Supporting Information

Highly Flexible, strong and dynamic recovery solid-solid phase change materials based on polyurea-polyurethane for thermal management

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Experimental and Characterization Apparatus

Fourier transform infrared spectroscopy (FT-IR) model number Nicolet 6700 is used to characterize the characteristic chemical bonds of PTUs. PTU After dissolving the PTU material with ethanol, a small amount was applied to the KBr sheet, and the test was carried out after the rapid volatilization of ethanol.

X-ray diffraction (XRD) patterns were obtained by a Japan Rigaku D max X-ray diffractometer equipped with graphite monochromatized high-intensity Cu Kα radiation (λ= 1.54178 Å).

Small angle X-ray Scattering (SAXS) Small-angle X-Ray scattering (SAXS) was carried on a Cu Kα X-ray radiation source (Xeuss 2.0, Xenocs). Exposure time to X-ray was 10 min for all the samples, the thickness of the samples was about 0.5-0.8mm.

The universal testing machine model Instron 1158 is used to test the mechanical properties of elastomers. At room temperature, the polyurea was cut into dumbbell-shaped splines that meet ASTM D412 at a stretching speed of 100 mm/min, and each sample was tested five times and the results were averaged.

The thermogravimetric analyzer (TGA) model Q5000 (TA Corporation, USA) is used to test the thermal stability of samples. The test was carried out in a nitrogen atmosphere with a heating rate of 20 °C/min and a temperature increase from 25 °C to 800 °C.

The differential scanning calorimeter (DSC) model is TA Q2000 DSC (TA company, USA), which is used to analyze the glass transition temperature of polyurea, etc. The tests were carried out under nitrogen at a heating rate of 10 °C/min and cycled from 0 °C to 80 °C.

The scanning electron microscope (SEM) model JSM-6700F (Japan Electronics Corporation) is used to characterize the topography of materials. The acceleration voltage is 20 KV during the test.

Thermocouples are used to record temperature changes in materials in real time.

The thermal imaging camera, the model FLIR A655sc (FLIR Inc., USA), is used to take thermal imaging of the surface of materials to record and determine the temperature distribution of the sample surface.

Recycling PCMs via degradation-re-crosslinking(Route 1): PCMs with the same composition were cut into pieces, placed in DMF solution at 80°C and stirred until dissolved, and transferred to a mold to dry to regain the film.

Recycling PCMs via hot-pressing and reprocessing(Route 2): PCMs with the same constituent were cut into pieces, and then placed in a mold with desired shapes. Subsequent hot-pressing was conducted at 130 °C for 5 mins under a pressure of 10 MPa. After cooling down to ambient temperature, a recycled material was obtained.

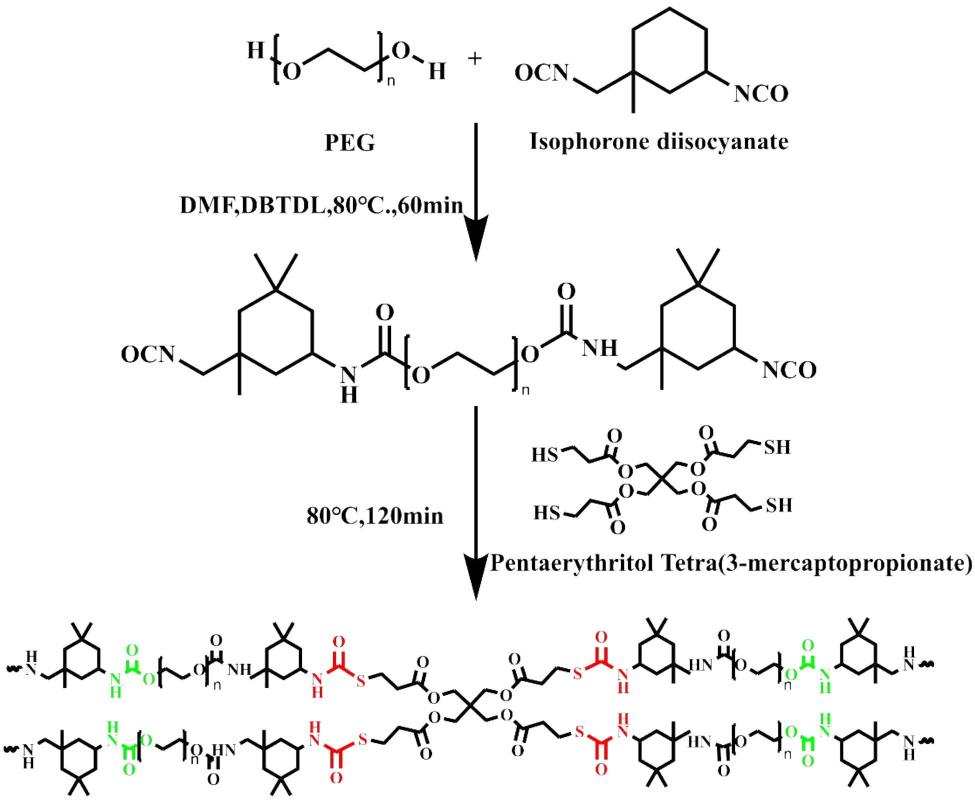


Figure S1. Synthesis of PTUs.

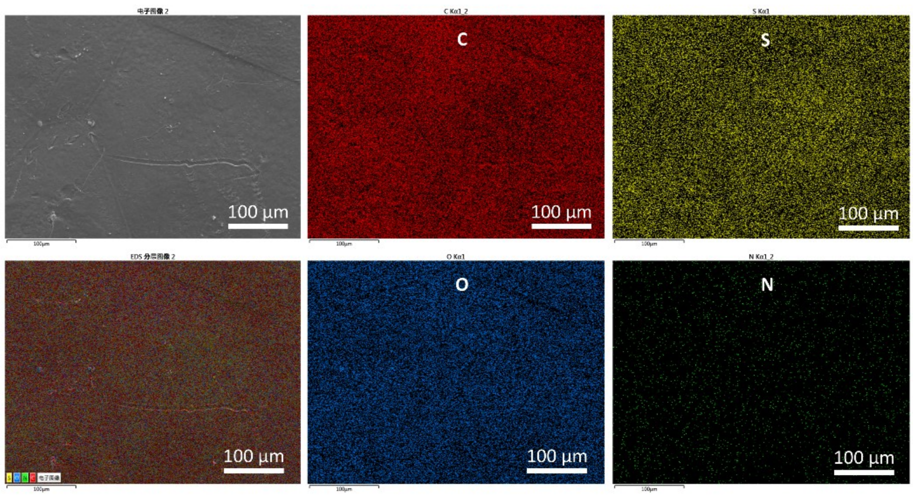
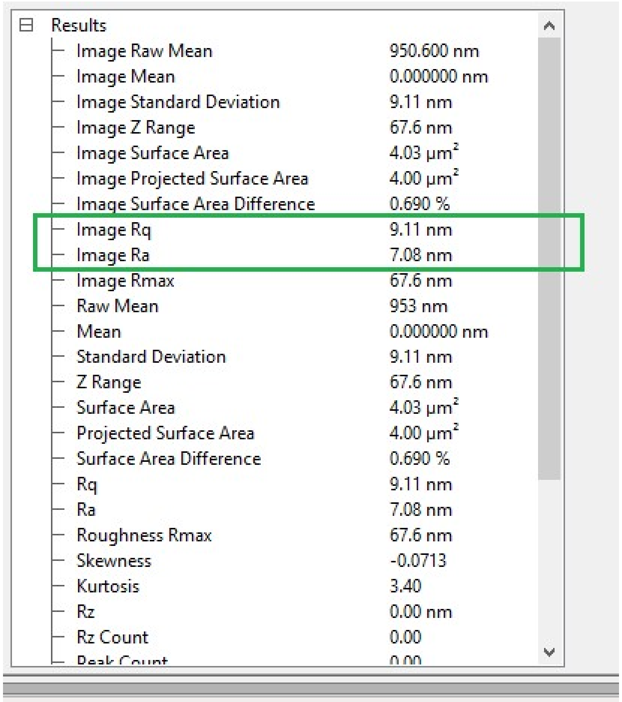


Figure S2. SEM and EDS of the PTUs.



Results	
Image Raw Mean	950.600 nm
Image Mean	0.000000 nm
Image Standard Deviation	9.11 nm
Image Z Range	67.6 nm
Image Surface Area	4.03 μm²
Image Projected Surface Area	4.00 μm²
Image Surface Area Difference	0.690 %
Image Rq	9.11 nm
Image Ra	7.08 nm
Image Rmax	67.6 nm
Raw Mean	953 nm
Mean	0.000000 nm
Standard Deviation	9.11 nm
Z Range	67.6 nm
Surface Area	4.03 μm²
Projected Surface Area	4.00 μm²
Surface Area Difference	0.690 %
Rq	9.11 nm
Ra	7.08 nm
Roughness Rmax	67.6 nm
Skewness	-0.0713
Kurtosis	3.40
Rz	0.00 nm
Rz Count	0.00
Peak Count	0.00

Figure S3. PTU surface roughness data diagram. (Rq is the root mean square roughness, Ra is the arithmetic mean roughness)

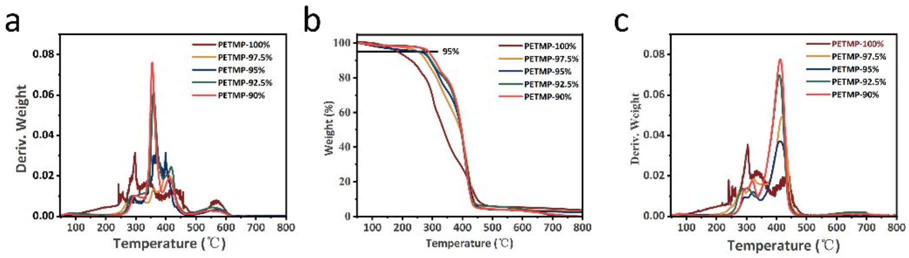


Figure S4. a) DTG(Air), b) TGA(N_2), c) DTG(N_2) of PTUs.

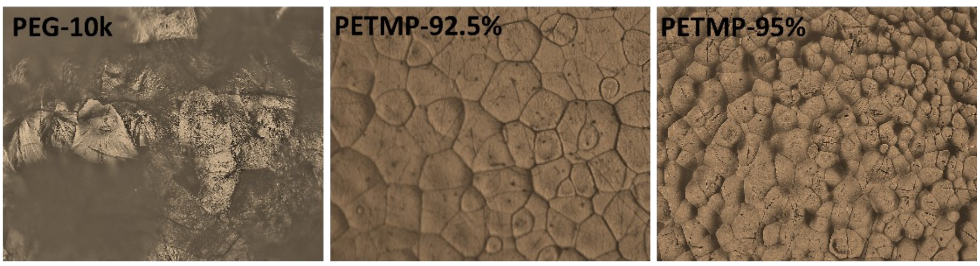


Figure S5. POM of PTUs.

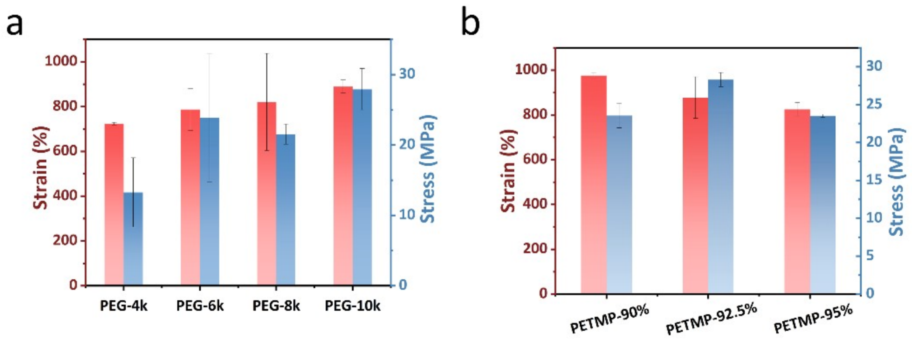


Figure S6. Mechanical histogram of PTUs.

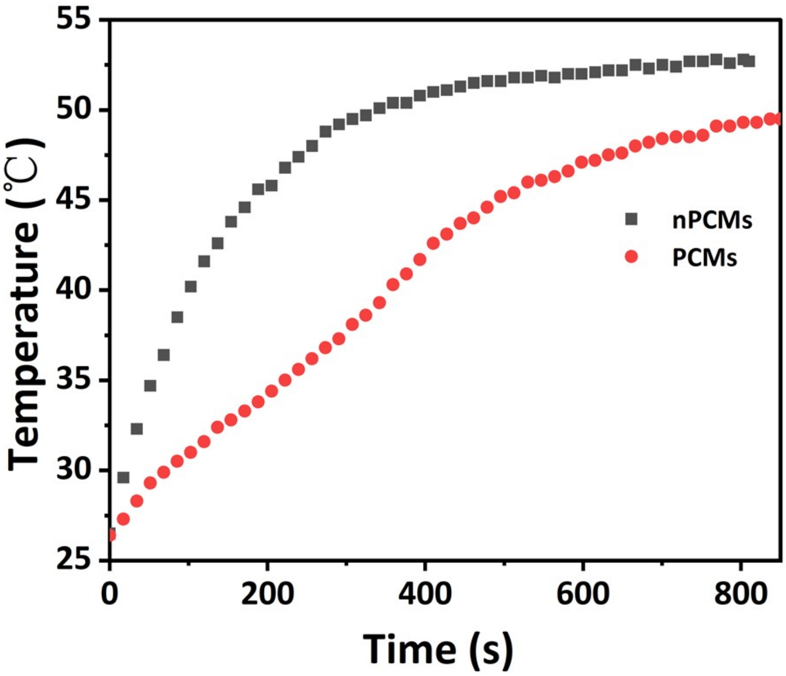


Figure S7. Thermal effect of PCM under forced heating on a hot stage.

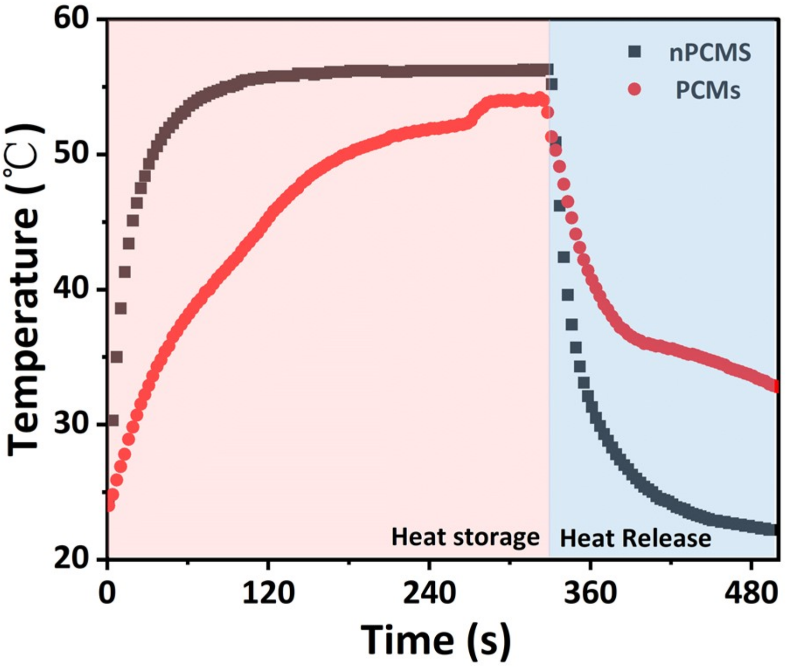


Figure S8. Temperature curve of PCM under forced heating of hot stage.

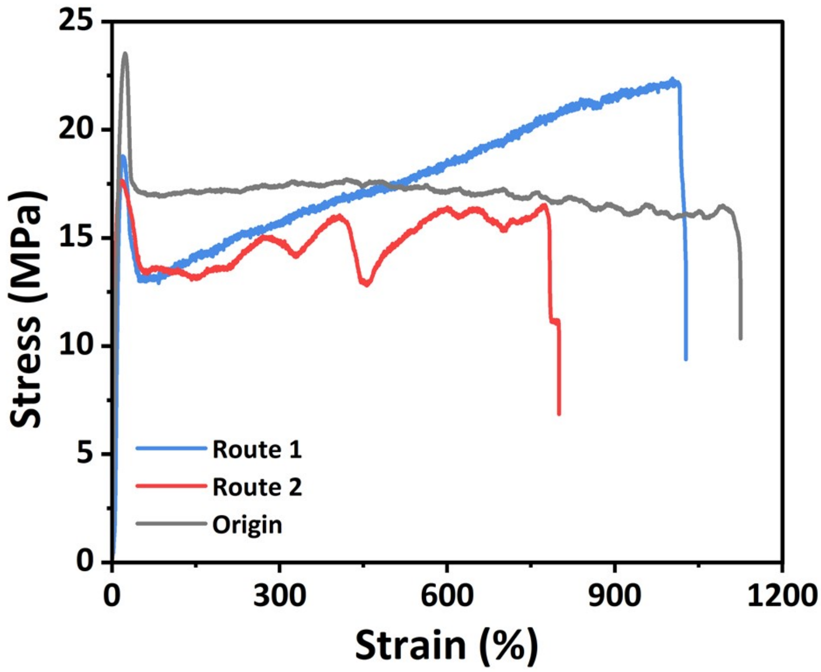


Figure S9. Mechanical tests before and after recovery in different ways.

Table S1 Formulation of the PTUs with different amounts of PEG-10k

Samples	IPDI (g)	PEG-10k (g)	PETMP (g)
PETMP-100%	3.668	0	3.665
PETMP-97.5%	3.668	3.750	3.570
PETMP-95%	3.668	7.500	3.482
PETMP-92.5%	3.668	11.250	3.390
PETMP-90%	3.668	15.000	3.298

Table S2 Formulation of the PTUs with different molecular weights of PEG

Samples	IPDI (g)	PEG-10k (g)	PETMP (g)
PEG-4k	3.668	4.500	3.665
PEG-6k	3.668	6.750	3.570
PEG-8k	3.668	9.000	3.482
PEG-10k	3.668	11.250	3.390

Table S3 Initial degradation temperature of PTUs

Samples	$T_{5\%}$(Air, ℃)	$T_{5\%}$(N_2, ℃)
PETMP-100%	178.5	184.3
PETMP-97.5%	248.0	251.6
PETMP-95%	248.4	256.0
PETMP-92.5%	286.4	275.4
PETMP-90%	282.6	287.5

Table S4 Mechanical properties of PTUs

Samples	Strain (%)	Stress (MPa)
PEG-4k	416.8±154.4	23.0±0.2
PEG-6k	751.0±287.0	25.0±3.0
PEG-8k	676.9±45.0	26.1±6.9
PEG-10k	877.3±92.7	28.3±0.9
PETMP-90%	976.0±13.3	23.6±1.6
PETMP-92.5%	877.3±92.7	28.3±0.9
PETMP-95%	825.9±30.1	23.5±0.2

Table S5 Activation energy of PTUs

Samples	Ea(kJ/mol)
PETMP-90%	127.5±11.8

PETMP-92.5%	154.6±2.0
PETMP-95%	158.6±11.9

Table S6 Enthalpy change of PTUs

Samples	△Hm		△Hc		Ts (℃)	λ (%)
	T (℃)	△H (J/g)	T (℃)	△H (J/g)		
PETMP-95%	46.1	45.9	15.2	39.9	30.9	49.7
PETMP-92.5%	51.5	68.5	36.3	49.3	15.2	53.2
PETMP-90%	51.2	77.0	26.6	57.0	24.7	63.0
PEG-10k	61.4	179.6	37.2	165.2	24.2	-

Note：Ts is the supercooling temperature, Ts = Tm-Tc, λ is latent heat efficiency, $\lambda=(\triangle H_{m-PTU})/(\triangle H_{m-PEG}\times\omega)\times 100\%$, where $H_{m\text{-}PTU}$ and $H_{m\text{-}PEG}$ are the melting latent heat of synthesized PSU and the used pristine PEG, respectively.

Table S7 The data of the PTU mechanics test was recovered

Samples	Strain (%)	Stress (MPa)	Recovery efficiency (calculated by Stress)
Origin	976.0±13.3	23.6±1.6	—
Route 1	906.7±57.1	22.2±2.1	94%
Route 2	785.2±42.0	19.0±1.3	80%

Table S8 Enthalpy change of chemical-recycle PSU and PEG

Samples	△Hm		△Hc		Ts (℃)
	T (℃)	△H (J/g)	T (℃)	△H (J/g)	
PETMP-95%	46.9	51.7	15.8	37.2	31.1
PETMP-92.5%	51.2	71.6	29.8	46.1	21.5
PETMP-90%	51.9	76.2	22.8	58.4	29.2
PEG-10k	61.4	179.6	37.2	165.2	24.2

Table S9 Enthalpy change of hot-pressed recycle PSU and PEG

Samples	△Hm		△Hc		Ts (℃)
	T (℃)	△H (J/g)	T (℃)	△H (J/g)	
PETMP-95%	39.2	36.9	21.8	10.4	17.5
PETMP-92.5%	45.4	44.3	29.7	46.1	15.7
PETMP-90%	45.2	51.8	24.2	56.2	21.0
PEG-10k	61.4	179.6	37.2	165.2	24.2