Supporting Information

Effective solar-driven interfacial water evaporation-assisted adsorption of organic pollutants by a activated porous carbon material

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1. Experimental Section

Materials. Sodium chloride, glucose monohydrate, potassium hydroxide, sodium hydroxide, Hydrochloric acid, rhodamine B (RhB), methyl orange (MO) and methyl blue (MB) were purchased from Aladdin Reagent Co., Ltd. (Shanghai, China), and Polyvinyl alcohol (PVA) sponge was purchased from Zhejiang Yuechang Co., Ltd. All aqueous solutions in the experiment were prepared by deionized water.

Water evaporation test. The Polyvinyl alcohol sponge (PS) was cut into cylinder with 3 cm diameter and 2 mm thickness. Then, the PC@PS and activated-PC@PS samples were prepared by the following steps. 10 mg PC or activated-PC sample was ultrasonic dispersed in an appropriate amount of anhydrous ethanol, and the resultant slurry was uniformly dropped on the PS and dried. The obtained samples were named as PC-A1 (A2, A3, A4, A5)@PS. As a comparison, 10 mg PC samples were loaded on the filter membrane (0.22 μm) with 3 cm diameter to evaluate the evaporation performance of the PC material. These samples were surrounded by thermal insulating polystyrene foam to prevent heat loss. The samples were irradiated by a solar simulator (CEL-HXUV300, CEAULIGHT) with 1, 2 and 3 sun irradiation for two hours. The mass change of the device was tested by a precision balance (EX223ZH, OHAUS) in the latter 60 min, which was equivalent to the mass change of water evaporation. The surface and water temperatures were measured by a dual-channel thermocouple.

Adsorption experiments. At first, the SDIWE device was placed on a small

magnetic stirrer, and the precision balance was below the magnetic stirrer. In order to achieve SDIWE-assisted heat adsorption, the PC-A4 sample was irradiated by a solar simulator with 0, 1 and 2 sun irradiations to make the water temperatures maintain at ~25°C (298 K), ~31°C (304 K) and ~36°C (309 K). Subsequently, 3 mg of the PC and activated PC adsorbents were added into 30 mL water solution containing different concentrations of rhodamine B (20-300 mg L^{-1}), methyl orange (20 and 50 mg L^{-1}) and methylene blue (20 and 50 mg L^{-1}). Meanwhile, the dark adsorption dye molecules occur at the stirring rate of 400 r/min, and the concentrations of RhB (MO, MB) were measured by UV-vis spectrometer at the characteristic absorption wavelength.

The adsorption behaviors of activated PC and kinetics of PC-A4 over time was conducted by using 20 and 50 mg L^{-1} dye solution at different conduction temperatures. The adsorption capacity at time t (Q_t, mg g^{-1}) was defined by the following equation [S1]:

$$Q_t = \frac{(C_0 - C_t)V}{m} \tag{S1}$$

where C_0 (mg L^{-1}) represents the initial dye concentration, C_t (mg L^{-1}) is the residual dye concentration at time t, V (mL) is the volume of the dye solution, m (g) is the adsorbent mass.

Adsorption isotherm experiments were performed by adsorbing the different initial concentrations of RhB solution (50-500 mg L^{-1}) for 12 h to achieve adsorption equilibrium at 298, 304 and 309 K, respectively. The lost water calculated on the basis of the evaporation rate was added in the nether water body every three hours.

The equilibrium adsorption amount Q_e (mg g^{-1}) was expressed by the equation [S2]:

$$Q_e = \frac{(C_0 - C_e)V}{m}$$ (S2)

Where C_e (mg L^{-1}) refers to the RhB concentration at equilibrium.

Effect of initial pH. 3 mg PC-A4 sample was added into the 30 mL RhB solution (300 mg L^{-1}), initial pH ranged from 2 to12. The adsorption experiment was conducted at the conduction temperature of 309 K. The equilibrium adsorption amount at different pH was measured by UV-vis spectrometer.

Recycling test. 3 mg PC-A4 sample was added into the 30 mL RhB solution (300 mg L^{-1}). After saturated adsorption, the adsorbent was separated from the solution by centrifugation at a rate of 12000 r/min and then treated by sonication in absolute ethyl alcohol for 1h. Then, the adsorbent was washed several times and dried for the next adsorption experiment. Finally, the centrifuged liquid was determined by UV-vis spectrometer.

2. Calculation of the energy efficiency (η) for solar-steam conversion

The energy efficiency (*η*) for solar-steam conversion can be described by the following equation:

$$\eta = \frac{\dot{m}H_e}{q_0 C_{opt}}$$ (S3)

where $\dot{m} = \frac{dm}{Adt}$ is water evaporation rate (kg h^{-1} m^{-2}, *dm* is the water mass change, *A* is the surface area of photothermal material，*t* is the evaporation time), C_{opt}, q_0, H_e are the optical concentration, the normal solar irradiation and equivalent evaporation enthalpy.

3. Calculation of heat loss [S3]

Radiation heat loss (η_{rad}). The heat radiation can be described by Stefan-Boltzmann Equation:

$$J_{rad} = \varepsilon\sigma\,(T_{pc}^4 - T_{env}^4) \tag{S4}$$

where ε and σ represent the emissivity (0.94) and Stefan-Boltzmann constant (5.67 × 10^{-8} W m^{-2} K^{-4}). T_{pc} is the steady-state temperature of the absorber surface (41.3 °C) and T_{env} is the air temperature above the absorber (38.7 °C) under 1 sun irradiation. In this respect, the radiation heat flux is calculated to be 17 W m^{-2}, which is ~1.7% of the input energy of 1 sun flux.

Conduction heat loss (η_{cond}). The conduction heat energy loss η_{cond} can be determined by the following equation:

$$J_{cond} = Q_{cond}/At = Cm\Delta T/At \tag{S5}$$

where C is the specific heat capacity of water (4.2 kJ kg^{-1} °C^{-1}), Q_{cond} and t are the heat energy conducted to bulk water and conductive time (1 h), respectively. The absorber area (A) and the water weight (m) are 7.065 × 10^{-4} m^2 and 30 g, respectively. ΔT (6.1 °C) represents the recorded temperature of water before and after stable solar steam generation. Therefore, the conductive heat loss is calculated as ~302.2 W m^{-2} under 1 kW m^{-2} solar irradiation, which is 30.22 % of the input energy of 1 sun flux.

Convection heat loss (η_{conv}). The convective heat loss is calculated by the following equation:

$$J_{conv} = h\,(T_{pc} - T_{env}) \tag{S6}$$

where h is the convection heat transfer coefficient (5 W m^{-2} K^{-1}). Thus, the convection heat loss η_{conv} is calculated to be 1.3 % under 1 sun irradiation.

4. conductive heat loss (η_{cond}), the energy efficiency (η) for solar-steam conversion and evaporation rate ($\dot{m}$) with different masses of water body (1 sun irradiation)

(1) m=30 g, ΔT =6.1 °C

Under 1 kW m^{-2} solar irradiation, the conductive heat loss is calculated to be ~302.2 W m^{-2}, which is 30.22 % of the input energy of 1 sun flux.

$\dot{m}$=1.42 kg m^{-2} h^{-1} $\qquad$ η=66.3 %

(2) m=20 g, ΔT =6.7 °C

Under 1 kW m^{-2} solar irradiation, the conductive heat loss is calculated to be ~221.2 W m^{-2}, which is 22.12 % of the input energy of 1 sun flux.

$\dot{m}$=1.59 kg m^{-2} h^{-1} $\qquad$ η=74.3 %

(3) m=10 g, ΔT =7.4 °C

Under 1 kW m^{-2} solar irradiation, the conductive heat loss is calculated to be ~122.1 W m^{-2}, which is 12.21 % of the input energy of 1 sun flux.

$\dot{m}$=1.78 kg m^{-2} h^{-1} $\qquad$ η=83.2 %

5. Adsorption kinetics

The nonlinear formulas are defined as the following equation:

$$ln(Q_e - Q_t) = lnQ_e - k_1 t \tag{S7}$$

$$\frac{t}{Q_t} = \frac{1}{k_2 Q_e^2} + \frac{t}{Q_e} \tag{S8}$$

where k_1 and k_2 are the rate constant for pseudo-first-order and pseudo-second-order models, respectively.

6. Langmuir and Freundlich models fitting

The Langmuir and Freundlich models were selected to fit the adsorption data of the PC-A4 sample, which are given by the following formula:

$$Q_e = \frac{Q_m K_L C_e}{1 + K_L C_e} \tag{S9}$$

$$Q_e = K_L C_e^{1/n} \tag{S10}$$

where Q_m (mg g^{-1}) is the maximum adsorption capacity, C_e (mg L^{-1}) is the solution concentration at equilibrium, K_L (L mg g^{-1}) and K_F (mg g^{-1}(L mg^{-1})$^{1/n}$) are the constants of the Langmuir and Freundlich models, respectively.

7. **thermodynamic analysis**

The thermodynamic parameters including Gibbs free energy of adsorption (ΔG), enthalpy change (ΔH), and standard entropy change (ΔS) were calculated at different conduction temperatures by the following equations:

$$\ln\left(\frac{Q_e}{C_e}\right) = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \tag{S11}$$

$$\Delta G = \Delta G - T\Delta S \tag{S12}$$

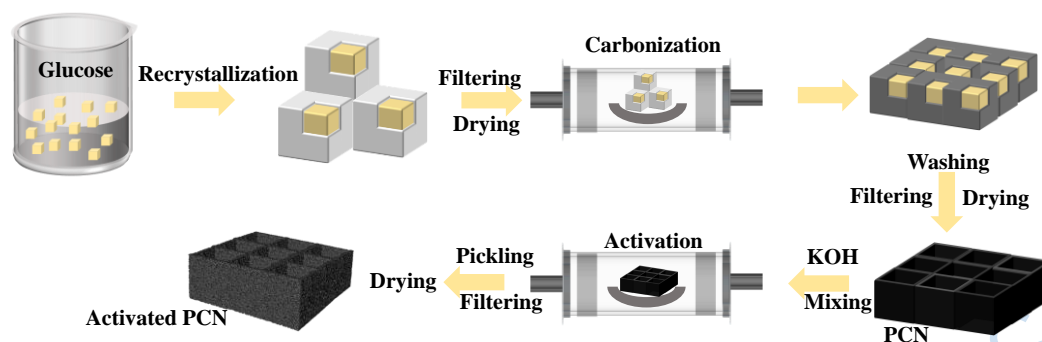where R is the gas constant (8.3145 J/mol·K) and T is the solution temperature (K).

Fig. S1 Schematic illustration of the preparation of PC and activated PC.
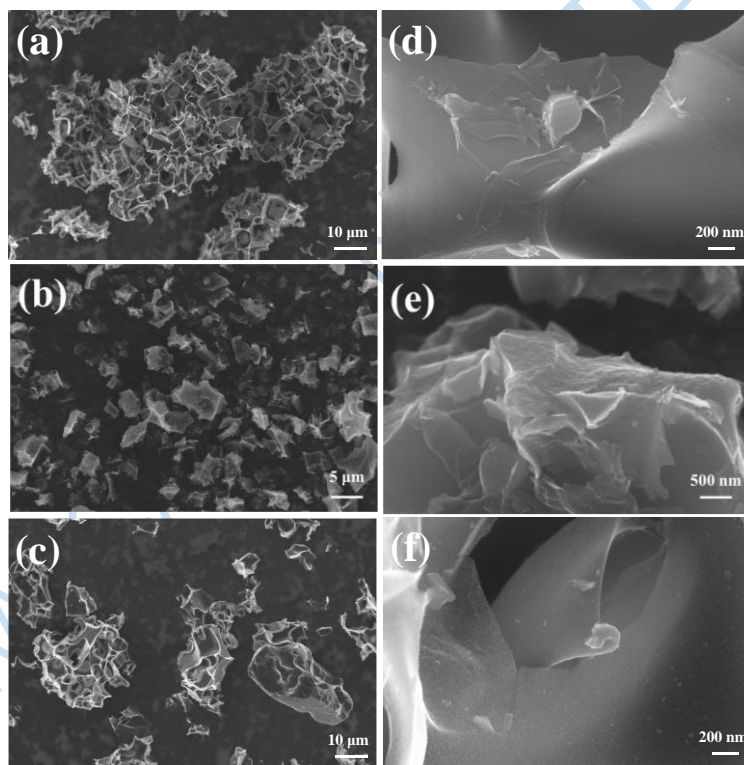

Fig. S2 (a, d) SEM image of PC-A1 sample at different magnifications; (b, e) SEM image of PC-A5 sample at different magnifications; (c, f) SEM image of PC-A3 sample at different magnifications.
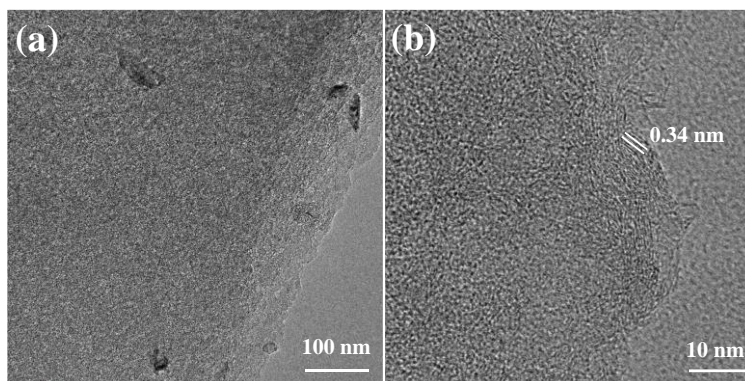
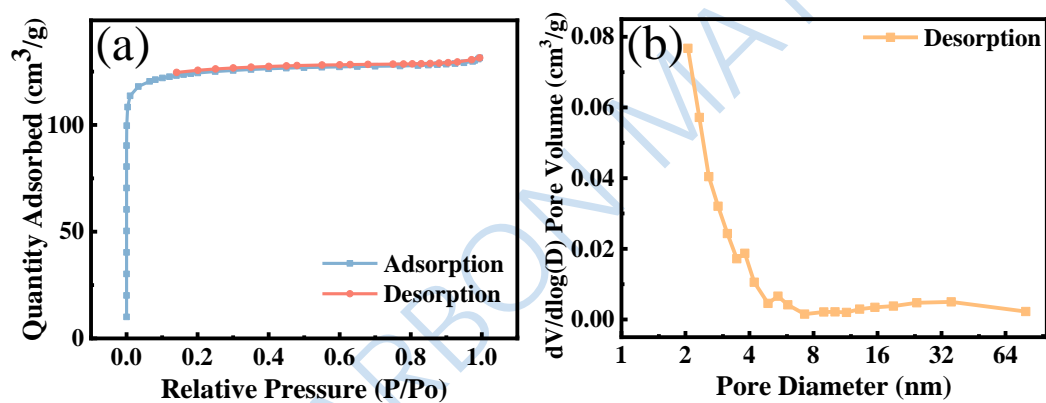Fig. S3 (a-b) TEM image of PC-A4 sample at different magnifications

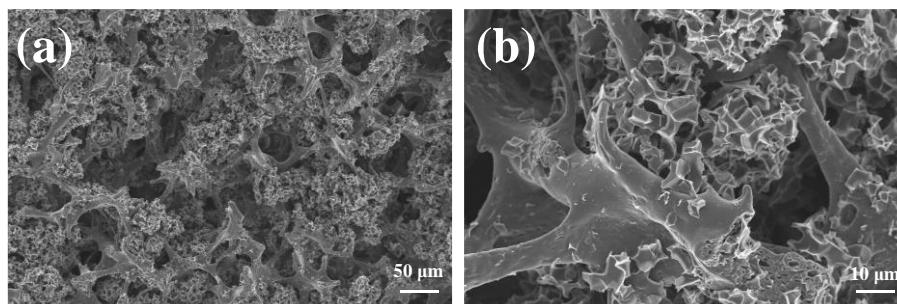Fig. S4 (a) N_2 adsorption-desorption isotherm of PC;(b) Pore size distribution of PC.

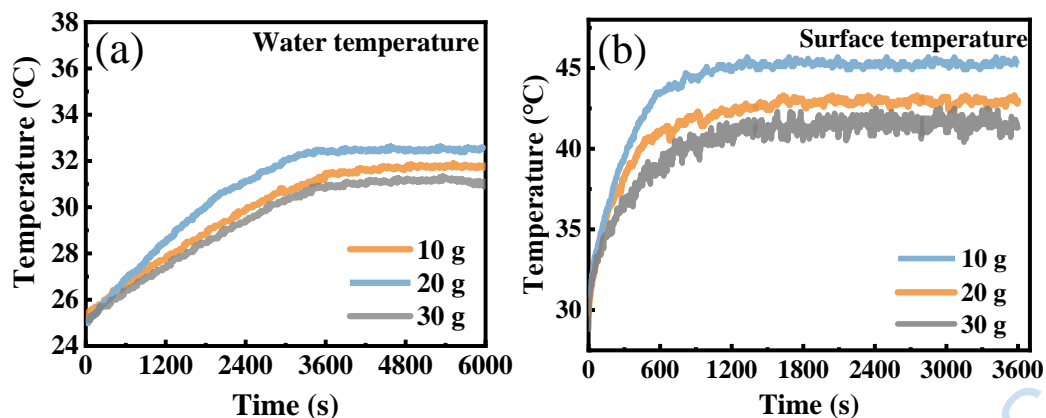Fig. S5 (a-b) SEM image of PC-A4@PS sample at different magnifications

Fig. S6 (a-b) The surface temperature changes of wetted PC-A4@PS with different masses of water body (1 sun irradiation); (d) Temperature change curves of different masses of bottom water (1 sun irradiation).
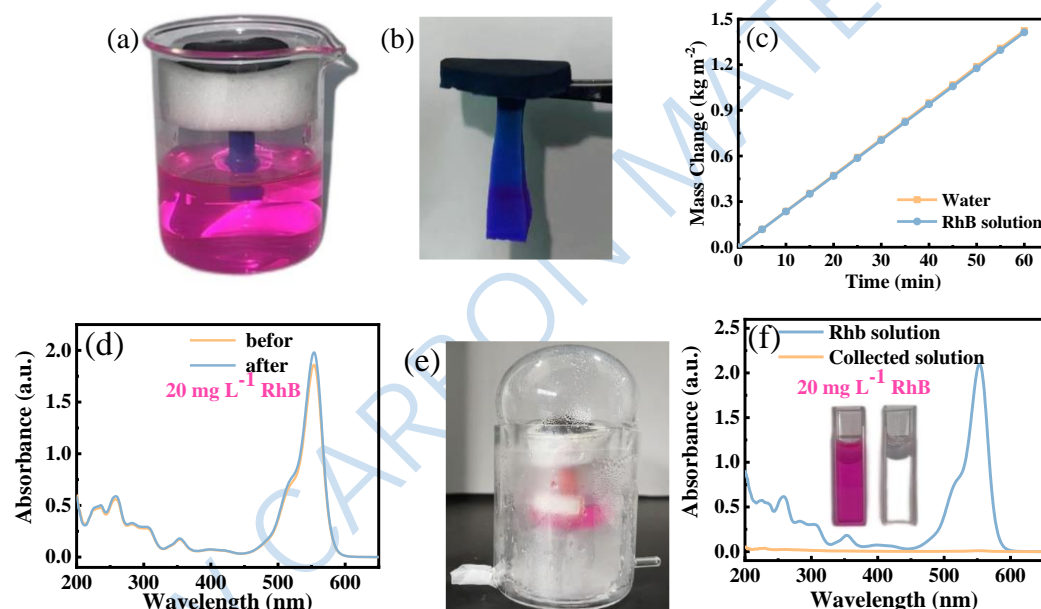

Fig. S7 (a-b) The colors of RhB solution and PVA sponge after photothermal water evaporation by using PC-A4@PS sample; (c) Water mass change as a function of time for different water supply sources under 1 sun irradiation; (d) The absorbance of RhB solution before and after photothermal water evaporation; (e) The collection device of the evaporative water; (f) The absorbance of the RhB solution and the collected solution.

Incipiently, the aqueous solution in SDIWE device (Fig. 2a-2b) was replaced by 30 mL RhB solution (20 mg L^{-1}) to test the photothermal ability. As shown in Fig. S7(a-b), the color of RhB solution barely changes after water evaporation, and only the soaking area of the PVA sponge appears magenta. Meanwhile, the evaporation rate remains unchanged, while the concentration of RhB solution slightly increases due to the water evaporation (Fig. S7(c-d)). The absorption of the collected water has no characteristic absorption peak, which is consistent with the water (Fig. S7(e-f)).
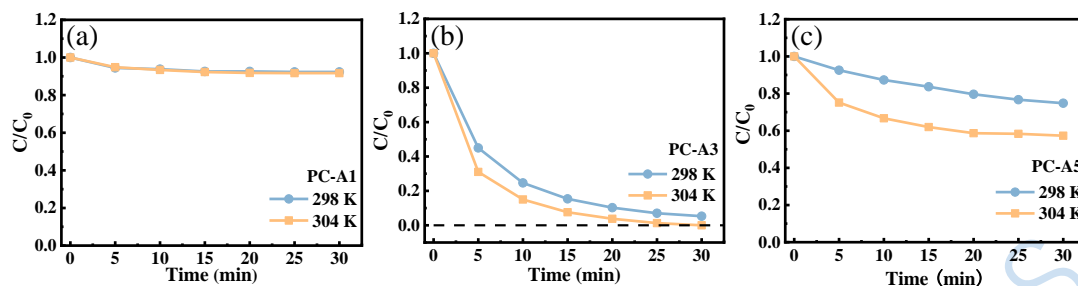
Fig. S8 Adsorption behaviors of (a) PC-A1, (b) PC-A3 and (c) PC-A5 samples to RhB (20 mg L^{-1}) at different conduction temperatures

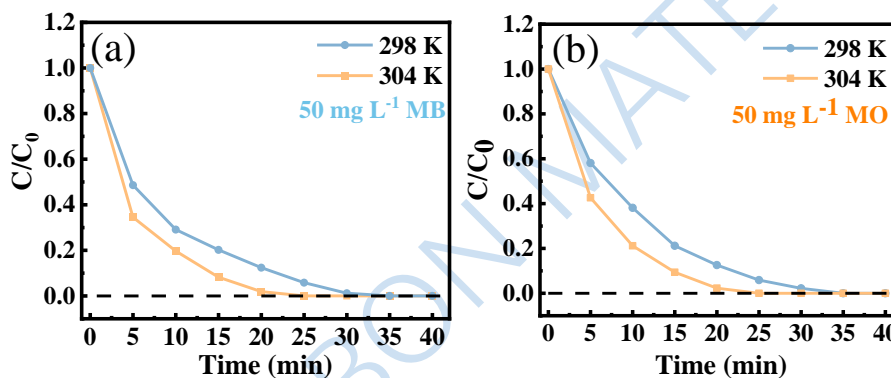Fig. S9 Adsorption behaviors of PC-A4 sample to (a) MO (50 mg L^{-1}), (b) MB (50 mg L^{-1}) at different conduction temperatures.
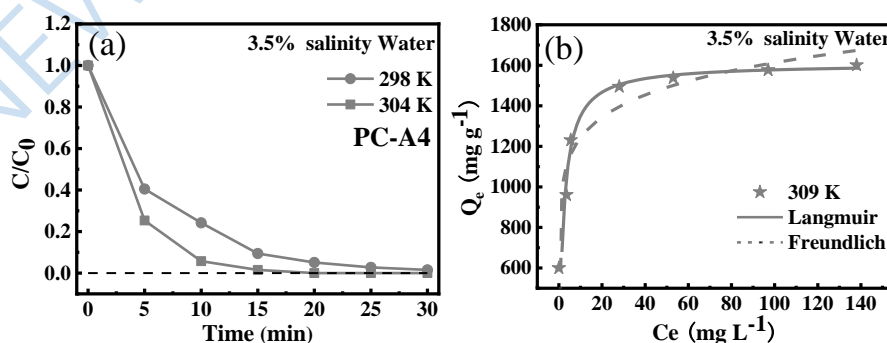

Fig. S10 (a) Adsorption behaviors of PC-A4 sample to RhB (20 mg L^{-1}) at 3.5% salinity water and different conduction temperatures; (b) Adsorption isotherms of PC-A4 to RhB (3.5% salinity water) and the corresponding nonlinear fitting by Langmuir and Freundlich models.

Table S1 Data for N_2 adsorption-desorption isotherms.

Samples	$S_{BET}(m^2\ g^{-1})$	$V_{total}\ (cm^3\ g^{-1})$	$V_{micro}\ (cm^3\ g^{-1})$
PC	491.36	0.21	0.17
PC-A4	1867.71	1.04	0.28

Table S2 Data for calculated equivalent enthalpy values.

Samples	Calculated equivalent enthalpy values	DSC test evaporation enthalpy	Evaporation rates under dark (kg m^{-2} h^{-1})
Water	2450	2440	0.09
PC	2183	2215	0.101
PC-A4	2080	2086	0.106
PC@PS	1807.3	1815	0.122
PC-A4@PS	1683	1674	0.131

Table S3 Fitting parameters of adsorption Kinetic models for RhB at different temperatures

Sample	Kinetic models		Constants		
			298 K	304 K	309 K
PC-A4	Pseudo-first-order	$Q_{e,cal}$ (mg g^{-1})	472	482	558
		R^2	0.79	0.77	0.78
		k_1 (min^{-1})	0.058	0.078	0.091
	Pseudo-second-order	$Q_{e,cal}$ (mg g^{-1})	507	509	510
		R^2	0.98	0.98	0.98
		k_2 (10^{-4} g mg^{-1} min^{-1})	1.93	2.81	3.43

Table S4 Adsorption isotherm parameters of Langmuir and Freundlich models.

Sample	Isotherm types		Constants		
			298 K	304 K	309 K
PC-A4	Langmuir	Q_m (mg g^{-1})	1435	1528	1607
		K_L (L mg g^{-1})	0.372	0.396	0.520
		R^2	0.99	0.99	0.99
	Freundlich	K_F (mg g^{-1}(L mg^{-1})1/n)	848	881	956
		1/n	0.109	0.117	0.113
		R^2	0.21	0.36	0.46

Table S5 Thermodynamic parameters for the adsorption of RhB onto PC-A4 sample.

Sample	Temperature	ΔG (kJ/mol)	ΔH (kJ/mol)	ΔS (J/mol K^{-1})
PC-A4	298 K	-5.414	16.882	74.826
	304 K	-5.865		
	309 K	-6.239		

References

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[S3] M. Q. Yang, C.F. Tan, W. Lu, et al., Adv. Funct. Mater. 30 (2020) 2004460.