
Supplementary information

Anisotropic conductive phase change composites enabled by parallel expanded graphite sheets for solar-thermal energy storage

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Note S1. Uncertainty analysis.

The solar-thermal (η_{s-t}) of the PCCs can be calculated using Eq. (S1)^[1,2].

$$\eta_{s-t} = \frac{m \cdot \Delta H}{P \cdot S \cdot \Delta t} \times 100\% \quad (S1)$$

where m is the mass of PCC, g; ΔH is the phase change enthalpy of PCC, J·g⁻¹; P is the simulated solar irradiance, W·m⁻²; S is the surface area of PCC, m²; Δt is the duration of the phase change process, s.

The magnitude of uncertainty reflects the closeness between the measurement result and the true value. In indirect measurements, the uncertainty typically consists of two components: uncertainties from direct measurements and propagation errors. The mathematical foundation of its propagation law lies in the total differential. Assuming the measured physical quantity (N) has a functional relationship with direct measurement quantities ($x, y, z, \dots$) expressed as $N = f(x, y, z, \dots)$, taking its logarithm followed by total differential yields the following Eq. (S2).

$$\frac{dN}{N} = \frac{\partial \ln f}{\partial x} dx + \frac{\partial \ln f}{\partial y} dy + \frac{\partial \ln f}{\partial z} dz + \dots \quad (S2)$$

When the uncertainties of directly measured quantities are mutually independent, the uncertainty (σ) can be derived in Eq. (S3). The relative uncertainty can be expressed as Eq. (S4).

$$\sigma = \sqrt{\left(\frac{\partial f}{\partial x}\right)^2 \sigma_x^2 + \left(\frac{\partial f}{\partial y}\right)^2 \sigma_y^2 + \left(\frac{\partial f}{\partial z}\right)^2 \sigma_z^2 + \dots} \quad (S3)$$

$$\frac{\sigma}{N} = \sqrt{\left(\frac{\partial \ln f}{\partial x}\right)^2 \sigma_x^2 + \left(\frac{\partial \ln f}{\partial y}\right)^2 \sigma_y^2 + \left(\frac{\partial \ln f}{\partial z}\right)^2 \sigma_z^2 + \dots} \quad (S4)$$

In experiments, the solar-thermal efficiency η_{s-t} of the PCCs is a function of the mass of the sample m (± 0.0001 g), the phase change enthalpy ΔH ($\pm 0.2\%$), the solar irradiation intensity P ($\pm 3\%$), the surface area of the sample S (± 0.0001 cm²), the phase change duration Δt (± 5 s). Thus, combining the above equation with these data yields the relative uncertainty (E_η) equation for solar-thermal efficiency η_{s-t} as Eq. (S5)^[3,4]. The maximum relative uncertainty (E_η) of solar-thermal efficiency η_{s-t} is 4.44% as shown in **Table S2**, indicating that the experimental results were reliable.

$$E_\eta = \sqrt{\left(\frac{\delta m}{m}\right)^2 + \left(\frac{\delta \Delta H}{\Delta H}\right)^2 + \left(\frac{\delta P}{P}\right)^2 + \left(\frac{\delta S}{S}\right)^2 + \left(\frac{\delta \Delta t}{\Delta t}\right)^2} \quad (S5)$$

Note S2. Calculations of the mass loss rate and solar absorptivity of the PCC.

Shape stability test was conducted using a constant-temperature heating plate, filter paper, and precision balance. The PCC block was placed on filter paper and subjected to constant-temperature heating at 80 °C for 30 min on the heating plate. The mass and morphology of each block were recorded every 5 min. During heating, some PW leaked from the PCC due to solid-liquid phase transition, which was absorbed by the filter paper, resulting in mass loss. The mass loss rate (γ) can be calculated by Eq. (S6)^[5].

$$\gamma = \frac{(m_t - m_0)}{m_0} \times 100\% \quad (S6)$$

where m_t is the mass of the PCC block at a time point during the heating process, g; m_0 is the initial mass of the PCC block, g.

The solar absorptivity (A) of the upper surface of each sample within the main solar spectrum (0.3-2.5 μm) can be calculated using Eq. (S7) based on its measured spectral absorptivity^[6].

$$A = \frac{\int_{0.3\mu\text{m}}^{2.5\mu\text{m}} A(\lambda) I_{\text{AM1.5}}(\lambda) d\lambda}{\int_{0.3\mu\text{m}}^{2.5\mu\text{m}} I_{\text{AM1.5}}(\lambda) d\lambda} \quad (S7)$$

where λ is wavelength, μm ; $A(\lambda)$ is the spectral absorptivity of the sample surface; $I_{\text{AM1.5}}(\lambda)$ is the spectral irradiance of the AM1.5 reference solar spectral irradiance^[7], $\text{W} \cdot \text{m}^{-2} \cdot \mu\text{m}^{-1}$.

Note S3. Parallel and series heat conduction models.

The volumetric proportion of EG (φ_v) in the PCC can be calculated from its mass proportion (φ_{wt}) through Eq. (S8).

$$\varphi_v = \frac{\frac{\varphi_{\text{wt}}}{\rho_{\text{EG}}}}{\frac{\varphi_{\text{wt}}}{\rho_{\text{EG}}} + \frac{1 - \varphi_{\text{wt}}}{\rho_{\text{PW-OBC}}}} \quad (S8)$$

where ρ_{EG} and $\rho_{\text{PW-OBC}}$ are the densities of the EG and PW-OBC sheets, respectively.

To define the upper and lower bounds of thermal resistance along the heat flow path, parallel and series thermal conduction models were established (**Figure S4**). These bounds directly determine the minimum and maximum achievable heat transfer rates. The lengthwise (R_{par}) and transverse (R_{ser}) thermal resistances are given in Eq. (S9) and Eq. (S10), respectively.

$$\frac{1}{R_{\text{par}}} = \frac{\varphi_v}{R_{\text{EG}}} + \frac{(1 - \varphi_v)}{R_{\text{PW-OBC}}} \quad (S9)$$

$$R_{\text{ser}} = \varphi_v R_{\text{EG}} + (1 - \varphi_v) R_{\text{PW-OBC}} \quad (\text{S10})$$

Where R_{EG} and $R_{\text{PW-OBC}}$ are the thermal resistances of the EG sheet and PW-OBC mixture, respectively.

The ratio λ_L/λ_T represents the thermal conductive anisotropy, where λ_T and λ_L denote the transverse and longitudinal conductivities, respectively. Based on the series and parallel heat conduction models, its expression can be given by Eq. (S11).

$$\frac{\lambda_L}{\lambda_T} = \left(\frac{\lambda_{\text{PW-OBC}}}{\lambda_{\text{EG}}} + \frac{\lambda_{\text{EG}}}{\lambda_{\text{PW-OBC}}} \right) \varphi_v (1 - \varphi_v) + (1 - \varphi_v)^2 + \varphi_v^2 \quad (\text{S11})$$

where λ_{EG} and $\lambda_{\text{PW-OBC}}$ refer to the thermal conductivities of the EG and PW-OBC sheets, respectively.

Supporting Figures.

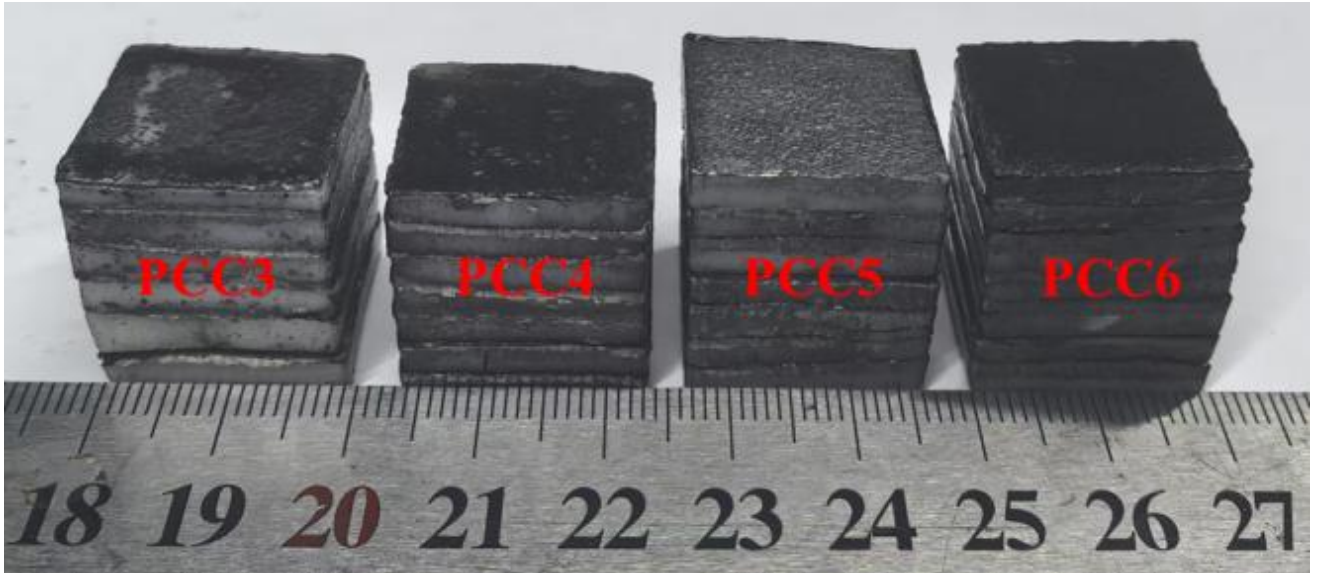


Figure S1. Photographs of PCC3-PCC6 samples with varying expanded graphite (EG) proportion.

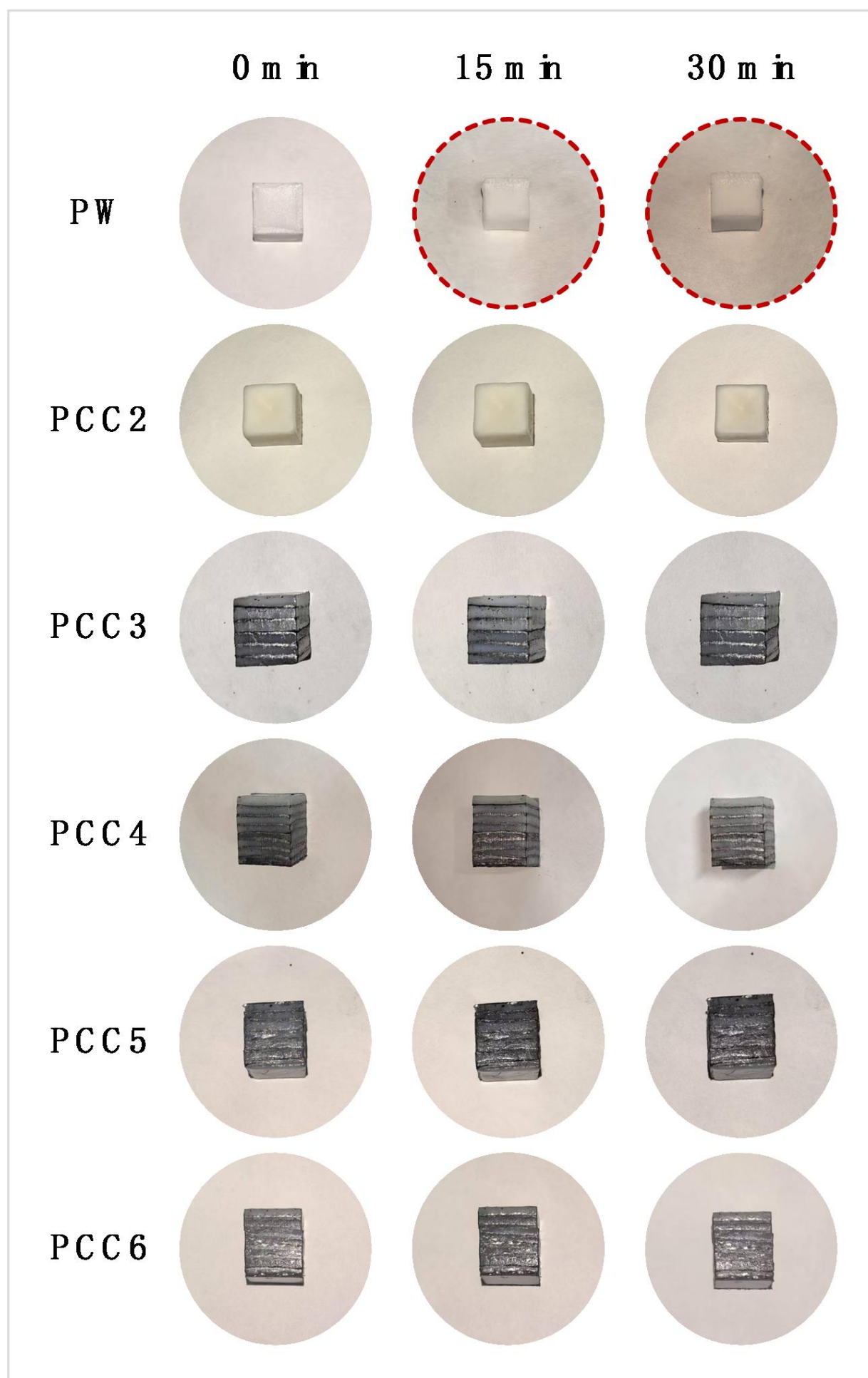


Figure S2. Time-lapse photos of PW and PCC samples with dimensions $20 \times 20 \times 20 \text{ mm}^3$.

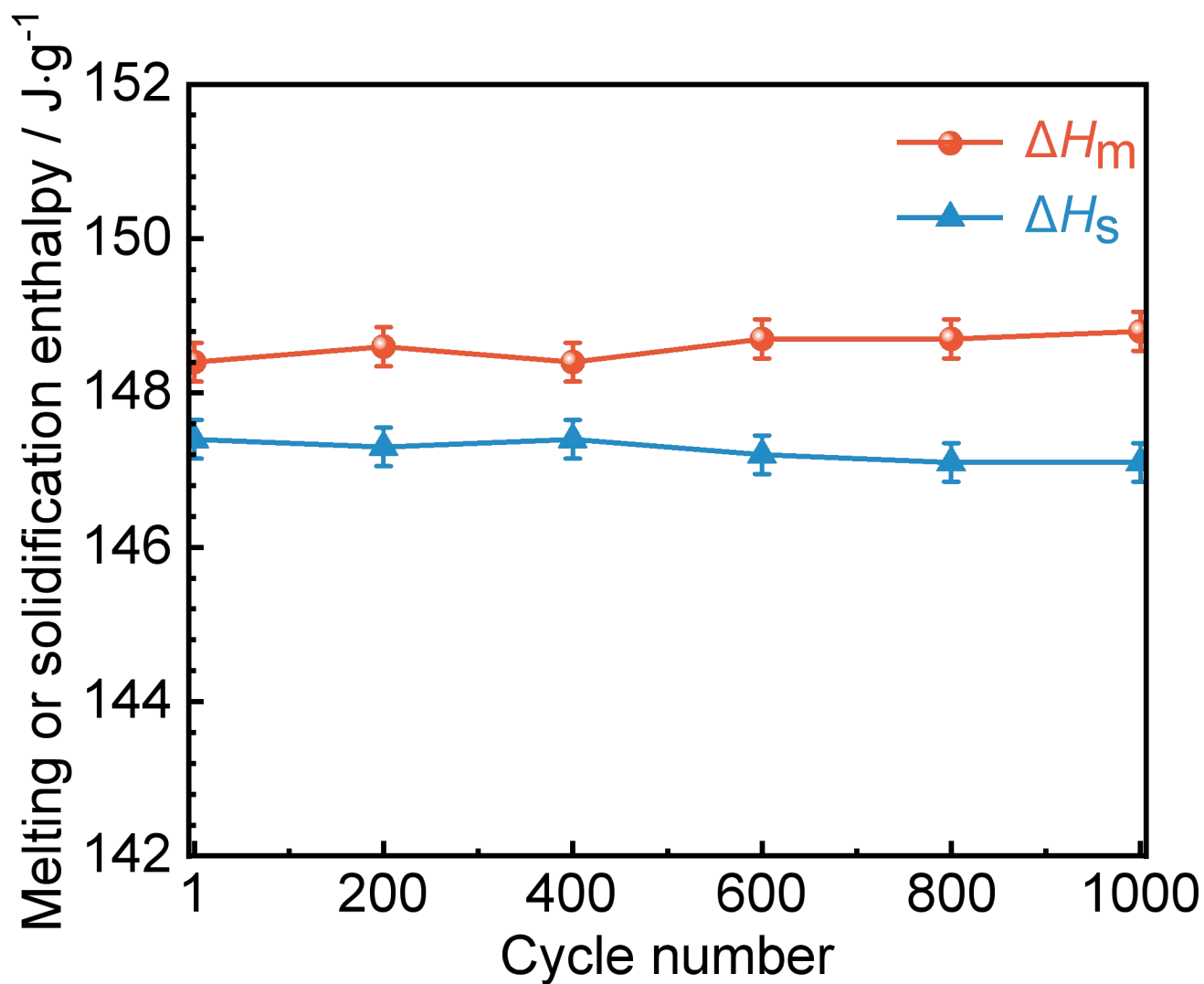


Figure S3. The Measured melting (ΔH_m) and solidification (ΔH_s) enthalpies of the PCC6 under 1,000 heating/cooling cycles within 20 °C-90 °C.

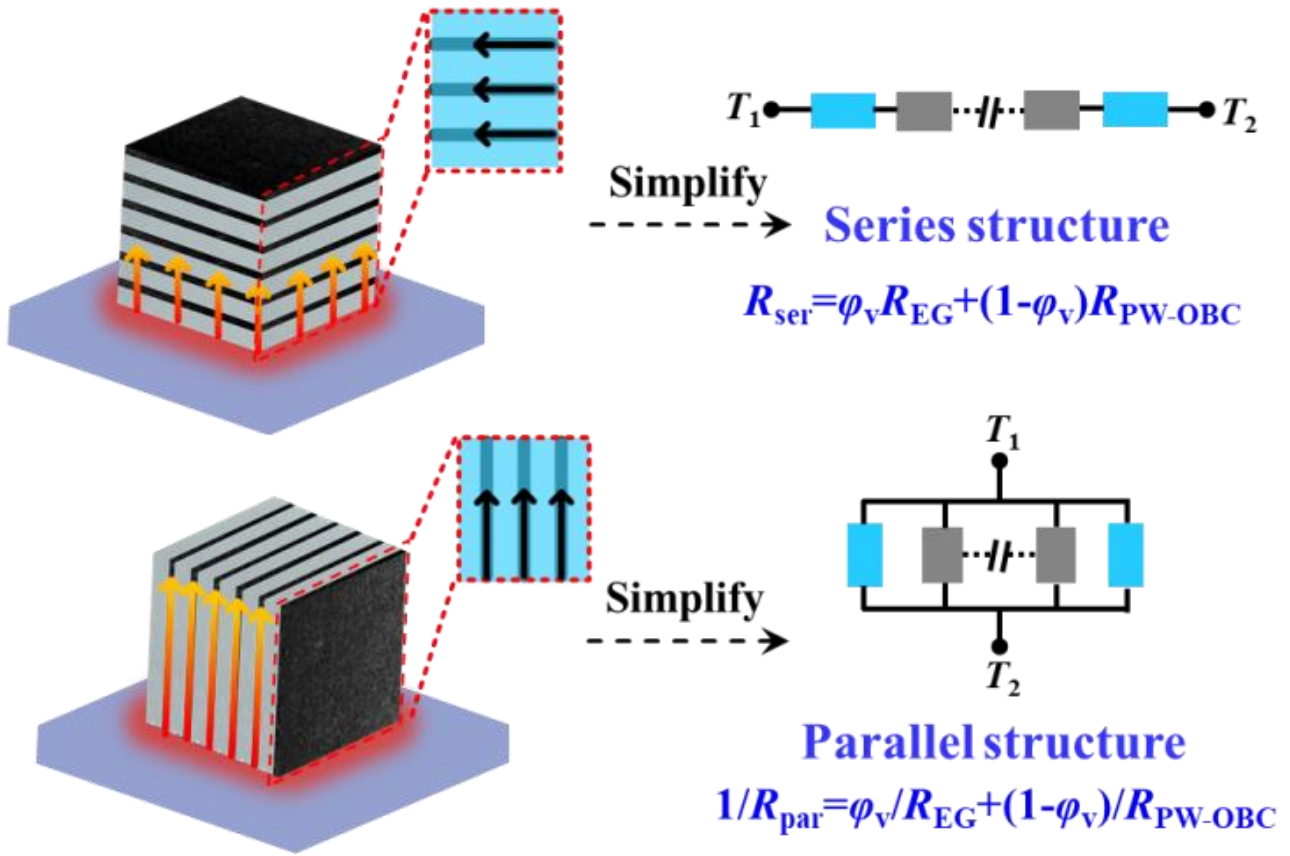


Figure S4. Schematic diagrams of the PCC's series and parallel thermal models.

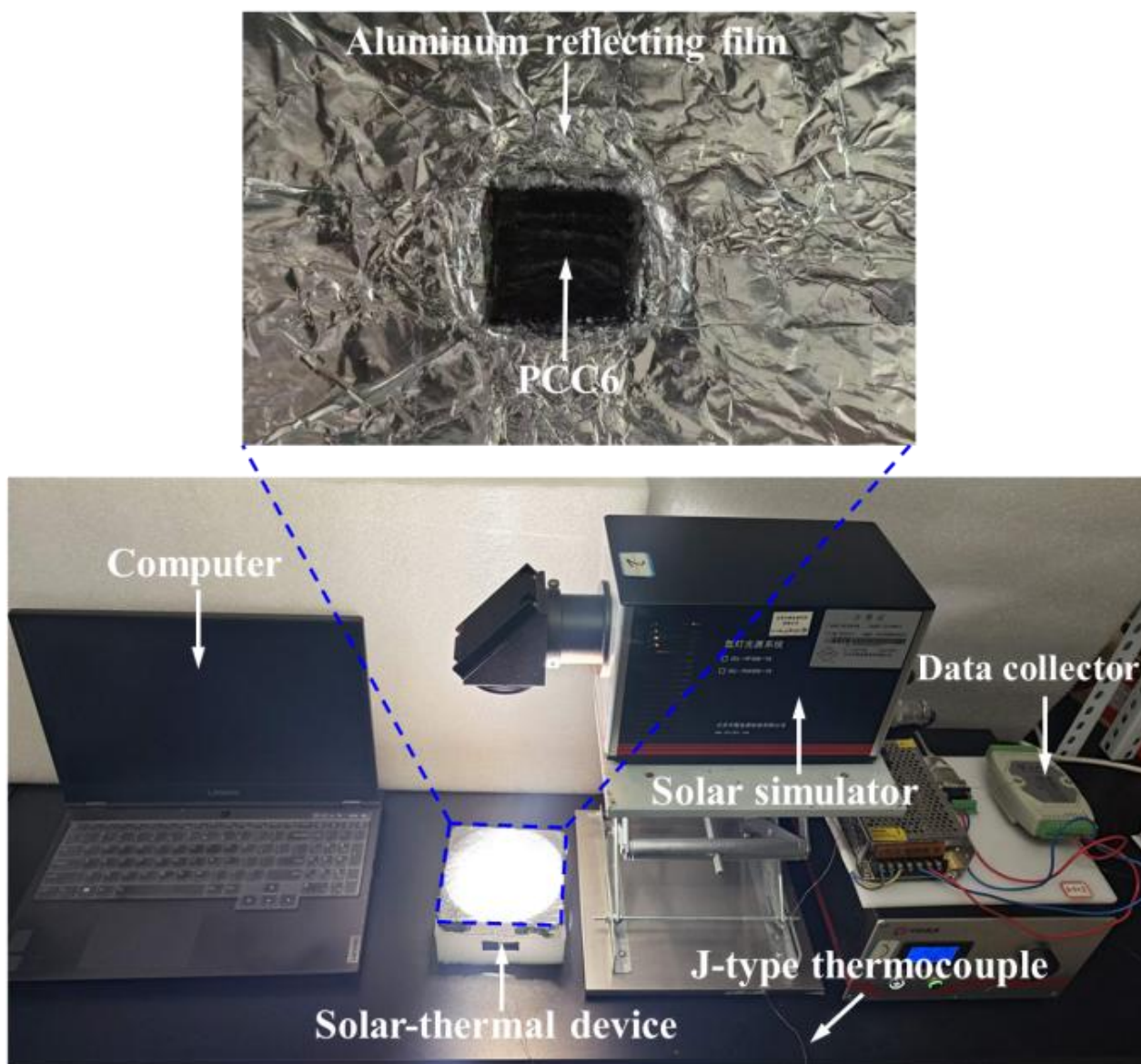


Figure S5. Photos showing the PCC-based direct solar-thermal energy storage device (left) and the solar-thermal testing setup (right).

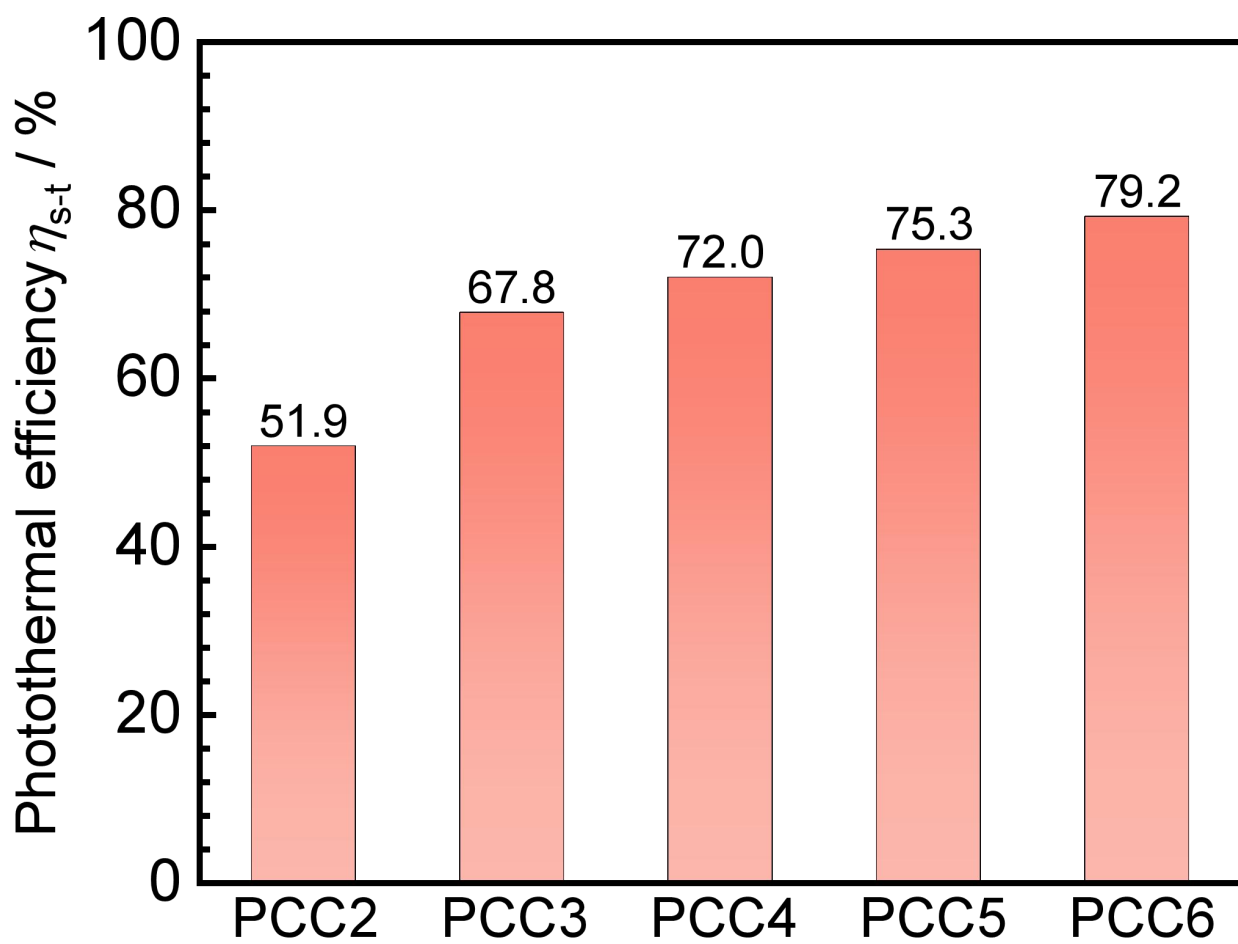


Figure S6. Solar-thermal efficiencies of the PCCs under 1 sun.

Supporting Tables.

Table S1. Thermal conductivities of present PCCs and some recently open-published PCCs.

$\lambda_L/W\cdot m^{-1}\cdot K^{-1}$	$\lambda_T/W\cdot m^{-1}\cdot K^{-1}$	Thermal conductive Anisotropy λ_L/λ_T	Reference
5.63	0.77	7.31	[3]
5.84	1.34	4.36	[6]
10.55	2.60	4.06	[8]
6.75	2.92	2.31	[9]
1.2453	0.1933	6.44	[10]
0.8	0.5	1.6	[11]
21.3	8.29	2.57	[12]
15.5	10.1	1.53	[13]
6.31	2.95	2.14	[14]
1.84	1.38	1.33	[15]
8.87	2.68	3.31	[16]
1.73	0.62	2.79	[17]
4.54	0.49	9.27	This work

Table S2. Results of uncertainty analysis.

Sample	m (g)	ΔH (J·g ⁻¹)	P (kW·m ⁻²)	S (cm ²)	Δt (s)	E_η (%)
PCC2	1.3343	189.5	1.0	4.0	1,217	3.03
PCC3	1.2279	180.1	1.0	4.0	815	3.07
PCC4	1.2770	169.7	1.0	4.0	752	3.08
PCC5	1.2091	161.0	1.0	4.0	646	3.10
PCC6	1.1935	148.4	1.0	4.0	559	3.14
PCC6 _L with glass	1.1935	148.4	1.0	4.0	556	3.14
PCC6 _H	1.3155	148.4	1.0	4.0	737	3.08
PCC6 _L	1.1935	148.4	1.5	4.0	353	3.32

PCC6 _L with glass	1.1935	148.4	1.5	4.0	351	3.33
PCC6 _H	1.3155	148.4	1.5	4.0	453	3.20
PCC6 _L	1.1935	148.4	2.0	4.0	247	3.62
PCC6 _L with glass	1.1935	148.4	2.0	4.0	248	3.62
PCC6 _H	1.3155	148.4	2.0	4.0	320	3.39
PCC6 _L	1.1935	148.4	2.5	4.0	189	4.00
PCC6 _L with glass	1.1935	148.4	2.5	4.0	191	3.99
PCC6 _H	1.3155	148.4	2.5	4.0	250	3.61
PCC6 _L	1.1935	148.4	3.0	4.0	153	4.44
PCC6 _L with glass	1.1935	148.4	3.0	4.0	156	4.39
PCC6 _H	1.3155	148.4	3.0	4.0	200	3.91

Table S3. Mass values of PW and different PCCs at different time points.

Time/min	$m_{\text{PCC1/g}}$	$m_{\text{PCC2/g}}$	$m_{\text{PCC3/g}}$	$m_{\text{PCC4/g}}$	$m_{\text{PCC6/g}}$
0	7.298	7.338	7.363	7.619	8.128
5	7.286	7.333	7.362	7.617	8.124
10	7.227	7.330	7.358	7.614	8.122
15	7.214	7.329	7.357	7.612	8.119
20	7.108	7.323	7.354	7.608	8.116
25	7.106	7.323	7.351	7.607	8.113
30	7.104	7.322	7.350	7.604	8.112

Table S4. Phase change temperatures and enthalpies of the PW and PCCs.

Sample	PW content/wt.%	$T_m/^{\circ}\text{C}$	$\Delta H_m, \Delta H_{pre}/\text{J}\cdot\text{g}^{-1}$	$T_s/^{\circ}\text{C}$	$\Delta H_s, \Delta H_{pre}/\text{J}\cdot\text{g}^{-1}$
PW	100.00	52.9	249.9, 249.9	46.3	248.9, 248.9
PCC2	75.00	53.7	189.5, 187.4	37.6	187.9, 186.7
PCC3	71.25	53.7	180.1, 178.1	38.9	177.6, 177.3
PCC4	67.50	51.2	169.7, 168.7	41.4	167.2, 168.0
PCC5	63.75	53.2	161.0, 159.3	40.4	160.3, 158.7
PCC6	60.00	52.1	148.4, 149.9	40.6	147.4, 149.3

Table S5. Thermal conductivity enhancing factors (α_i) in this work and existing studies.

Base material	Filler	$\phi_{wt}/\%$	$\lambda_{PCM}/\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$	$\lambda_{PCC}/\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$	$\alpha_i/\%$	Reference
PW/OBC	CFs	10	0.21	2.23	106.19	[3]
OC	CF/CNT	11	0.26	1.31	45.80	[18]
Erythritol	SCFs	10	0.77	3.91	50.78	[19]
SAT	EPCu	5.7	0.59	3.22	95.75	[20]
SA	rGO	3	0.41	1.25	101.63	[21]
Mg(NO ₃) ₂ ·6H ₂ O/ g-C ₃ N ₄	GS	10	0.38	2.43	63.95	[22]
PEG	PVA-rGO	3.1	0.221	0.348	50.80	[23]
PW	SCA	13	0.23	0.43	14.38	[24]
mE/2DNH	EG	20	0.714	12.36	86.55	[25]
C14/PCL	EG	15	0.224	3.702	110.18	[26]
PA/PE	EG	10	0.25	1.9955	79.82	[27]
PW	HPC2.0	16.04	0.27	0.33	7.62	[28]

PA	CFs	20	0.23	5.21	113.26	[14]
Paraffin wax/ Olefin block EG copolymer		0	0.18	-	-	This work
		5	0.18	1.20	135.33	This work
		10	0.18	2.28	126.67	This work
		15	0.18	3.24	120.00	This work
		20	0.18	4.54	126.11	This work

Table S6. The solar-thermal energy conversion and storage performance of the PCC6_L.

Solar irradiation $P/W \cdot m^{-2}$	1,000	1,500	2,000	2,500	3,000
Phase change duration $\Delta t/s$	559	353	247	189	153
Surface area S/cm^2	4.0				
Mass m/g	1.19				
Input energy/J	223.6	211.8	197.6	189.0	183.6
Stored energy/J	177.1				
Solar-thermal efficiency $\eta_{s-t}/\%$	79.2	83.6	89.6	93.7	96.5

Table S7. The solar-thermal energy harvesting performance of the PCC6_T.

Solar irradiation $P/W \cdot m^{-2}$	1,000	1,500	2,000	2,500	3,000
Phase change duration $\Delta t/s$	737	453	320	250	200
Surface area S/cm^2	4.0				
Mass m/g	1.32				
Input energy/J	294.8	271.8	256.0	250.0	240.0
Stored energy/J	195.2				
Solar-thermal efficiency $\eta_{s-t}/\%$	66.2	71.8	76.3	78.1	81.3

Table S8. The solar-thermal energy harvesting performance of the PCC6_L with glass.

Solar irradiation $P/W \cdot m^{-2}$	1,000	1,500	2,000	2,500	3,000
Phase change duration $\Delta t/s$	556	351	248	191	156
Surface area S/cm^2	4.0				
Mass m/g	1.19				
Input energy/J	222.4	210.6	198.4	191.0	187.2
Stored energy/J	177.1				
Solar-thermal efficiency $\eta_{s-t}/\%$	79.6	84.1	89.3	92.7	94.6

Table S9. Solar-thermal efficiencies of present PCCs and some open-published PCCs.

PCC	Phase change temperature/ $^{\circ}C$	Solar irradiation /sun	Solar-thermal efficiency $\eta_{s-t}/\%$	Reference
TME/EG	83.1	3.0	69.49	[29]
OC/CF/CNT	58.21	3.0	82.6	[18]
SAT/EPCu	50.89	1.2	80.81	[20]
Ag-MWCNTs/PW@CNS	60.91	1.2	73.1	[30]
PEG/PVA-GO	59.1	1.2	82.3	[23]
LA/GA	43.31	1.0	74.6	[31]
CNT/PDA@SiO ₂ -MEPCM	37.3	3.0	90.1	[32]
mE/2DHN/EG	119.57	3.0	75.72	[25]
Erythritol-PAAS/GO	119	1.0	69.2	[8]
HPC2.0/PW	52.36	2.5	38.0	[28]
HPC2.0/PW	52.36	3.0	89.0	[28]
PW/OBC/CFs	56	1.0	79.17	[3]
PW-OBC/EG	52.1	1.0	79.2	This work
PW-OBC/EG	52.1	1.5	83.6	This work
PW-OBC/EG	52.1	2.0	89.6	This work
PW-OBC/EG	52.1	2.5	93.7	This work
PW-OBC/EG	52.1	3.0	96.5	This work

Noting: TME-Trimethylolethane, EG-Expanded graphite, OC-octadecanol, CF-Carbon foam, CNT-Carbon nanotube, SAT- Sodium acetate trihydrate, EPCu-Modified expanded graphite, Ag-MWCNTs/PW@CNS-Silver-decorated carboxyl multi-wall carbon nanotubes/paraffin wax

@Carbon nanotube sponge, PEG-Poly ethylene glycol, PVA-Polyvinyl alcohol, GO-Graphene oxide, LA-Lauric acid, GA-Graphene aerogel, PDA-Polydopamine, SiO₂-MEPCM-Silica hierarchical shell, mE-Meso-erythritol, 2DHN-two-dimensional heterogeneous nanohybrids, PAAS-Polyamic acid salt, HPC-Hierarchical porous carbon, PW-Paraffin wax, OBC- Olefin block copolymer, CFs-Carbon fibers.

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