Supplementary Information (SI) for Journal of Materials Chemistry A.

Supplementary Information

Asymmetric structure endows thermal radiation and heat conduction of graphene film for enhancing dual-mode heat dissipation

Ruo-Han Niu[a,b], Ye-Wen Li[a], Shi-Long Ma[c], Zong-Lin Yi[a], Li-Jing Xie[a], Fang-Yuan Su[a], Hui Jia[a]*, Cheng-Meng Chen[a,d]*

[a] Shanxi Key Laboratory of Carbon Materials, Institute of Coal Chemistry, Chinese Academy of Sciences, Taiyuan, 030001, China.

[b] University of Chinese Academy of Sciences, Beijing 100049, China.

[c] College of Materials Science and Engineering, Taiyuan University of Technology, Taiyuan, 030024, PR China.

[d] Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China.

*Corresponding authors

E-mail addresses: jiahui@sxicc.ac.cn (Hui Jia)

chencm@sxicc.ac.cn (Cheng-Meng Chen)

Scherrer equation:

$$L_c = \frac{K\lambda}{\beta_{002}\cos\theta_{002}}$$

Where K and λ are sample shape constant and X-ray source wavelength (Cu-target, 0.154 nm), respectively. β and θ are full width at half maximum of the (002) peak and Bragg diffractive angle.

Cançado Formula:

$$L_a = (2.4 \times 10^{-10})\lambda_{lasur}^{4}(\frac{I_D}{I_G})^{-1}$$

Where L_a refers to crystalline sizes. E_l is the excitation laser energy used in the Raman experiment in eV units. λ_{laser} is wavelength (532 nm) of laser in Raman testing.

Arithmetic average roughness:

$$R_a = 1/L\int|Z(x)|dx$$

Where R_a refers to arithmetic average roughness, L refers to reference length.

$$S_a = 1/A\int|Z(x,y)|dx\,dy$$

Where S_a refers to amplitude parameter, A refers to reference area.

Table S1. The parameters of thermal conductivity layer and thermal radiation layer for the calculation.

Parameter	Thermal conductivity layer	Thermal radiation layer
Thickness (d)	150 μm	20 μm
Density (ρ)	2.10 g/cm^3	0.33 g/cm^3
The cross-sectional area (S)	2.5 cm*10 cm	
Through-plane thermal conductivity (k)	7 W/(m·K)	7 W/(m·K)

interfacial thermal resistivity ($\theta = d/(kS)$)	0.002 $K \cdot W^{-1}$	0
In-plane thermal conductivity (k)	1000 $W/(m \cdot K)$	

Table S2. Spectral parameters for Raman bands of samples: band position, peak area, and the D band peak aera intensity ratios relative to the G band.

	Parameters	GF	GF@GPI	GF@CBPI
G	Position (cm^{-1})	1583.57	1593	1580
	Area	224549.15	349546.78	505383.83
D1	Position (cm^{-1})	-	1353	1360
	Area		504396.21	1142269.29
	I_{D1}/I_G		1.44	2.26
D2	Position (cm^{-1})		1609	1620
	Area		202285.22	392630.43
	I_{D2}/I_G		0.58	0.78
D3	Position (cm^{-1})		1494	1501
	Area		429170.06	251861.56
	I_{D3}/I_G		1.23	0.50
D4	Position (cm^{-1})		1267	1208
	Area		354296.24	258657.88
	I_{D4}/I_G		1.01	0.51
I_D/I_G	（D1+D2+D3+D4）/G	-	4.26	4.05

Table S3. Elemental content of raw material and films.

Sample	C (wt.%)	H (wt.%)	O (wt.%)	N (wt.%)
graphene	94.52	0.71	0.48	0
carbon black	97.40	0.27	0.14	0
GF	99.80	0.15	0.05	0
GF@PI	71.74	1.86	22.75	3.65
GF@GPI	84.04	0.89	12.91	2.16
GF@CBPI	84.23	0.77	12.26	2.74

Table S4. C1s XPS spectra data of films.

Sample	–C–C (%)	–C–N (%)	–C–O (%)	–C=O (%)
GF	99.95	0	0.05	

GF@PI	50.3	34.82	10.39	4.48
GF@GPI	72.29	6.25	11.91	9.56
GF@CBPI	59.98	10.17	22.69	7.16

Table S5. O1s XPS spectra data of films.

Sample	C=O (%)	C–O–C (%)
GF	-	-
GF@GPI	74.88	25.12
GF@CBPI	68.17	31.83

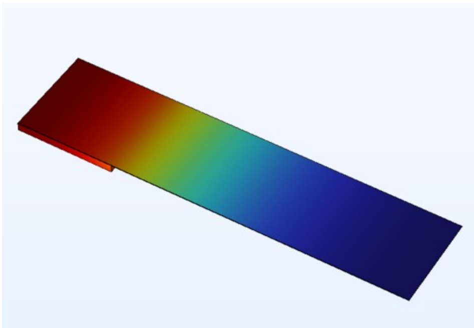

Fig. S1. 3D-finite-element model of the graphene composite film.

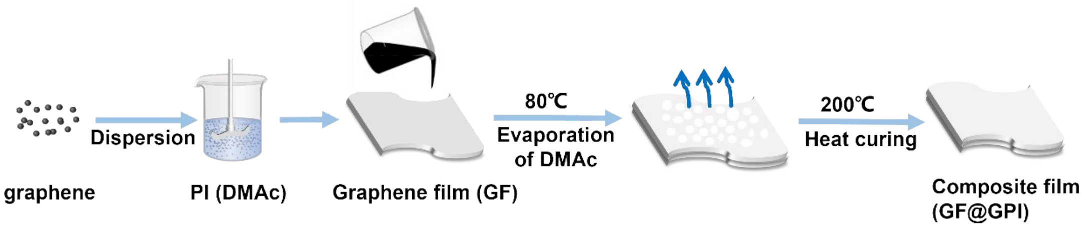

Fig. S2. Fabrication of GF@GPI.

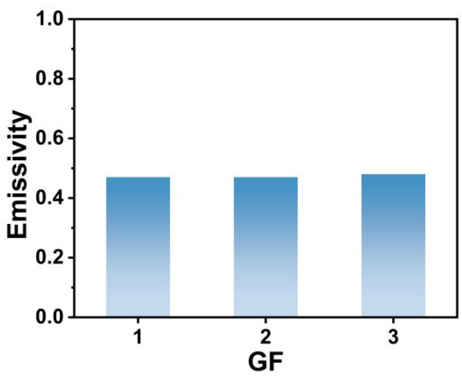

Fig. S3. The emissivity of the graphene film.

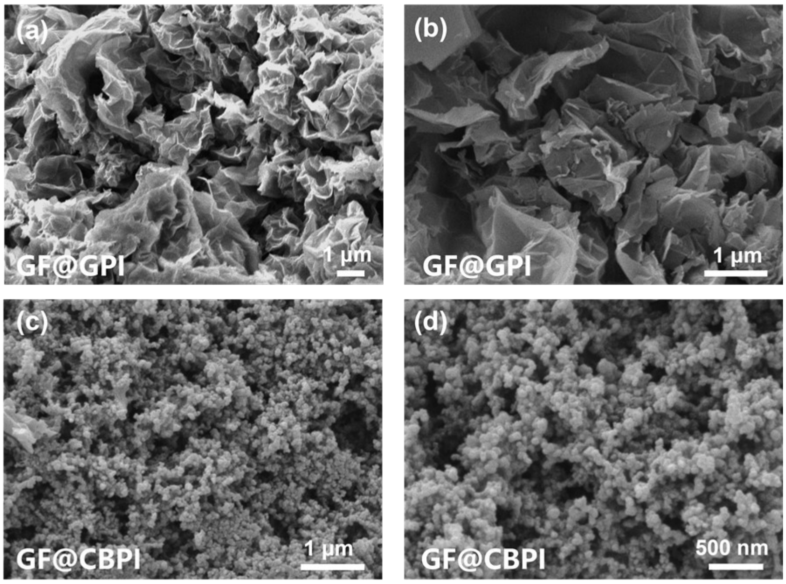

Fig. S4. SEM of GF@GPI and GF@CBPI.

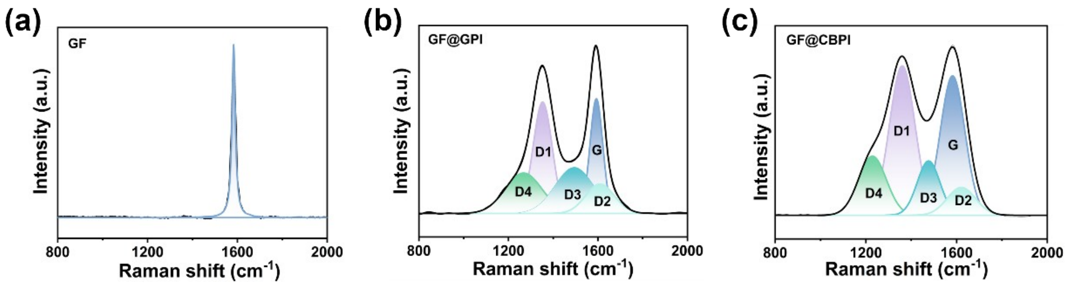

Fig. S5. The Raman spectra of (a) GF, (b) GF@GPI and (c) GF@GPI.

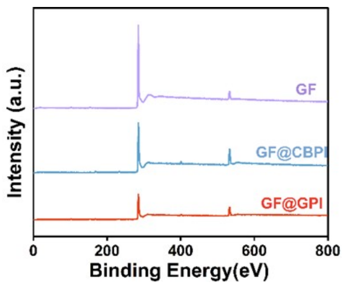

Fig. S6. The XPS spectra of GF, GF@GPI and GF@GPI.

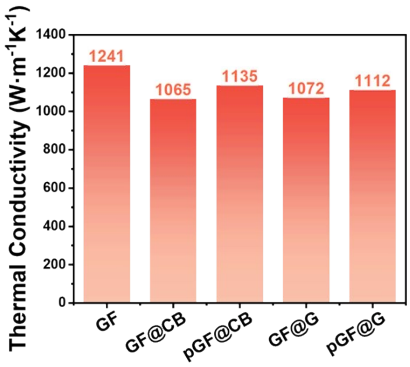

Fig. S7. The thermal conductivity of GF, GF@GPI, pGF@GPI, GF@CBPI, and pGF@CBPI.

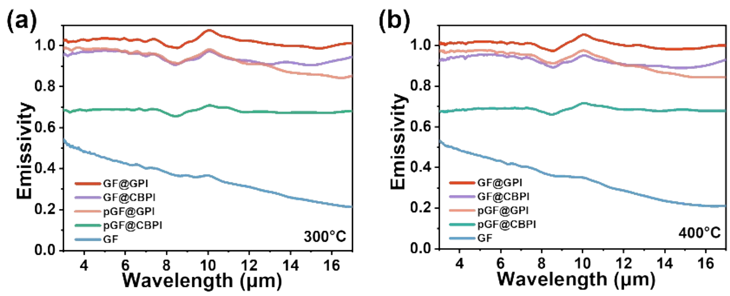

Fig. S8. The emissivity spectrums of films in the wavelength of 2.5–18 μm at (a) 300°C and (b) 400°C.

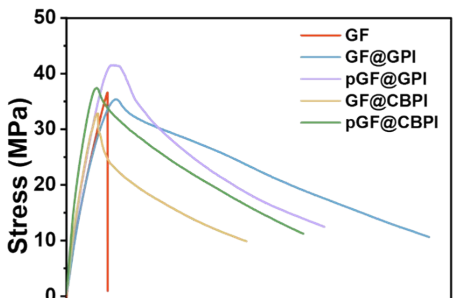

Fig. S9. The tensile strength of GF, GF@GPI, pGF@GPI, GF@CBPI, pGF@CBPI.

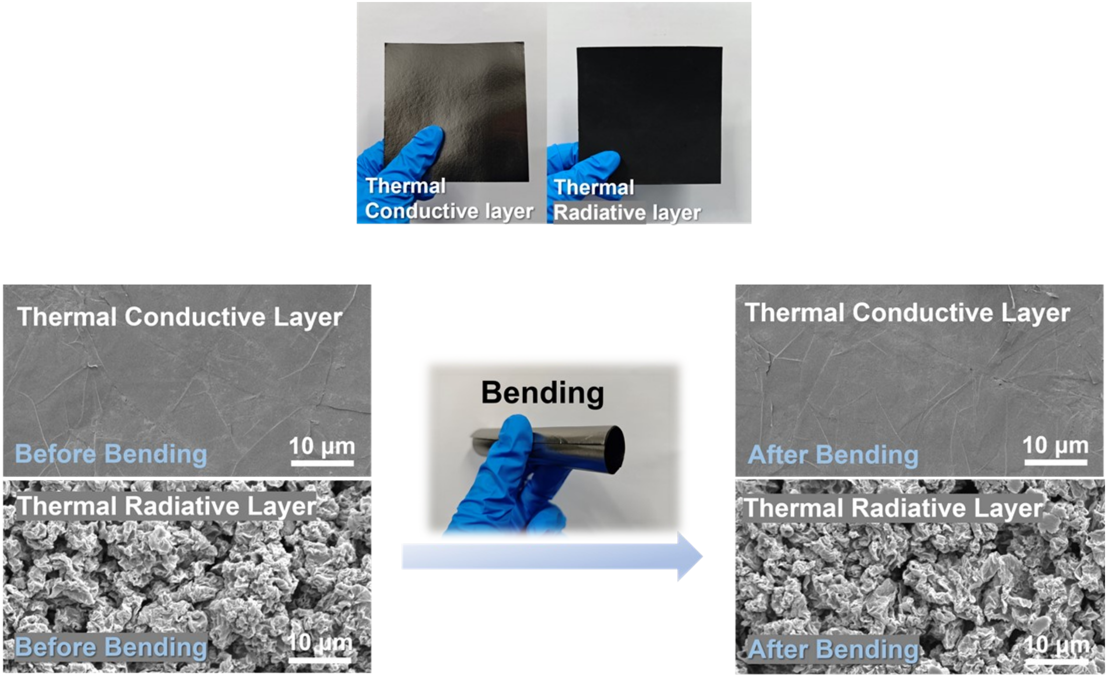

Fig. S10. The digital photograph of GF@GPI and its flexibility.

Fig. S11. Experimental setup of apparent temperature measurement with an input power.

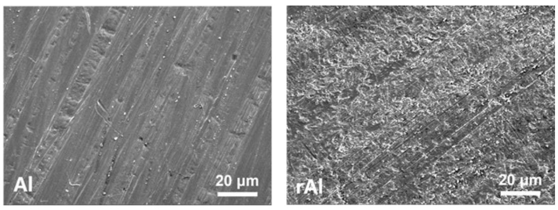

Fig. S12. SEM of Aluminium and Aluminium after roughening.