Bio-inspired spiral-structured poly(vinyl alcohol)/liquid metal/ carbon nanofiber composites for efficient thermal management

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Table S1 Formulations of S-PVA/LM$_x$-CNFs composite films with different LM content.

Sample	PVA (g)	LM (g)	CNFs (g)
S-PVA/LM$_{4.5}$CNFs	6.30	1.43	0.20
S-PVA/LM$_{6.5}$CNFs	6.30	2.11	0.20
S-PVA/LM$_{8.5}$CNFs	6.30	2.82	0.20
S-PVA/LM$_{10.5}$CNFs	6.30	3.56	0.20
S-PVA/LM$_{12.5}$CNFs	6.30	4.33	0.20

Table S2 The detailed $K_{\perp}$ values for the previously reported flexible polymer-based TIMs are systematically summarized.

Sample	Filler	Filler content	$K_{\perp}$	Ref
PDMS/BN	BN	30 wt%	1.24	16
WPU/CNT@LM	CNT@LM	40 vol%	2.19	17
PDMS/PW/BN	BN	13 wt%	2.16	18
PDMS/BN/MXene	BN/MXene	15 wt%	2.03	19
PDMS/BN/LM	BN/LM	40 wt%	2.1	20
PVA/SiC/LM	SiC/LM	40 wt%	0.35	21
PVA/Al_2O_3	Al_2O_3	50 wt%	1.8	22
PVA/MXene@Ag	MXene@Ag	60 wt%	0.41	23
PVA/BNNS/MXene	BNNS/MXene	30 wt%	1.74	24
PVA/LM-CNFs	LM-CNFs	12.5 vol%	2.75	This work
TPU@MD/PVA	MDs	39 vol%	3.31	25
LSR/BN/C-CF	BN/C-CF	10 wt%	3.32	26

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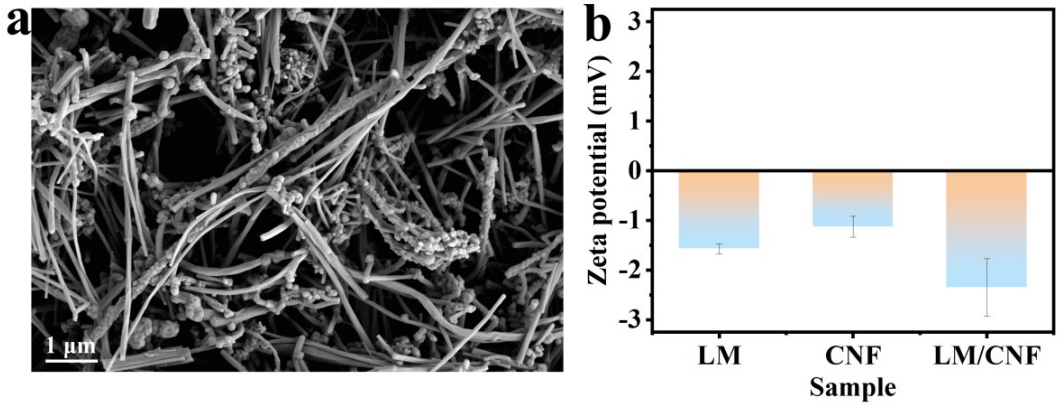

Fig. S1. (a) SEM images of the CNF. (b) Zeta potential of LM, CNF, and LM@CNF.

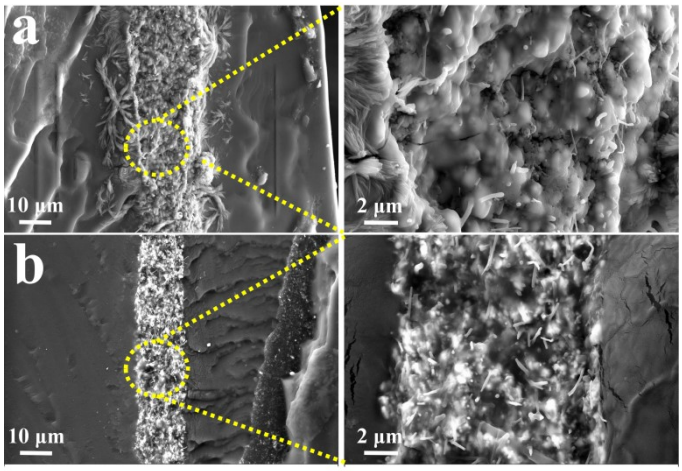

Fig. S2. SEM images of the S-PVA/ $LM_{12.5}$-CNFs composite sample (a) before compression and (b) after compression at 55°C.

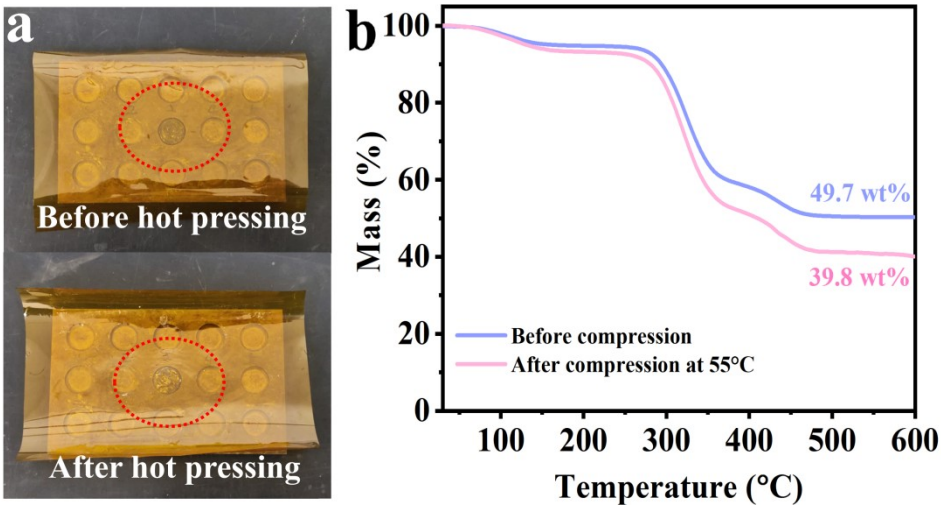

Fig. S3. (a) Digital picture and (b) TG curves of the S-PVA/ $LM_{12.5}$-CNFs composite sample before and after compression at 55°C.

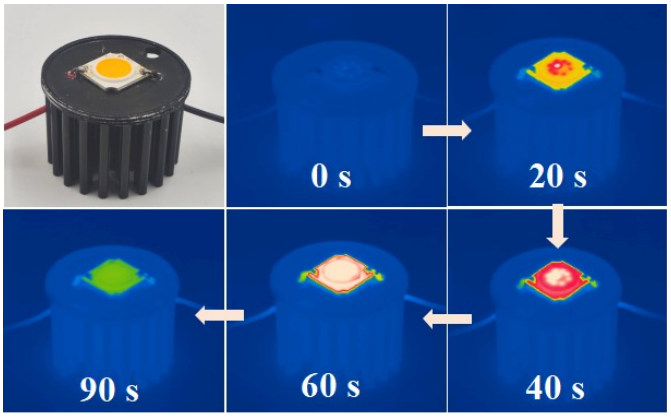

Fig. S4. Infrared thermal images of LED chip for absence of a TIM.

Supplementary method-Finite Element Model

The heat transfer process was simulated using COMSOL Multiphysics 6.1. The models were configured as follows: the chip (heat source) measured 5 cm × 5 cm × 0.3 cm; the TIM 5 cm × 5 cm × 0.2 cm; and the aluminum radiator comprised a 5 cm × 5 cm × 0.3 cm base with 10 fins. The thermal conductivity of the aluminum heat sink was set to 238 W/m·K. Four cases were analyzed: pure PVA, B-PVA/$LM_{12.5}$/CNFs and S-PVA/$LM_{12.5}$-CNFs as a TIM, along with an ideal contact condition. The thermal conductivities were set as follows: 0.49 W/m·K for pure PVA, 1.02 W/m·K for B-PVA/$LM_{12.5}$/CNFs, and 2.75 W/m·K for S-PVA/$LM_{12.5}$-CNFs. A steady-state finite element analysis was employed. The external boundaries of the chip and TIM were defined as adiabatic. The chip power was 5.2 W, with an initial uniform temperature of 297.15 K. A convective heat transfer coefficient of 3 W/(m²·K) was applied to the aluminum radiator, based on a typical value. The ambient temperature and pressure were set to 296.15 K and 1 atm, respectively.

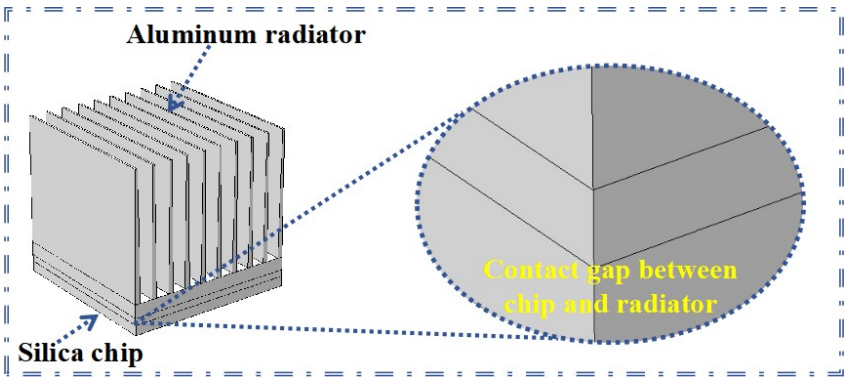

Fig. S5. Schematic representation showing the assembly of an aluminum radiator and chip in the finite element

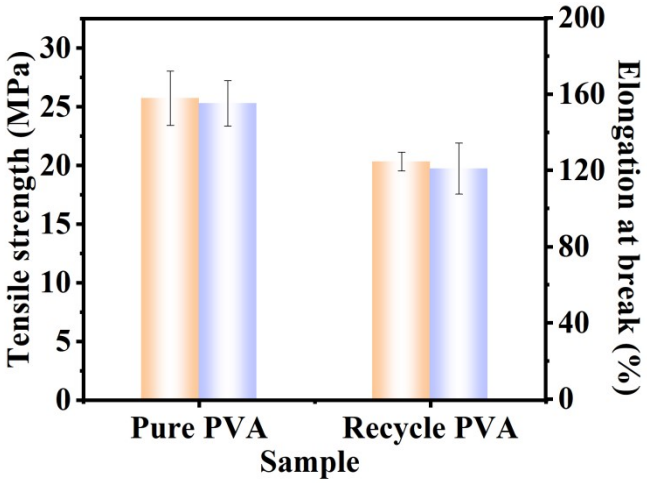

Fig. S6. The tensile strength and elongation at break of S-PVA/ $LM_{12.5}$-CNFs composite .