854.

2. C. W. Nan, G. Liu, Y. H. Lin and M. Li, *Applied Physics Letters*, 2004, **85**, 3549-3551.

3. M. B. Bryning, D. E. Milkie, M. F. Islam, J. M. Kikkawa and A. G. Yodh, *Applied Physics Letters*, 2005, **87**, 3.

4. Y. Liu, M. Lu, Z. Hu, L. Liang, J. Shi, X. Huang, M. Lu and K. Wu, *Chem. Eng. J.*, 2020, **382**.

5. J. T. Hu, Y. Huang, Y. M. Yao, G. R. Pan, J. J. Sun, X. L. Zeng, R. Sun, J. B. Xu, B. Song and C. P. Wong, *ACS Appl. Mater. Interfaces*, 2017, **9**, 13544-13553.

6. Z. Liu, J. H. Li and X. H. Liu, *ACS Appl. Mater. Interfaces*, 2020, **12**, 6503-6515.