Supplementary Materials

Bioinspired roll-to-roll solar-thermal energy harvesting within form-

stable flexible composite phase change materials

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Note S1 Simulation of charging and discharging processes

A COMSOL model was built to analyze the heat transfer during charging and discharging processes. The roll-to-roll charging process is approximated as heating the composite PCM with a moving heating source. The heating power density is assumed to be 3.76 kW m⁻², which is obtained by multiplying the incident solar power density (4 kW m⁻²) by the solar absorption coefficient of the composite PCM (0.94). The velocity of the moving heating source is increasing with time and is expressed as

$$v = \omega \cdot (r + n \cdot d) \tag{S1}$$

where ω is the angular velocity of motor (2.5 r/min), r is the radius of shaft (8 mm), d is the thickness of the PCM composite (3 mm), and n is the number of rolled layers.

The heat diffusion of the system can be described by

$$\rho c_p \frac{\partial T}{\partial t} + \rho \Delta H_m \frac{1}{\partial t} = \nabla \cdot (k \nabla T) - Q$$
(S2)

where ρ is the density of storage materials (0.8 g cm⁻³), $^{C_{p}}$ is the heat capacity of the PCM composites (2.4 J g⁻¹ K⁻¹), k is the thermal conductivity of the composites (0.55 W m⁻¹ K⁻¹), and $^{\Delta H_{m}}$ is the phase change enthalpy of the storage materials (160 J g⁻¹). Q is the heat loss from the system, which includes convection heat loss with a convection coefficient of 5 W m⁻² K⁻¹ and radiation heat loss with an emissivity of 0.9.

The natural cooling discharging process was simulated in a similar way. At the beginning time, the charged composites were set to have uniform temperature distribution ($T_0 = 56$ oC). Both the extended and rolled composites were placed in air with the surrounding temperature $T_{\infty} = 20$ oC. The heat releasing process was considered to be dominated by the free convection and radiation heat transfer. The convection coefficient was assumed to be 5 W m⁻² K⁻¹, and the emissivity of composite

PCMs was assumed to be 0.9 when calculating the radiation heat releasing. As the discharging proceeds, the temperature of the composite PCMs will decrease. Compared to the extended sample, the rolled state has significantly reduced surface area, thus lowering the convection and radiation heat dissipation. By solving Equation (S2), the evolution of temperature T with discharging time t can be obtained for every points of the sample under both extended and rolled state.



Figure S1. Deposition of GO onto PU sponge and characterization of PU-rGO Sponge. (a) Schematic surface modification of PU sponge with GO by polydopamine (PDA) chemistry and subsequent reduction. (b) FTIR spectra of PU, PU-PDA, PU-PDA-GO, PU-PDA-rGO sponge. (c) TGA curves of PU-GO and PU-rGO sponge. (d) Raman spectra of PU-GO and PU-rGO sponge.



Figure S2. Preparation of large-sized PU-rGO sponge. (a) Photograph of a commercial yellow-colored PU sponge with a diameter of 10 cm. (b) Photograph of black-colored PU-rGO sponge.



Figure S3. SEM images of PU and PU-PDA sponge. (a) SEM image of pristine PU sponge. (b) SEM image of PU-PDA sponge. The inset SEM images at a high magnification show the smooth surface.



Figure S4. Wettability of PU and PU-rGO sponge. (a) Photographs showing a water droplet and a lubricating oil droplet on the surface of PU sponge. (b) Photographs showing a water droplet and a lubricating oil droplet on the surface of PU-rGO sponge.



Figure S5. DSC curves of PU-rGO-PCM composites before and after cycling tests. (a) PU-rGO-paraffin composites. (b) PU-rGO-stearic acid composites.



Figure S6. Static form-stability of stearic acid composites. Photographs showing stearic acid, PU-stearic acid, PU-rGO-stearic acid before and after melting. The dotted line shows the leaked liquid stearic acid from the composite sample.



Figure S7. Discharging temperature profiles of PU-PCM and PU-rGO-PCM composites. (a) Discharging temperature profiles of PU-paraffin and PU-rGO-paraffin composites. (b) Discharging temperature profiles of PU-stearic acid and PU-rGO-stearic acid composites.



Figure S8. Twistable and foldable PU-rGO-paraffin composites. The left shows the photograph and the infrared image of the charged twisted composite (82 wt%). The right shows the photograph and the corresponding infrared image of the charged Chinese tie-shaped composite.



Figure S9. Durability test of PU-rGO-paraffin composites. (a) Comparative photographs showing the appearance of PU-rGO-paraffin composite before and after rolling for 4000 cycles. (b, c) A cross-sectional SEM image of PU-rGO-paraffin composite before the rolling test and an SEM image at a high magnification showing the decoration of rGO sheets on the inner surface of PU sponge. (d, e) A cross-sectional SEM image of PU-rGO-paraffin composite and an SEM image at a high magnification showing the robust attachment of rGO sheets after the rolling test.

Sample	$T_{\rm m}$ (°C)	$\Delta H_{\rm m} ({\rm J~g^{-1}})$	$T_{\rm s}$ (°C)	$\Delta H_{\rm s} ({\rm J g^{-1}})$
Paraffin	52.4	168.3	59.6	170.6
PU-paraffin (96.4 wt%)	53	158.1	59.2	159.3
PU-rGO-paraffin (95.2 wt%)	53.7	159.3	59.2	160.5
Stearic acid	67.7	254.5	65.2	254.1
PU-stearic acid (97.9 wt%)	67.6	220.3	65	218.7
PU-rGO-stearic acid (96.7 wt%)	68.1	223.4	65.9	227.5

Table S1 Thermophysical properties of neat PCM and composite PCMs