

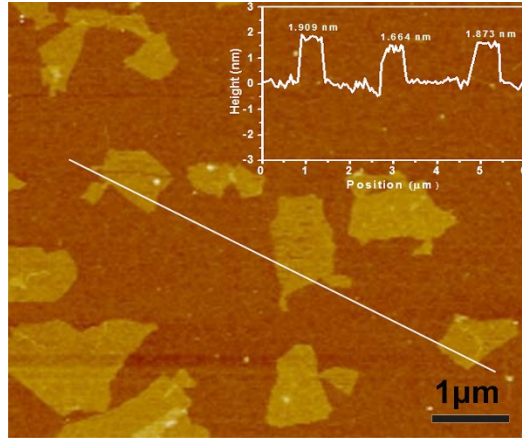
## Supporting Information

### **Surface Modification and Magnetic Alignment of Hexagonal Boron Nitride Nanosheets for High Thermally Conductive Composites**

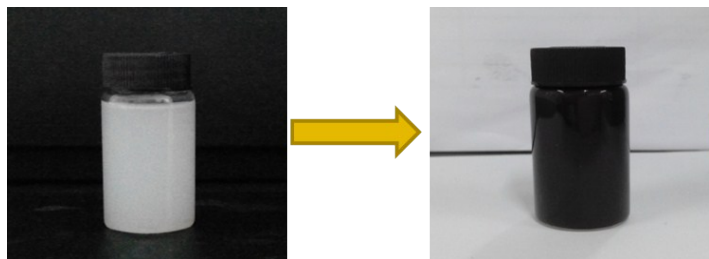
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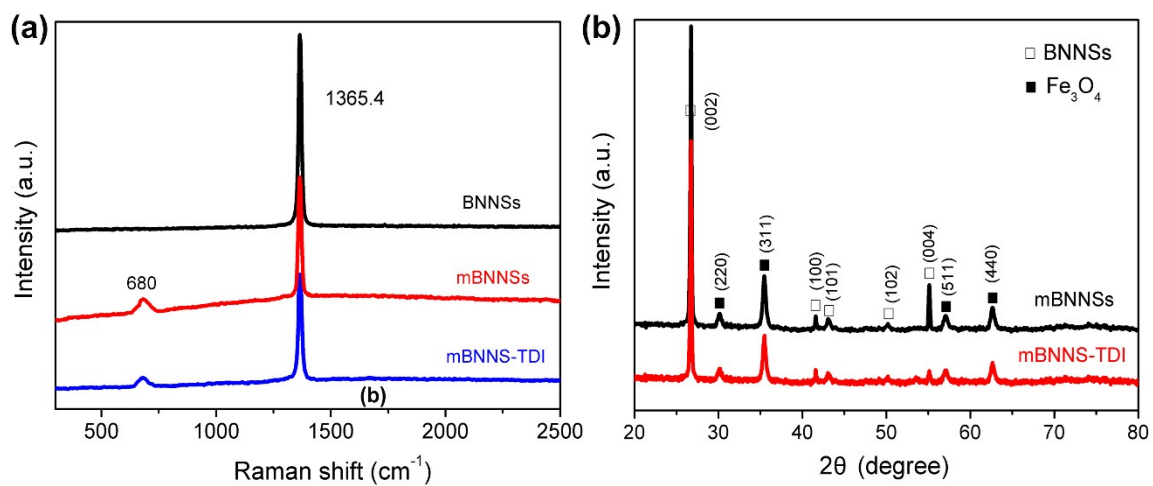
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**Fig. S1.** AFM image and height profiles of BNNSs.



**Fig. S2.** Photographs of BNNSs (left) and mBNNSs (right) dispersed in water.



**Fig. S3** (a) Raman spectra of BNNs, mBNNs and mBNNs-TDI. (b) XRD patterns of mBNNs and mBNNs-TDI, the hollow labels refer to diffraction peaks from BNNs, and the solid labels refer to peaks from  $\text{Fe}_3\text{O}_4$  nanoparticles.

$$T_m = 2\pi\mu_o\chi_\alpha^2[(h+d)(l+d)^2 - hl^2]H_m^2\sin^2\theta/3(\chi_\alpha + 1) \quad (S1)$$

where  $\mu_o$  the magnetic permeability of free space ( $4\pi \times 10^{-7} \text{ Hm}^{-1}$ ),  $\chi_\alpha$  is the magnetic susceptibility of mBNNS-TDI,  $h$  and  $l$  are the half of the thickness and diameter of mBNNS-TDI,  $d$  is the particle size of  $\text{Fe}_3\text{O}_4$ ,  $\theta$  is the angle between mBNNS-TDI long axis and the magnetic vector.

$$T_g = 2\pi hl^2(\rho_f + \rho_m)gl\cos\theta \quad (S2)$$

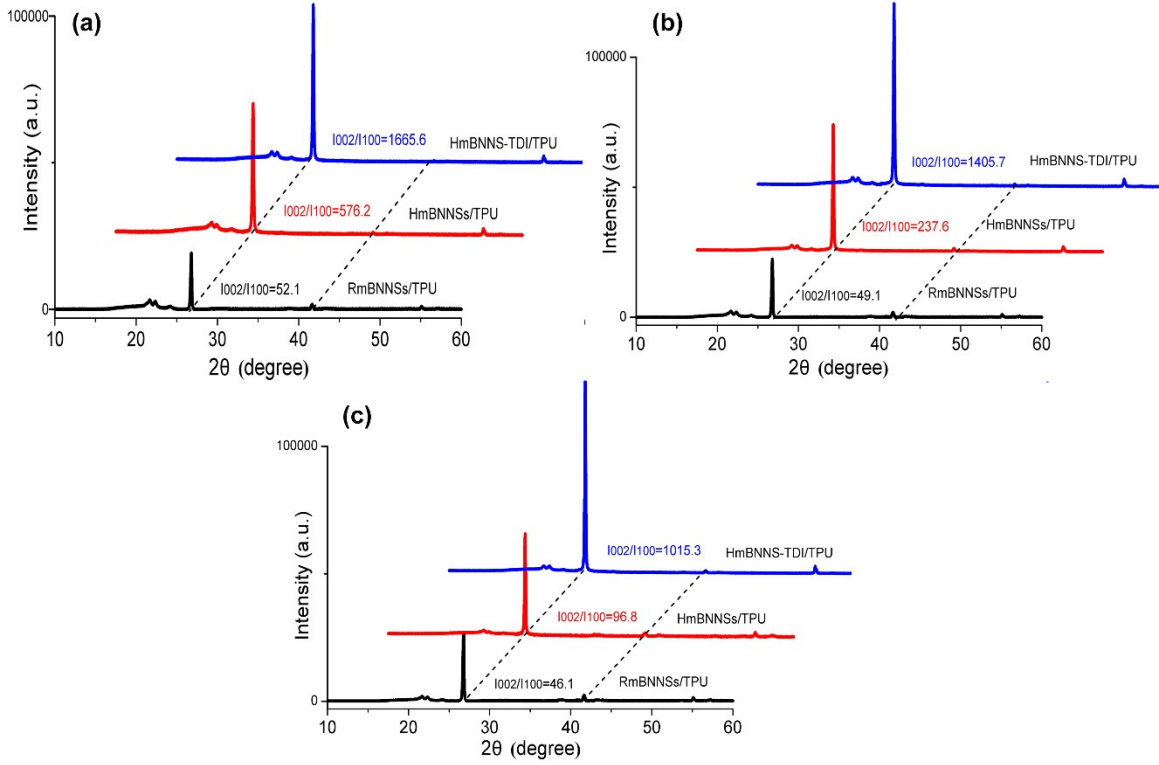
where  $\rho_f$  and  $\rho_m$  are the density of mBNNS-TDI and TPU matrix, respectively,  $g$  is a constant.

$$T_v = 12\pi\eta hl^2(d\phi/dt)(f/f_o)$$

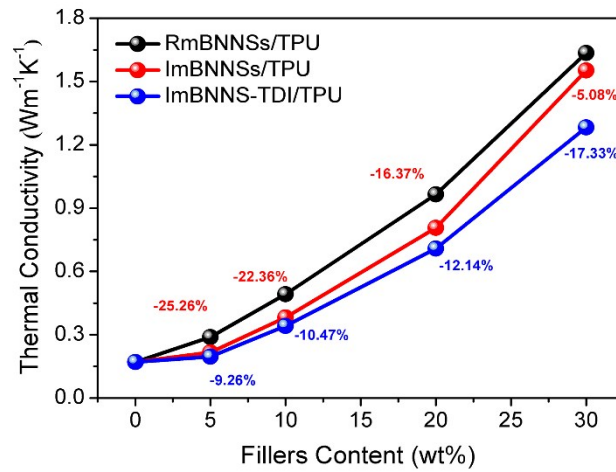
$$\text{where, } f/f_o = 4(1 - \delta^2)/[3(2 - hR\delta^2)],$$

$$R = (2/h)(\delta^2 - 1)^{-1/2}\tan^{-1}[(\delta^2 - 1)^{1/2}] \quad (S3)$$

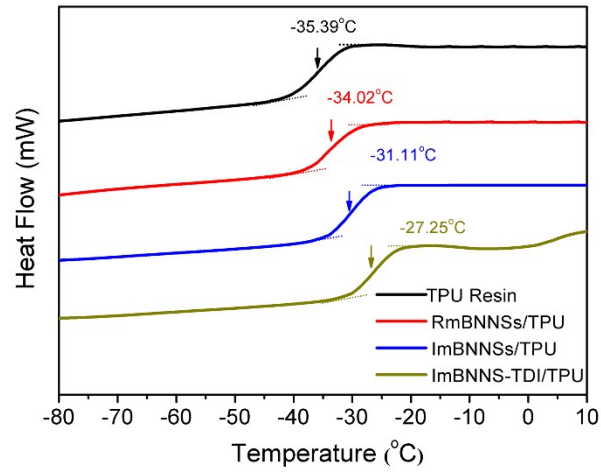
where  $\eta$  is the viscosity of resin,  $d\phi/dt$  is the angular frequency of mBNNS-TDI,  $\delta$  is the aspect ratio of mBNNS-TDI.



**Fig. S4** XRD patterns of different composites with (a) 5 wt% fillers, (b) 20 wt% fillers, (c) 30 wt% fillers.

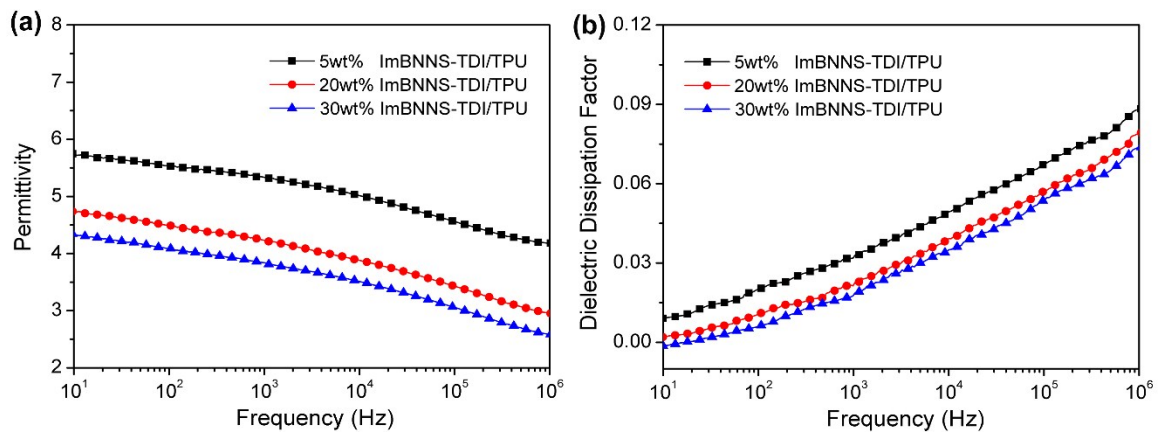


**Fig.S5** Through-plane thermal conductivities of RmBNNs/TPU, ImBNNs/TPU and ImBNNs-TDI/TPU composites. The red numbers show the through-plane thermal conductivity enhancement of ImBNNs/TPU in comparison with the RmBNNs/TPU composites. The blue numbers show the through-plane thermal conductivity enhancement of ImBNNs-TDI /TPU in comparison with the ImBNNs/TPU composites.



**Fig. S6** DSC curves of TPU resin and various composites with 10 wt% loading.





**Fig. S7** Frequency dependence of dielectric constant (a) and dielectric dissipation factor (b) of ImBNNs-TDI/TPU composites with various filler loadings.