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Supplementary Materials for
A new strategy for balancing thermal stability and latent heat of solid-liquid PCMs via a solid-solid PCM as a supporting skeleton

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Exploration experiment on the optimal ratio of PESH-CPCM

In order to explore the optimal ratio of SH-skeleton and PA, as well as the optimal amount of EG added, some exploratory experiments were conducted in advance. Firstly, SH-skeleton was used as the matrix material and different contents of EG were compounded, denoted as SH-skeleton/EG-xwt% (x represents the quality proportion of EG, x=0，2，4，6，8). The thermal conductivity of SH-skeleton/EG-xwt% was tested, and the test results are shown in **Table S1**. As the amount of EG added continues to increase, the thermal conductivity of SH-skeleton/EG-xwt% also increases. In the case of 8wt% EG added, the thermal conductivity of SH-skeleton/EG-8wt% reaches 2.45 W·m^{-1}·K^{-1}. However, during the experiment, we found that compared to the addition of 6 wt% EG, the addition of 8 wt% EG

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significantly prolonged the stirring time to achieve uniformity, which reduced the preparation efficiency. In addition, according to a large number of reported works, when the thermal conductivity of CPCM is greater than 2 $W \cdot m^{-1} \cdot K^{-1}$, it can meet many application scenarios, including battery thermal management. Therefore, based on the comprehensive evaluation of thermal conductivity and preparation efficiency, the optimal amount of EG added was determined to be 6wt%. Next, determining the optimal ratio of SH Skeleton and PA is another key issue. Samples with different PA contents were prepared and denoted as SH-skeleton/EG/PA ywt% (y represents the quality proportion of PA, y=0, 10, 20, 30, 40, 50). The specific composition information is shown in **Table S2**. During the preparation process, we found that when PA reached 50wt%, SH-skeleton/EG/PA-50wt% could not polymerize after heating. This is due to the high PA content, which prevents the formation of a continuous cross-linked network between styrene acrylate (SA) and hexamethylene diacrylate (HA). The latent heat of SH-skeleton/EG/PA-ywt% (y=0, 10, 20, 30, 40) was tested using DSC, and the test results are shown in **Figure S1a-S1e** and **Table S2**. SH-skeleton/EG/PA-40wt% has the highest latent heat, which is attributed to the higher latent heat of pure PA compared to SH-skeleton. In addition, the thermal stability of SH-skeleton/EG/PA-ywt% (y=0, 10, 20, 30, 40) was also evaluated by heating them on a 60 °C platform for 2 h, recording the mass loss rate after heating, and taking photos to compare the leakage traces before and after heating, as shown in Figure S1f and S2. Compared to the SH-skeleton/EG/PA-30wt%, the mass loss rate of SH-skeleton/EG/PA-40wt% is as high as 0.14 wt%, an increase of 20 %, and its leakage traces are also more clearly visible.

After considering the thermal conductivity, preparation efficiency, thermal stability, and other properties of CPCM, the composition ratio of PESH-CPCM was determined to be 64wt% SH Skeleton, 30wt% PA, and 6wt% EG, which were used for subsequent performance characterization and application research.

Table S1. Composition ratio and thermal conductivity of SH-skeleton/EG-xwt%.

	SH-skeleton (wt%)	EG (wt%)	Thermal conductivity ($W \cdot m^{-1} \cdot K^{-1}$)
SH-skeleton/EG-0wt%	100	0	0.29
SH-skeleton/EG-2wt%	98	2	1.20
SH-skeleton/EG-4wt%	96	4	1.67
SH-skeleton/EG-6wt%	94	6	2.16
SH-skeleton/EG-8wt%	92	8	2.45

Table S2. Composition ratio and latent heat of SH-skeleton/EG/PA-ywt%

	SH-skeleton (wt%)	PA (wt%)	EG (wt%)	Latent heat ($J \cdot g^{-1}$)
SH-skeleton/EG/PA-0wt%	94	0	6	88.79
SH-skeleton/EG/PA-10wt%	84	10	6	97.26
SH-skeleton/EG/PA-20wt%	74	20	6	103.19
SH-skeleton/EG/PA-30wt%	64	30	6	112.02
SH-skeleton/EG/PA-40wt%	54	40	6	125.80
SH-skeleton/EG/PA-50wt%	44	50	6	--

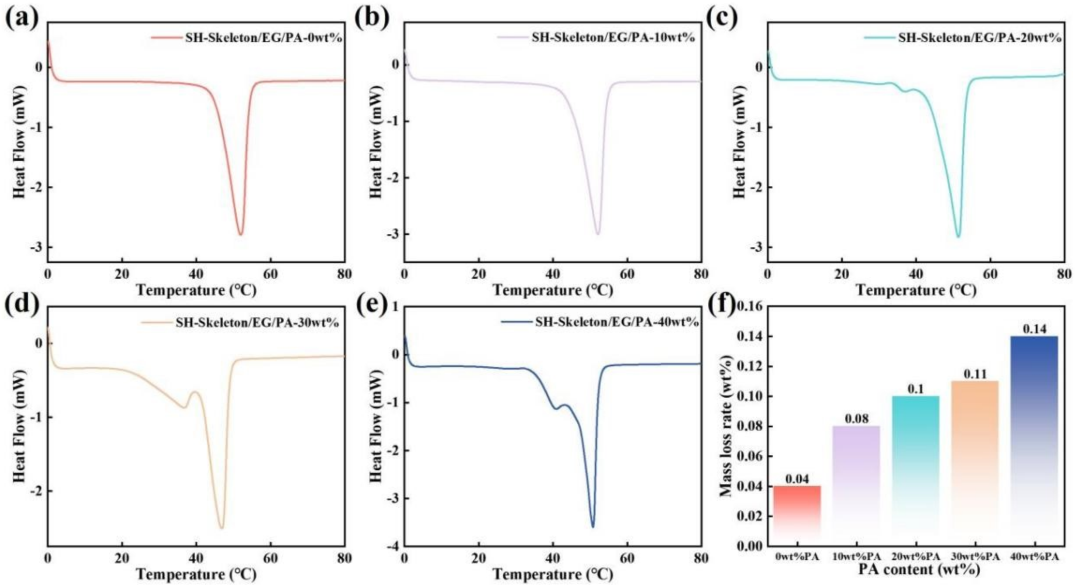

Figure S1. DSC curves of SH-skeleton/EG/PA-ywt% (y=0, 10, 20, 30, 40), (a) 0 wt%PA; (b) 10 wt%PA; (c) 20 wt%PA; (d) 30 wt%PA; (e) 40 wt%PA. (f) Mass loss rate of SH-skeleton/EG/PA-ywt% (y=0, 10, 20, 30, 40) after heating at 60 °C for 2 h.

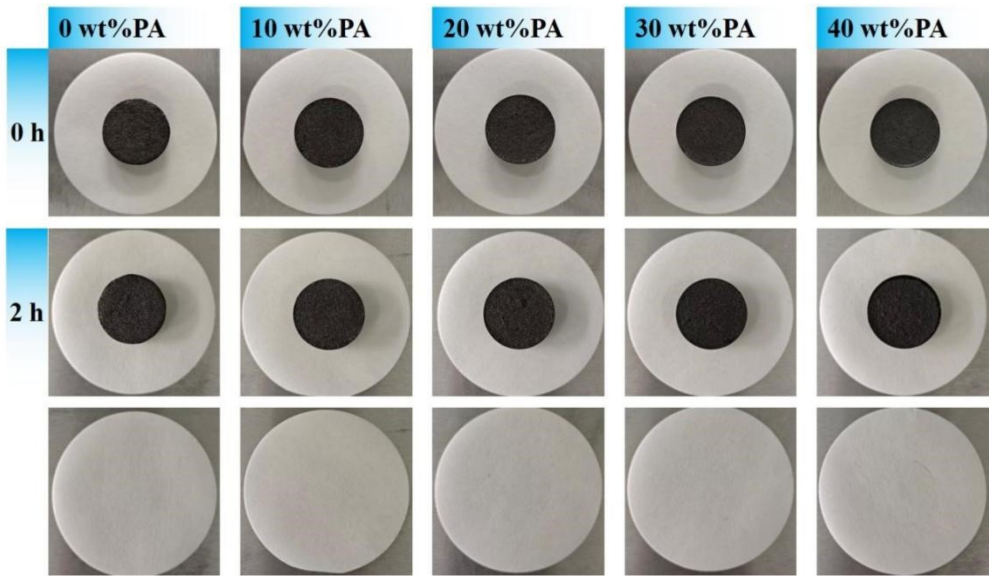

Figure S2. Shape changes and leakage traces of SH-skeleton/EG/PA-ywt% (y=0, 10, 20, 30, 40) after heating

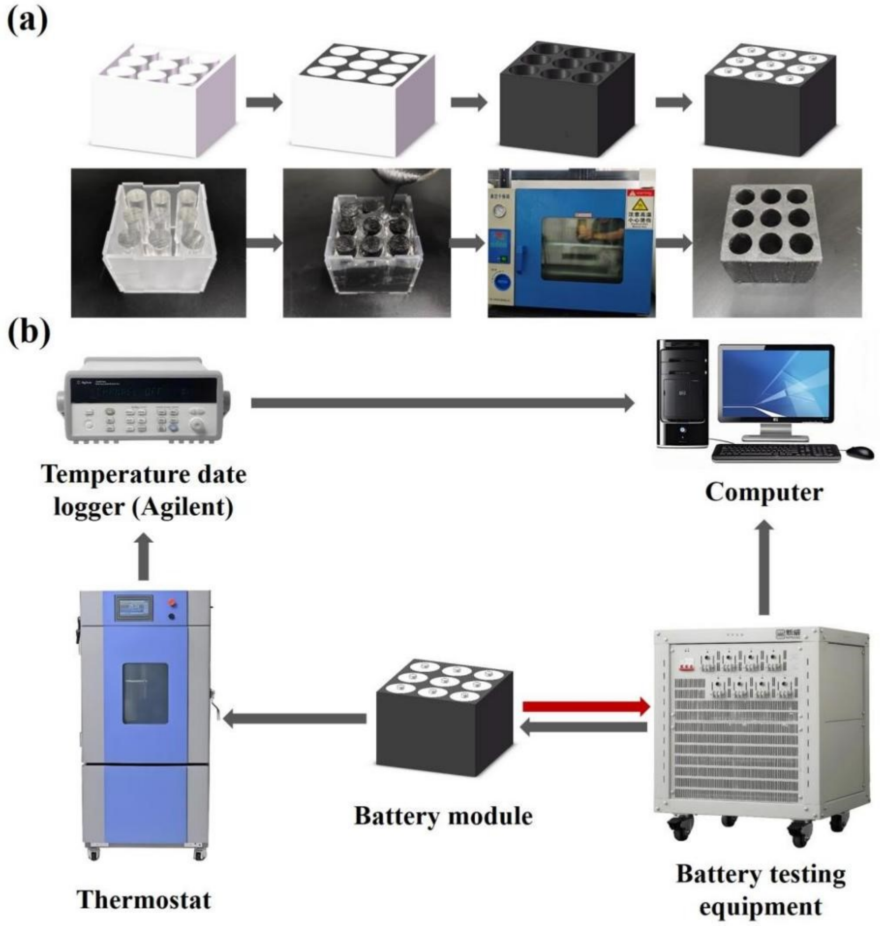

Figure S3 (a) The assembly process of battery module. (b) The schematic diagram of battery module testing platform.

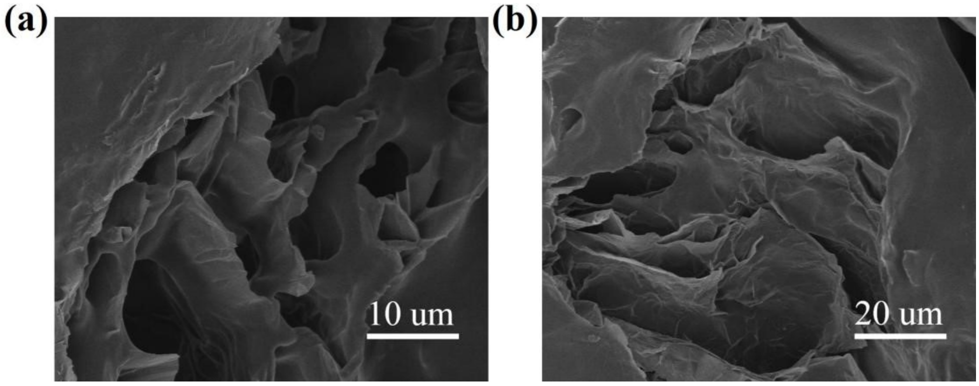

Figure S4. SEM image of PESH-CPCM (PA removed)

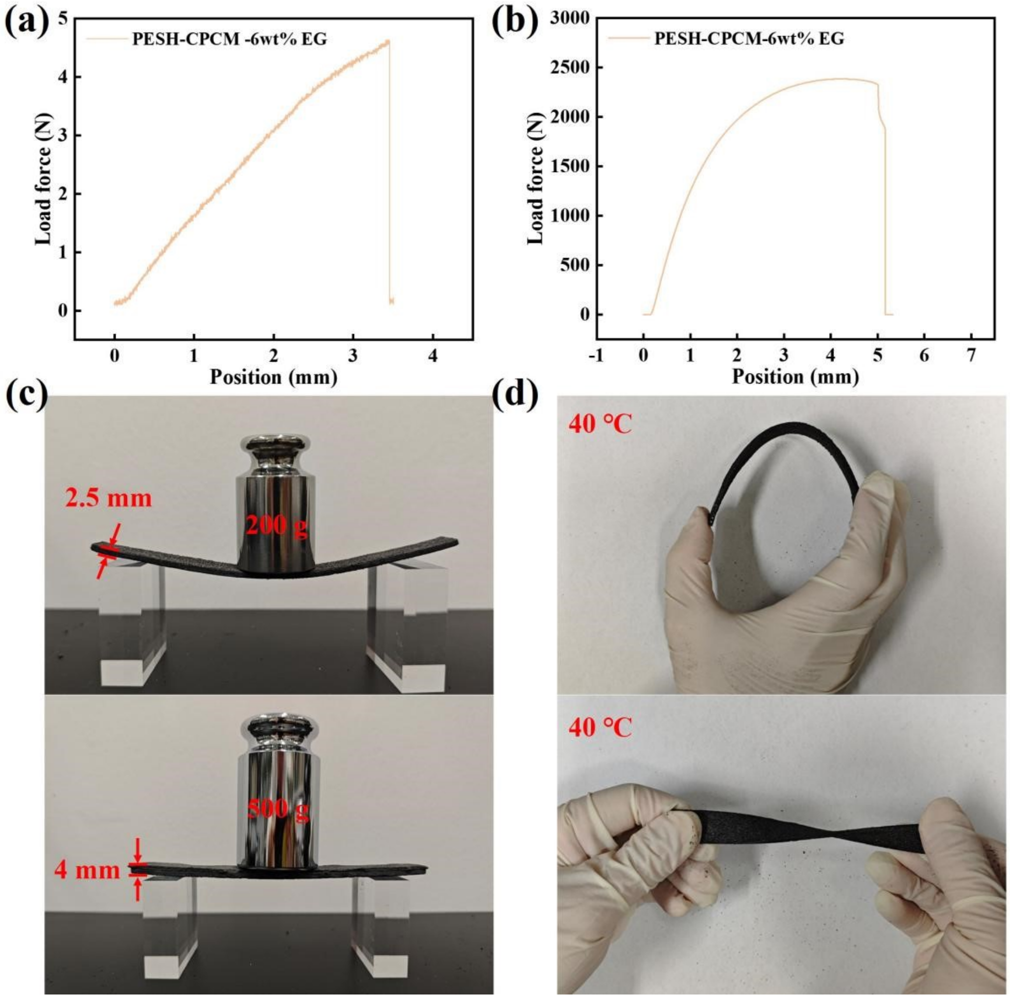

Figure S5. Bending (a) and compression (b) test curves of PESH-CPCM-zwt% EG (z=0, 2, 4, 6); (c) Load testing of PESH-CPCM-6wt% EG; (d) Flexibility testing of PESH-CPCM-6wt% EG after heating at 40 °C.

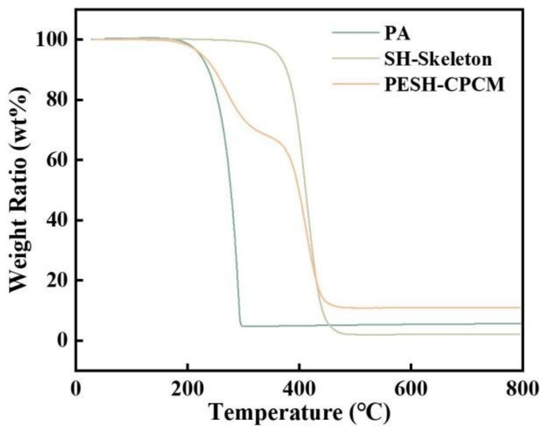

Figure S6. TG curves of PA, SH-skeleton, PESH-CPCM

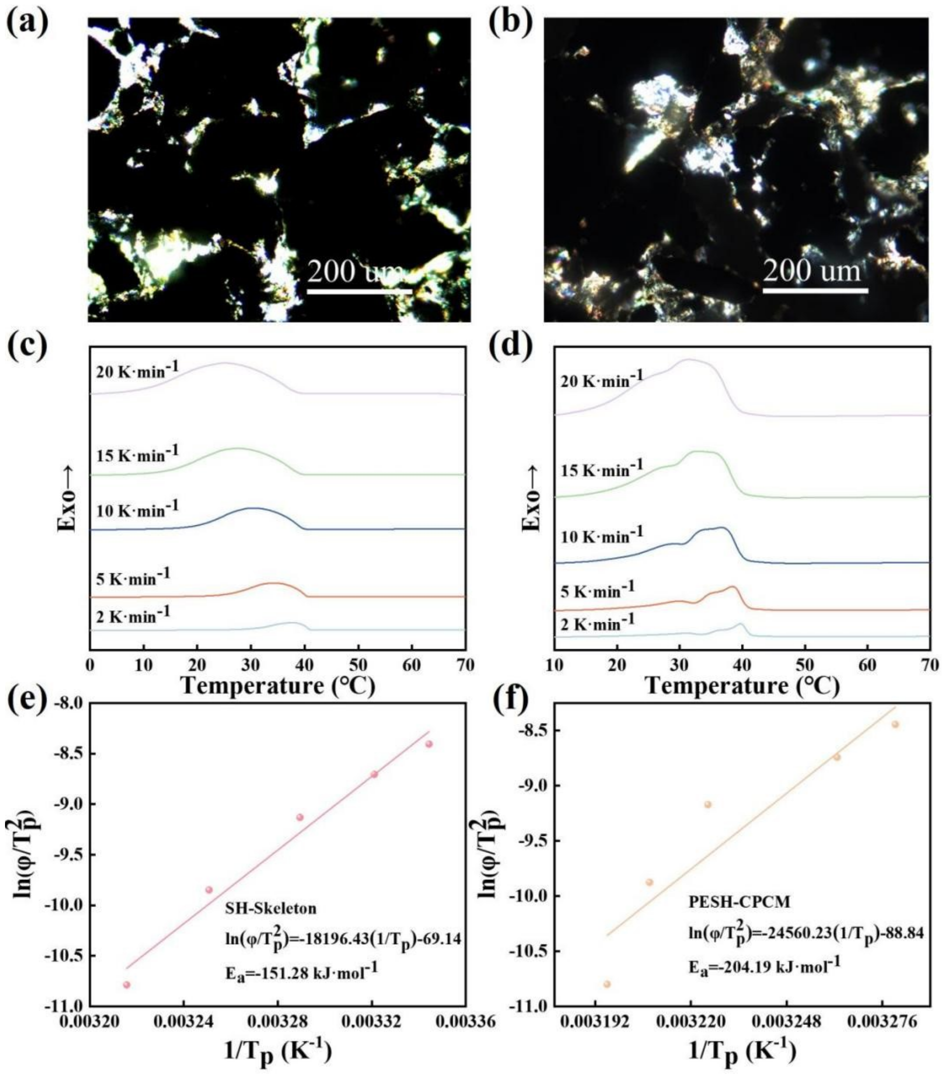

Figure S7. POM images of SH-skeleton (a) and PESH-CPCM (b); non-isothermal DSC curves of SH-skeleton (c) and PESH-CPCM (d); nucleation activation energy diagrams of SH-skeleton (e) and PESH-CPCM (f).

Table S3. Various parameters used to calculate E_a.

	φ ($K \cdot min^{-1}$)	T_P (K)	1/ T_P (K^{-1})	ln(φ/T_p^2)	Ea ($kJ \cdot mol^{-1}$)
SH-skeleton	-2	310.97	0.003215744	-10.787	
	-5	307.62	0.003250764	-9.848	
	-10	304.00	0.003289474	-9.131	151.28
	-15	301.10	0.003321156	-8.706	
	-20	299.01	0.003344369	-8.405	
PESH-CPCM	-2	312.94	0.003195501	-10.799	
	-5	311.73	0.003207904	-9.875	204.19
	-10	310.08	0.003224974	-9.171	
	-15	306.49	0.003262749	-8.742	

	-20	304.90	0.003279764	-8.444

Table S4. The price of raw materials with industrial grade purity.

Material	Price (USD • kg^{-1})
Stearyl acrylate (SA)	~4.22
Hexamethylene diacrylate (HA)	~3.34
Paraffin (PA)	~2.12
N, N-dimethyl-p-toluene (DMPT)	~10.26
Benzoyl peroxide (BPO)	~2.52
Expanded graphite (EG)	~45.82

Figure S8. Oil bath pot (a), mechanical stirrer (b) and incubator (c) for laboratory; mechanical stirrer (d) and incubator (e) for industry.