Supporting Information

Nacre-like composite films with high thermal conductivity, flexibility, and solvent stability for thermal management applications

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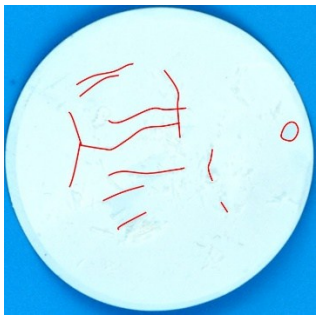

Fig. S1 Digital photograph of BNNS/PVA film with the 75 wt% BNNS after slight bending. The red lines represent the cracks on the surface.

Fig. S2 The special accessory used to measure the in-plane diffusivity.

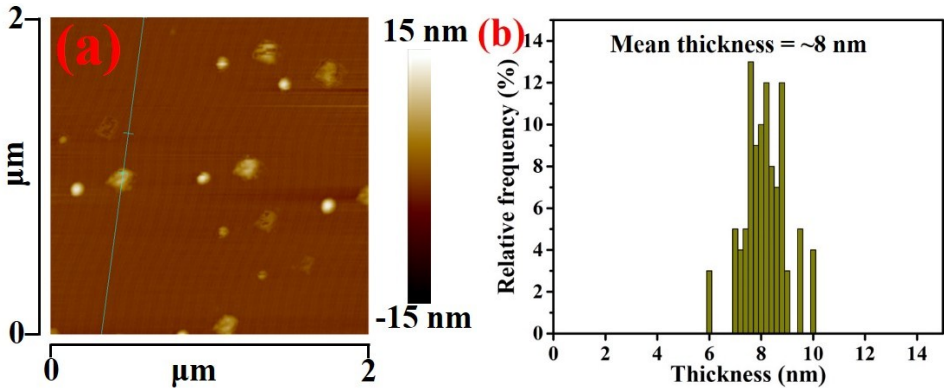

Fig. S3 (a) More AFM images of BNNSs and (b) the distribution of thickness of BNNSs obtained by the statistical calculation.

AFM was used to obtain the average thickness of BNNSs by measuring 30 BNNSs and the corresponding distribution is shown in Fig. S3a-b. We find that most of BNNSs have the thickness in the range of 7 ~9 nm, showing the average thickness of ~8 nm.

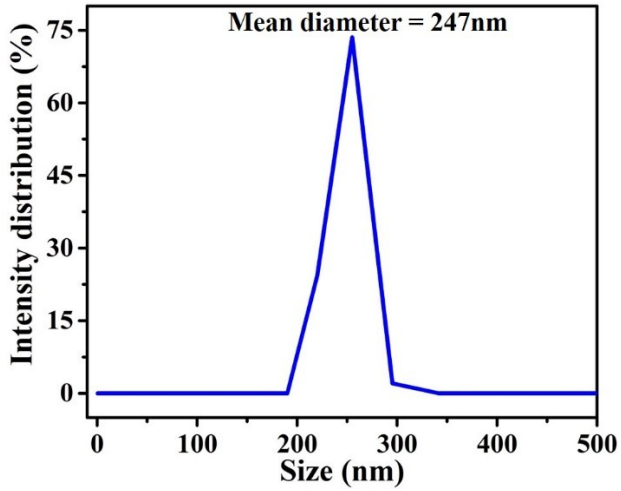

Fig. S4 The size distribution of BNNS measured by dynamic light scattering.

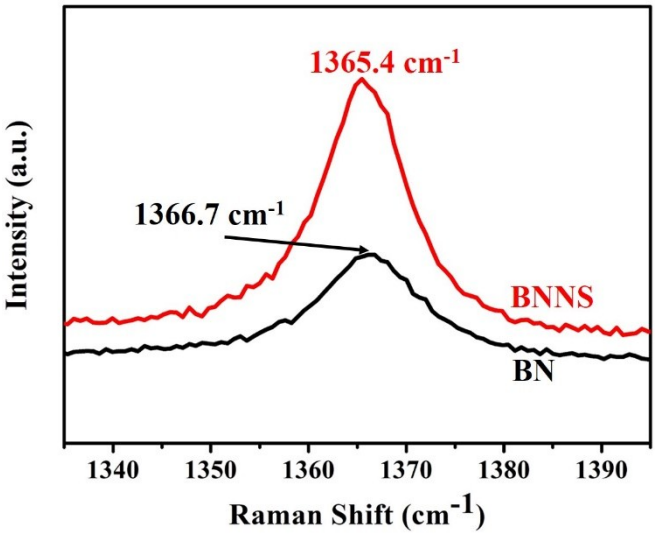

Fig. S5 Raman spectra of (a) BN platelets and (b) BNNS.

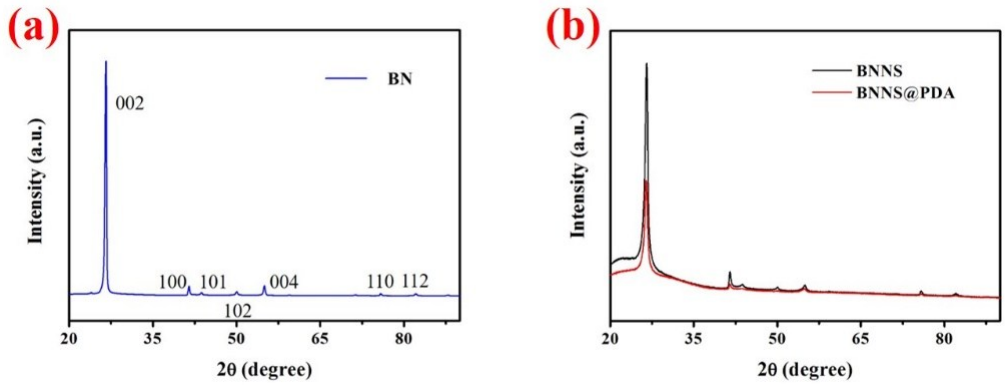

Fig. S6 XRD patterns of (a) BN, BNNS and (b) BNNS@PDA.

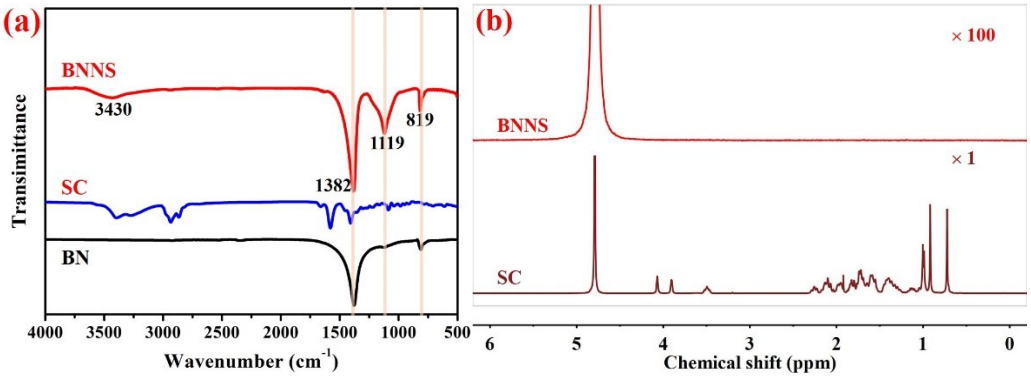

Fig. S7 (a) FTIR spectra of BNNSs, SC and BN platelets; (b) ^{1}H-NMR spectra of SC and BNNSs.

The structure groups of BN platelets, SC, and BNNSs were analyzed using an FTIR spectrometer (Model 6700, Thermo Scientific, USA). It could be observed that there are no featured peaks of SC for the BNNSs (Fig. S7a). The 1H NMR spectra of sodium cholate (SC) and BNNSs dispersed in D_2O were recorded using an NMR spectrometer (Bruker Avance III 400, USA), and chemical shifts were calibrated using the solvent residual signal at 4.79 ppm (D_2O, Macklin Inc, China). As plotted in Fig. S7b, there is no featured peak of SC in BNNSs even after 100 magnification, suggesting that SC is not adsorbed and all removed from BNNSs. These results demonstrate that all the SC

is removed from the BNNSs. The role of SC was to assist the exfoliation of BN platelets in aqueous solution.

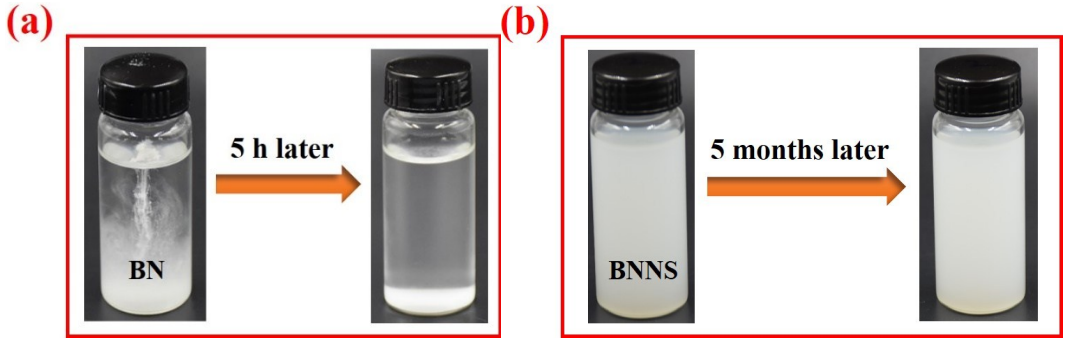

Fig. S8 Optical images of (a) BN and (b) BNNS aqueous dispersion.

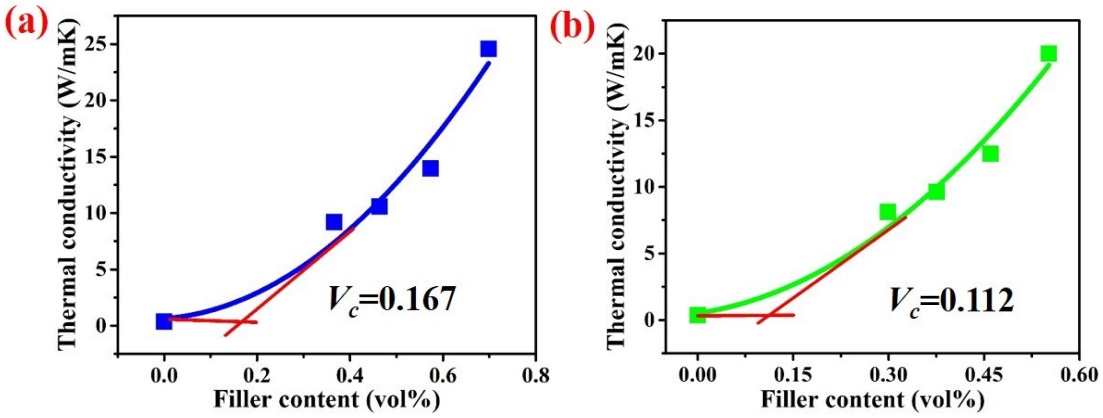

Fig. S9 The tangent process of experimental data for (a) BNNS@PDA/PVA and (b) BNNS/PVA films to obtain V_c.

When the V_f is equal to the V_c, the TCs of BNNS@PDA and BNNS/PVA is equal to that of pure PVA, according to Equation (1) in the manuscript. Here, V_c represents the critical volume fraction of BNNSs or BNNS@PDA in the composites, where the TC of composite films begins to increase quickly. The tangent curves were shown in Fig. S9.

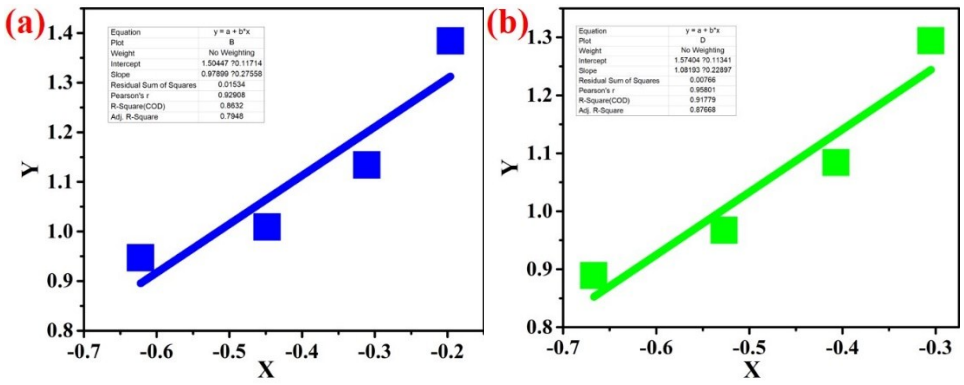

Fig. S10 The fitting process of Foygel model for (a) BNNS@PDA/PVA and (b) BNNS/PVA films.

To fit the Foygel model, we first transform Equation (1) into a linear form: $y=a+bx$, where a equals to $\lg K_0$, b equals to τ, x equals to $\lg[(V_f-V_c)/(1-V_c)]$. y equals to $\lg(K-K_m)$, respectively. The values of a and b can be gained by fitting experimental data. On the basis of the results, the values of K_0 and τ are calculated, as presented in Table S1.

Table S1. Parameters obtained from Foygel's model for BNNS@PDA/PVA and BNNS/PVA films.

Sample	K_0 (W/mK)	τ
BNNS@PDA/PVA	31.95	0.98
BNNS/PVA	37.50	1.08

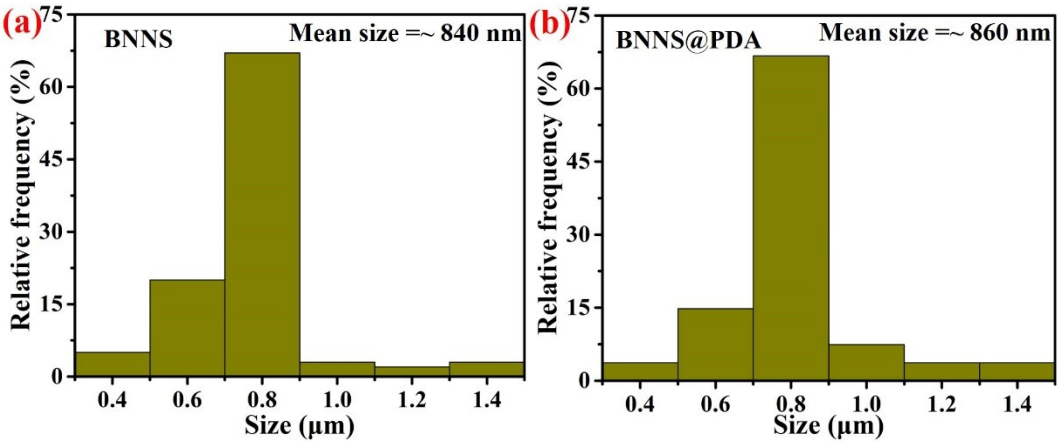

Fig. S11 The distribution of size of BNNSs and BNNS@PDA obtained by the statistical calculation.

The contact area (*S*) was defined as the overlapping area between adjacent nanoplatelets in the equation. Considering the reassembly of BNNSs and BNNSs@PDA during the vacuum-assisted filtration, it is more proper to calculate *S* using the size of 2D nanofillers in the composite films. The assembled size is the actual size to construct orderly lamellar structure and thermal conductive pathways (Fig. S11).

Therefore, we statistically count the assembled size of BNNSs and BNNS@PDA. The mean size of BNNSs and BNNS@PDA is 860 and 840 nm.

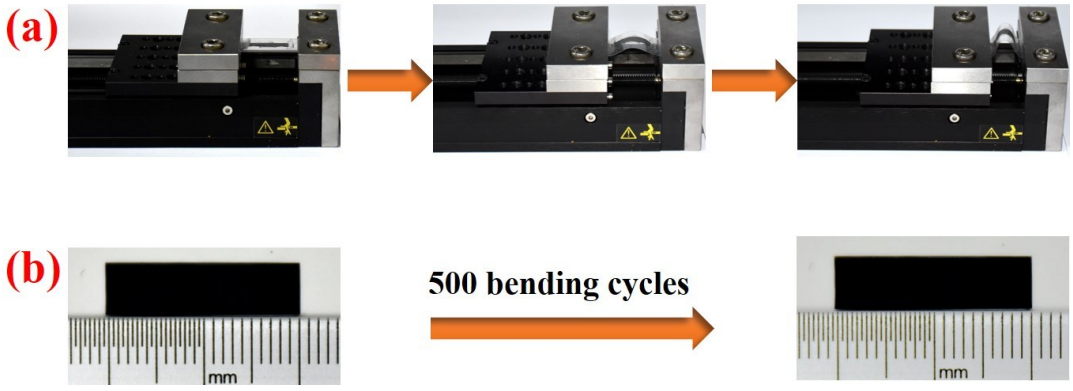

Fig. S12 Optical images of (a) a whole bending process and (b) the 70 wt% BNNS@PDA/PVA film before and after 500 bending cycles.