## **Supplementary material**

Constructing Binder-Free 3D Thermal Networks with Hexagonal

Boron Nitride of Varying Sizes to Enhance Polydimethylsiloxane

## **Composites: A Comparative Study**

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## **Calculation of BN contents**



Figure S1. TGA curves of pure PDMS, pure BN and BP-3.

It is worth mentioning that the residual mass percentage of PDMS in TGA analysis was 27.72%, and the calculation basis of BN loading in composite was given in the following example. Taking the calculation of h-BN content in BP-3 as an example:

Simultaneous equations: 
$$\begin{cases} M_{BN} + 0.2772 \times M_{PDMS} = 0.7190 \\ M_{BN} + M_{PDMS} = 1 \end{cases}$$

Where  $M_{BN}$  is the mass fraction of h-BN in BP-3,  $M_{PDMS}$  is the mass fraction of PDMS in BP-3.

Get 
$$M_{BN} = 61.12 \text{ wt\%}$$
  
Based on  $\rho_{BN} = 2.29 g \cdot cm^{-3}$  and  $\rho_{PDMS} = 1.00 g \cdot cm^{-3}$ 

And then, the volume fraction of BN in BP-3 was computed  $as V_{BN} = 40.70 \ vol\%$ 

Table S1. Detailed data of thermal conductivity parameters of BP series composites.

Sample	Density (g	Specific heat	Thermal diffusion	TC
	cm <sup>-3</sup> )	(J·g <sup>-1</sup> K <sup>-1</sup> )	coefficient (mm <sup>2</sup> ·s <sup>-1</sup> )	(Wm <sup>-1</sup> K <sup>-1</sup> )
BP-1	1.486	1.050	1.361±0.094	2.123±0.146

BP-2	1.457	1.050	2.071±0.032	3.169±0.049
BP-3	1.463	1.050	2.532±0.117	3.889±0.180
BP-4	1.414	1.050	2.497±0.069	3.707±0.103

## **3D** finite element simulation

Model	Dimension (µm)	TC of PDMS (Wm <sup>-1</sup> K <sup>-1</sup> )	TC of BN (Wm <sup>-1</sup> K <sup>-1</sup> )	TC of composite (Wm <sup>-1</sup> K <sup>-1</sup> )	Specific heat (J·g <sup>-1</sup> K <sup>-1</sup> )
PDMS	300*300*300	0.190	/	/	1.466
rBP-3	300*300*300	/	/	1.170	1.050
BP-3	300*300*300	0.190	50	/	1.050

Table S2. The parameters of the finite element simulation models.

It is worth noting that each grid in the rBP-3 Model is considered as a uniform mixture of BN and PDMS in the modeling process. Therefore, it is only necessary to set the overall TC of this kind of composite.



Figure S2. Arrow direction of heat flux in different models.



Figure S3. Temperature changes on the top surface of the models.



Figure S4. Optical image of heating stage.