### Supporting Information

Leakage-proof nacre-like boron nitride nanosheet/phase change microcapsule

composites with enhanced thermal conduction and thermal energy storage for

advanced thermal managements

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### **Supplementary Figures**



Fig. S1 SEM images and number particle size distribution curves (insets) of T-PCMCs.



**Fig. S2** SEM images of (a) the original BN powder and (b) BNNSs (the inset in b is the magnified SEM image of BNNSs).



Fig. S3 Photos of BNNS/PCMC scaffolds without CMC (a) and with CMC (b).



**Fig. S4** (a) XRD patterns of the nacre-like and random BNNS/PCMC-0 composites (the insets are the schematic representation of relationship between the (002) plane and XRD pattern (left) and the magnified XRD patterns of (100) plane (right), respectively.). (b) SEM images of nacre-like BNNS/PCMC-0 scaffold (I) and composite (II). (c) SEM images of random BNNS/PCMC-0 scaffold (I) and composite (II).



**Fig. S5** (a) Photos of the pure paraffin, 65-BNNS/paraffin-35 and nacre-like 65-BNNS/PCMC-35 composites before and after heated at 80 °C. (b) Mass preservation ratios of the nacre-like 65-

BNNS/PCMC-X and the 65-BNNS/paraffin-35 composites during 100 heating-cooling cycles.



**Fig. S6** DSC thermograms of 65-BNNS/PCMC-X composites (a, crystallization process; b, melting process).



Fig. S7 (a, b) SEM images of the y-z plane of nacre-like 28-BNNS/PCMC-X composites. (c, d) SEM images of the y-z plane of nacre-like 44-BNNS/PCMC-X composites.

BNNS	/PCMC-	0							L			
16.3	29.2	41.8	52.6	60.9	65.9	69.1	70.3	71.6	i	74.4	74.6	80 ∘C
44-BN	NS/PCM	1C-35							L			
16.7	28.4	34.8	36.6	38.1	42.6	51.8	60.7	<mark>63.9</mark>	i	72.5	73.1	
44-BN	NS/PCM	IC-50										
16.5	25.5	32.1	35.7	37.8	38.3	40.8	45.1	51.8	İ	71.2	71.7	
44-BN	NS/PCM	IC-75							ļ			
16.7	24.7	31.0	34.4	37.3	37.9	38.3	38.8	40.3	! i	70.9	71.5	
44-BN	NS/PCM	1C-100							÷			
16.6	23.9	29.6	35.3	38.1	39.1	39.4	39.6	40.6	 	66.7	69.1	10 ºC
0 s	10 s	20 s	30 s	40 s	50 s	60 s	70 s	80 s		160 s	190 s	

Fig. S8 Time-dependent infrared images of the nacre-like 44-BNNS/PCMC-X composites heated on an 80 °C hot plate.

BNNS/	PCMC-0	)										
16.3	29.2	41.8	52.6	60.9	65.9	69.1	70.3	71.6	l i	74.4	74.6	80 °C
28-BN	NS/PCM	C-35							1			
16.8	27.6	30.9	40.4	54.4	61.6	66.6	69.4	71.5	i	73.4	73.5	
28-BN	NS/PCM	C-50							÷			
16.1	25.5	27.9	28.7	30.9	38.8	49.2	55.5	62.2	i	72.1	72.4	
28-BN	NS/PCM	IC-75							i			
16.4	24.0	26.7	27.9	29.3	30.1	31.3	44.3	52.2	ł	71.6	71.8	
28-BN	NS/PCM	IC-100							i			
16.1	21.9	25.9	28.9	29.4	30.7	31.8	34.6	43.6	į	69.0	70.3	10 °C
0 s	10 s	20 s	30 s	40 s	50 s	60 s	70 s	80 s	Ι	160 s	190 s	

Fig. S9 Time-dependent infrared images of the nacre-like 28-BNNS/PCMC-X composites heated on an 80 °C hot plate.



**Fig. S10** (a) Compressive performance of the nacre-like 65-BNNS/PCMC-X composites. (b) Repeated compressive performance of the 65-BNNS/PCMC-35 composite.



**Fig. S11** Application demonstration of the nacre-like 44-BNNS/PCMC composites as the TIMs: (a) The TIMs were integrated between the LED chip and heat sink for heat dissipation demonstration. (b) Time-dependent infrared images of the LED chips using the commercial silicone pad and nacre-like 44-BNNS/PCMC-X composites as the TIMs with a size of 20 mm  $\times$  20 mm  $\times$  20 mm  $\times$  2 mm.



**Fig. S12** Application demonstration of the nacre-like 28-BNNS/PCMC composites as the TIMs: (a) The TIMs were integrated between the LED chip and heat sink for heat dissipation demonstration. (b) Time-dependent infrared images of the LED chips using the commercial silicone pad and nacre-like 28-BNNS/PCMC-X composites as the TIMs with a size of 20 mm  $\times$  20 mm  $\times$  20 mm  $\times$  2 mm.



**Fig. S13** (a) Time-dependent surface temperature curves of the LED chip integrated with the nacre-like 65-BNNS/PCMC-35 composite during 30 on–off cycles. (b) The first and 30th time-dependent surface temperature curves of the LED chip integrated with the nacre-like 65-

BNNS/PCMC-35 composite. (c) DSC thermograms of the original and cycled 65-BNNS/PCMC-X composites. (d) SEM images of the cycled 65-BNNS/PCMC-35 composites (I, y-z plane; II, x-z plane).

# **Supplementary Tables**

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samples	BNNS	CMC	T-PCMCs	$H_2O$
	(g)	(g)	(g)	(g)
BNNS/PCMC-0	5.5	0.55	0	97.5
BNNS/PCMC-10	5.5	0.55	$1.0^{a}$	96.5
BNNS/PCMC-35	5.5	0.55	3.0	94.5
BNNS/PCMC-50	5.5	0.55	5.5	92.0
BNNS/PCMC-75	5.5	0.55	16.5	81.0

### Table S1 Recipes for preparation of the BNNS/PCMC/CMC suspension

<sup>*a*</sup>65-PCMCs.

# Table S2 Properties of the 65-BNNS/PCMC-X composites

	-			
Samples	BNNS density (g·cm <sup>-3</sup> )	CMC density (g·cm <sup>-3</sup> )	65-PCMCs density (g·cm <sup>-3</sup> )	volume fraction of BN (vol%)
65-BNNS/PCMC-0				88
65-BNNS/PCMC-10				64
65-BNNS/PCMC-35	2.23	1.60	0.94	41
65-BNNS/PCMC-50				29
65-BNNS/PCMC-75				12

# Table S3 Thermal diffusivity values of the T-BNNS/PCMC-X composites

Nacre-like samples	$lpha_{ m In-plane}$ $( m mm^2 \cdot s^{-1})$	$lpha_{ m Through-plane} \ ( m mm^2\cdot s^{-1})$
BNNS/PCMC-0	$14.97\pm0.28$	$0.99\pm0.07$
65-BNNS/PCMC-35	$10.28\pm0.31$	$1.36\pm0.07$
65-BNNS/PCMC-50	$4.97\pm0.24$	$1.28\pm0.06$
65-BNNS/PCMC-75	$1.32\pm0.17$	$0.22\pm0.05$
44-BNNS/PCMC-35	$10.67\pm0.23$	$1.19\pm0.07$
44-BNNS/PCMC-50	$5.64\pm0.18$	$1.19\pm0.06$
44-BNNS/PCMC-75	$1.32\pm0.15$	$0.30\pm0.05$
28-BNNS/PCMC-35	$9.86\pm0.23$	$1.29\pm0.07$
28-BNNS/PCMC-50	$4.70\pm0.18$	$1.27\pm0.07$

	DN series (	DCM contract	Isotropic TC	latent heat	DC	
samples	BN content	PCM content	$(W{\cdot}m^{-1}{\cdot}K^{-1})$	$(J \cdot g^{-1})$	References	
PW-WPU-BN-2.5	25.2 wt%	71.5 wt%	0.96	149.0	40	
PLR-PEG-40BN	9.1 wt%	68.2 wt%	1.70	104.2	41	
PLR-PEG-60BN	13.0 wt%	65.2 wt%	2.72	91.2	41	
PW-POE-BN	29.8 vol%	_	3.08	72.0	42	
			through-plane:			
	10 10/	90 vol%	2.62;	121.0	42	
PEG-HBA	10 vol%		in-plane:	121.9	43	
			2.42			
PEG-CS-BN	1.9 wt%	93.7 wt%	0.4	165.3	44	
PUPEG-PW@SiO <sub>2</sub> -BN	16.7 wt%	77.9 wt%	0.675	112.2	45	
OCT-PVA-BN	25.4 wt%	72.7 wt%	0.839	172.0	46	
PEG-MWCNTs-BN	10 wt%	_	1.15	109.1	47	
65-BNNS/PCMC-35	41 vol%	53 vol%	7.89	72.0		
65-BNNS/PCMC-50	29 vol%	68 vol%	4.12	100.3	This work	
65-BNNS/PCMC-75	12 vol%	86 vol%	1.14	151.1		

Table S4	Comparison	of TC values	and laten	t heats of	f 65-BNNS	S/PCMC-X	composites	with
those of th	e reported PO	CM/BN-base	d composi	tes.				

Table S5 Parame	ters for fitting via	the Foygel	nonlinear model

Parameter	Nacre-like 65-BNNS/PCMC-X composites	Random 65-BNNS/PCMC-X composites
$K_0$	13.85	9.81
$V_{\rm c}$	0.012	0.012
τ	1.015	1.095
Р	50	50
L	5 µm	5 µm

D	0.1 µm	0.1 µm
$\overline{A}_{s}$	2.8×10 <sup>-14</sup> m <sup>2</sup>	$2.8 \times 10^{-14} \text{ m}^2$