

Hygroscopic Wood Sponge with Dual Phase Change Function for Enhanced All-Weather Atmospheric Water Harvesting

Corresponding Author: Professor Ruzhu Wang

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

Reviewer #2

(Remarks to the Author)

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The manuscript can be accepted now for publication.

Reviewer #3

(Remarks to the Author)

The authors conducted lots of supplementary experiments and made a comprehensive revision to the manuscript. Two small questions need to be clarified before publication.

1.How about the repetitiveness of the water uptake tests. The error bars in Fig. R10, Fig. R18 and Fig. R20 should be included.

2.PEG as the PCM might leak during desorption process as raised by the previous reviewer, the TOC in Figure R18 seems a significant increase in the collected water. The reviewer suggest a simple test with filter paper, to check the state of the composite when heating.

Version 2:

Reviewer comments:

Reviewer #3

(Remarks to the Author)

The authors have appropriately addressed the comments. Therefore, the manuscript is suitable for publication.

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Reviewer #1

The manuscript presents a novel dual-functional hygroscopic wood sponge (PHW) for all-weather atmospheric water harvesting (AWH), integrating LiCl-based sorption with a photothermal PCM hydrogel for energy management. The work is comprehensive, combining material synthesis, structural characterization, simulation, and outdoor validation. The topic is relevant for sustainable water technologies. However, certain experimental, analytical, and presentation aspects need to be addressed before acceptance as a part of Major revision.

Response:

We sincerely thank you for the constructive feedback and valuable suggestions. We greatly appreciate your careful evaluation of our work. In response, we have carefully addressed each of your points and made the necessary revisions to enhance the clarity and quality of the manuscript. We trust that these changes will meet your expectations and contribute to the overall improvement of the paper.

1. While the integration of wood sponge and PCM hydrogel is interesting, the manuscript should better differentiate its novelty from prior works that have already used wood-based sorbents or PCM-assisted AWH systems (e.g., references [22], [25], [38], [39]).

Response:

We sincerely thank you for your valuable comments. We have reviewed the references you mentioned and conducted further research on related literature. In the revised manuscript, we have provided a clearer and more rigorous comparison between our work and prior studies on wood-based sorbents, PCM-assisted AWH systems, and even PCM-assisted interfacial evaporation systems. We have compared the core innovations of our work with those of these studies, as shown in [Table R1](#).

Table R1. Comparison between our work and prior wood-based sorbents and PCM-assisted AWH systems.

Category	Prior work	Main Features	Limitations
Wood-based sorbents	Zhang et al. ¹ WLG-15	Machine learning used for performance prediction.	Lacks PCM-based thermal management at the material level, preventing stable and fast water release under weak or lack of solar radiation desorption without solar radiation.
	Liang et al. ² DWM-PM	Thermoresponsive hydrogels for low-enthalpy water release.	
	He et al. ³ AWPH	Split atmospheric water to produce hydrogen.	
PCM-assisted AWH systems	Agrawal et al. ⁴ PCM-integrated AWH system combined with a double-ended vacuum tube solar air heater	Store excess heat with PCM during the day to enable desorption at night, even without solar radiation.	Focus on PCM integration for thermal management improvements at the system level, rather than targeted energy management design for specific sorbent materials, often requiring large-scale equipment for implementation.
	Agrawal et al. ⁵ Integrating PCM with the solar air heater		
	Agrawal et al. ⁶ PCM-encapsulated both-sides open evacuated tube solar air heater		
	Agrawal et al. ⁷ PCM-integrated atmospheric water extraction		
Our work	Ji et al. PCM-integrated hygroscopic wood	Sorbent material design that integrates fast sorption kinetics with energy management for all-weather water production. It incorporates a multi-cycle operation design, verified through long-term outdoor experiments, along with heat and mass transfer simulations, and assessments of global water production potential and economic viability.	N/A

Regarding wood-based sorbents:

In further literature research, several key studies on wood-based sorbents were identified. Zhang et al. developed LiCl-modified wood sorbents and used interpretable machine learning models to predict moisture sorption and desorption performance. However, the sorbent lacks effective thermal management capabilities, making it unable to address the energy limitations for desorption caused by fluctuations in environmental conditions. Liang et al. designed multi-scale composite materials, combining delignified wood channels, MOF-801 microporous regions, and PNIPAM-MXene photothermal interfaces, which allow solar-driven desorption. However, this design relies entirely on solar radiation and lacks thermal energy storage, preventing continuous operation during the night or under non-sunny conditions. He et al. developed a hybrid water collection system that integrates photothermal evaporation and photocatalytic hydrogen generation, combining carbonized wood and ZnCl_2 . While the system shows great potential for hydrogen production, it lacks thermal energy storage, making it unable to operate continuously under nighttime or cloudy conditions, thus limiting its application in continuous water harvesting and hydrogen production for AWH.

Regarding PCM-assisted AWH systems:

Anshu Agrawal et al. conducted four studies on PCM-assisted AWH systems, with a focus on integrating PCM with solar air heaters to enhance water collection efficiency. In their research, they designed a PCM-integrated system combined with a double-ended vacuum tube solar air heater, enabling the system to operate at night by storing solar energy during the day. The integration of PCM provides thermal energy storage, allowing the system to operate efficiently even in the absence of sunlight, significantly extending its operating time. Their innovations were primarily centered around system design and PCM integration, rather than advancements in hygroscopic materials. By optimizing thermal and mass transfer, they improved water production efficiency. Despite significant progress in system integration, no notable innovations were made in the development of hygroscopic materials.

In summary, we propose an enhanced water-harvesting approach that couples solar energy with PCM latent heat to drive water release at the materials level. We use a wood sponge featuring vertically aligned multilayered channels as the matrix. The lower layer is loaded with lithium chloride and functions as the sorption zone. Its vertical and multilayered ordered structure reduces the vapor transport resistance and accelerates the sorption kinetics. The upper layer is impregnated with a photothermal PCM hydrogel, which serves as the energy management zone. When sunlight is strong, it can store solar energy; even under weak or no sunlight, it can release this stored energy to support desorption. This study addresses the challenges faced by solar-driven adsorbents under day-night alternation and fluctuating solar radiation, making all-weather atmospheric water collection possible. Furthermore, our research goes beyond material design, including multi-cycle operation, heat and mass transfer simulations, large-scale outdoor real-world testing, as well as global water production potential assessment and

economic analyses. This provides a system-level framework rarely explored in previous studies.

- [1] Zhang, X. *et al.* Development and characterization of novel wood-based composite materials for solar-powered atmospheric water harvesting: a machine intelligence supported approach. *J. Clean Prod.* **497**, 145061 (2025).
- [2] Liang, J. *et al.* Synergistic structural alignment and interfacial switching enable fast and low-energy water release in solar-driven atmospheric water harvesting. *Adv. Funct. Mater.*, e18960 (2025).
- [3] He, L. *et al.* A hybrid photocatalytic system splits atmospheric water to produce hydrogen. *Adv. Funct. Mater.* **34**, 2313058 (2025).
- [4] Agrawal., A. *et al.* Sorption based solar atmospheric water harvesting integrated with a direct evaporative cooler: an experimental approach. *J. Water Process Eng.* **76**, 108237 (2025).
- [5] Agrawal., A. *et al.* Experimental comparison and 6E analyses of double-ended evacuated tube collector based atmospheric water harvesting with and without PCM. *Sol. Energy Mater. Sol. Cells* **282**, 113343 (2025).
- [6] Agrawal., A. *et al.* Harvesting freshwater from atmospheric air using thermal energy storage enabled solar air heater. *J. Energy Storage.* **73**, 109008 (2023).
- [7] Agrawal., A. *et al.* Atmospheric water extraction based on solar air heater encapsulated with phase change material: An experimental investigation. *Sol. Energy Mater. Sol. Cells* **269**, 112764 (2024).

Changes made:

On page 11 of the revised manuscript, we have added a description distinguishing our work from previous wood-based sorbents and PCM-assisted AWH systems, with the relevant sections highlighted in red. The following is mentioned:

“However, existing wood-based sorbents often lack effective thermal management, particularly during the desorption phase, resulting in reduced water release efficiency.^{45,47,48} Traditional PCM-assisted AWH systems typically utilize PCM at the system level rather than within the sorbent material itself.⁴⁹⁻⁵² In contrast, our design integrates a PCM-based photothermal hydrogel into the upper layer of the sorbent matrix, functioning as an energy management zone that stores latent heat (Fig. 4a). This integration facilitates continuous desorption, independent of complex external systems, even under low solar irradiance or in the absence of sunlight.”

2. A quantitative comparison (in Table form) summarizing uptake capacity, sorption rate, desorption efficiency, and stability versus recent literature (2023–2025) would make the novelty clearer. Further, the incorporation of the recent literature (<https://doi.org/10.1016/j.jwpe.2025.108237>, <https://doi.org/10.1016/j.solmat.2024.113343>) would additionally benefit the manuscript.

Response:

We sincerely thank you for your insightful comments. In response, we have reviewed recent literature (2023–2025) and added a comparison table ([Table R2](#)) summarizing uptake capacity, sorption rate, desorption efficiency, and stability. We have also incorporated the suggested references and included additional studies on PCM-assisted AWH systems to strengthen the literature comparison and further highlight the novelty of our work.

Table R2. Comparison of different kinds of hygroscopic sorbents (from 2023 to 2025) with their water sorption capacity, water sorption rate, desorption efficiency, stability, outdoor water production and operational duration.

Sorbent	RH (%)	Sorption capacity (g g ⁻¹)	Sorption rate (g g ⁻¹ h ⁻¹) (Within 2 h)	Desorption ratios (%)	System-level energy efficiency (%)	Stability (Number of cycles)	Outdoor water production (L _{water} kg _{sorbent} ⁻¹ day ⁻¹)	Outdoor operational stability (Day)
CNF/LiCl-1.5 ¹	75	1.48	0.1873	80.3% (1 Sun, within 120min)	N/A	10	0.63	1
TCP-Li35 ²	60	1.93	0.3727	95.7% (1.0 Sun, within 36 min)	N/A	15	1.55	14
Pollen-LiCl@PNIPAM-b-PPy	80	1.52	0.657	82% (1.0 Sun, within 30 min)	N/A	10	N/A	N/A
PPy-COF@Trilayer-LiCl ⁴	60	1.37	0.6919	95% (1.0 Sun, within 10 min)	N/A	25	3.91 (kg _{water} m _{sorbent} ⁻² day ⁻¹)	1
PVF-PPy-MnC ⁵	70	0.86	0.3634	97.5% (1.0 Sun, within 30 min)	N/A	15	0.8799 (g _{water} m _{sorbent} ⁻² day ⁻¹)	1
HEMHs ⁶	70	2.36	1.1231	85% (1.0 Sun, within 120 min)	N/A	24	2.85	1
PCP@LiCl ⁷	60	1.48	0.7314	62.25% (1.0 Sun, within 60 min)	N/A	6	N/A	N/A
CAL gel ⁸	30	0.79	1.74	88.24% (70 °C, within 30 min)	N/A	10	8.96	1
MAWH ⁹	70	2.65	0.2054	92.23% (1.0 Sun, within 60 min)	N/A	20	N/A	N/A
SMPH aerogel ¹⁰	70	1.45	0.5118	N/A	N/A	10	0.8	7
WLG-15 ¹¹	70	1.6	0.4850	73.7%	N/A	10	1.43	1

					(1.0 Sun, within 600 min)				
MTC ¹²		60	0.45	0.225	81.28% (60 °C, within 10 min)	N/A	30	N/A	N/A
PCM-integrated AWH system ¹³	Slica gel	78.47	N/A	0.0229	N/A	14.05%	12	0.195	N/A
	Molecular sieve	72.95		0.026		8.48%	12	0.11	
AWH system with PCM-encapsulation ¹⁴	Slica gel	35.1	N/A	0.0223	N/A	9.17%	N/A	0.17	7
	Molecular sieve	30.8		0.0241	N/A	7.87%	N/A	0.134	7
Harvesting Fresh Water from Atmospheric Air (HFWA) ¹⁵	Slica gel	30.5	N/A	0.0288	N/A	10.29%	N/A	0.1875	4
	Molecular sieve	30.3		0.0314	N/A	10.29%		0.1705	4
Atmospheric Water Extraction with PCM ¹⁶	Slica gel (15kg)	35.9	N/A	0.0276	N/A	7.64%	N/A	0.1867	N/A
	Slica gel (20 kg)	33.3		0.0261	N/A	10.48%		0.205	
This work		80	2.48	0.7330			76.4% (Under 1.0 Sun for 60 min and in the dark for 60 min)	0.96-1.72	4

- [1] Zhu, P. et al. 3D printed cellulose nanofiber aerogel scaffold with hierarchical porous structures for fast solar-driven atmospheric water harvesting. *Adv. Mater.* **36**, 2306653 (2024).
- [2] Zou, H. et al. Solar-driven scalable hygroscopic gel for recycling water from passive plant transpiration and soil evaporation. *Nat. Water* **2**, 663–673 (2024).
- [3] Zheng, X. et al. Rape pollen-based composite sorbent with thermo-responsive and photothermal properties for atmospheric water harvesting. *Adv. Funct. Mater.* **34**, 2407127 (2024).
- [4] Xia, M. et al. Biomimetic hygroscopic fibrous membrane with hierarchically porous structure for rapid atmospheric water harvesting. *Adv. Funct. Mater.* **33**, 2214813 (2023).
- [5] Shan, H. et al. Integrating rooftop agriculture and atmospheric water harvesting for water–food production based on hygroscopic manganese complex. *Adv. Funct. Mater.* **34**, 2402839 (2024).
- [6] Sun, J. et al. Entangled mesh hydrogels with macroporous topologies via cryogelation for rapid atmospheric water harvesting. *Adv. Mater.* **36**, 2314175 (2024).
- [7] Wang, J. et al. Tillandsia-inspired ultra-efficient thermo-responsive hygroscopic nanofibers for solar-driven atmospheric water harvesting. *Adv. Mater.* **36**, 2408977 (2024).
- [8] Zhou, H. et al. Solar-driven drum-type atmospheric water harvester based on bio-based gels with fast adsorption/desorption kinetics. *Adv. Mater.* **36**, 2403876 (2024).
- [9] Cai, C. et al. Biomimetic dual absorption–adsorption networked MXene aerogel-pump for integrated water harvesting and power generation system. *ACS Nano* **18**, 4376–4387 (2024).
- [10] Wang, X. et al. High solar-thermal conversion aerogel for efficient atmospheric water harvesting. *Small* **20**, 2307416 (2023).
- [11] Zhang, X. et al. Development and characterization of novel wood-based composite materials for solar-powered atmospheric water harvesting: a machine intelligence supported approach. *J. Clean Prod.* **497**, 145061 (2025).
- [12] Luo, Q. et al. An atmospheric water-harvester with ultrahigh uptake–release efficiency at low humidity. *ACS Nano* **18**, 14650–14660 (2024).
- [13] Agrawal, A. et al. Sorption based solar atmospheric water harvesting integrated with a direct evaporative cooler: an experimental approach. *J. Water Process Eng.* **76**, 108237 (2025).
- [14] Agrawal, A. et al. Experimental comparison and 6E analyses of double-ended evacuated tube collector based atmospheric water harvesting with and without PCM. *Sol. Energy Mater. Sol. Cells* **282**, 113343 (2025).
- [15] Agrawal, A. et al. Harvesting freshwater from atmospheric air using thermal energy storage enabled solar air heater. *J. Energy Storage*. **73**, 109008 (2023).
- [16] Agrawal, A. et al. Atmospheric water extraction based on solar air heater encapsulated with phase change material: An experimental investigation. *Sol. Energy Mater. Sol. Cells* **269**, 112764 (2024).

Changes made:

We have added the **Supplementary Table 5 in the revised Supplementary Information, pages 10-11**, summarizing water sorption capacity, water sorption rate, desorption

efficiency, stability, outdoor water production and operational duration. These results are compared with the recent literature from 2023 to 2025. Additionally, we have incorporated the latest four studies by Anshu Agrawal on PCM-assisted AWH systems into the table, further clarifying the novelty of our research.

3. The study emphasizes latent heat contribution from PCM but lacks a detailed energy balance quantifying solar-to-thermal conversion efficiency, stored latent heat utilization rate, and effective desorption enthalpy.

Response:

We sincerely appreciate the valuable comments from you. Indeed, clarifying the **solar-to-thermal conversion efficiency, effective desorption enthalpy, and stored latent heat utilization rate**, is crucial for understanding the performance of our sorbent during photothermal desorption, especially in evaluating the photothermal conversion efficiency and the utilization of stored latent heat for water desorption under dark conditions.

Solar-to-thermal conversion efficiency

The photothermal conversion efficiency (η) refers to the proportion of received light energy converted into heat energy, which is an important parameter for assessing the photothermal performance of the material. It is calculated using the following formula:

$$\eta = \frac{M\Delta H_m}{PS\Delta t}$$

where M (g) is the mass of the energy management zone of the PHW, ΔH_m (J g⁻¹) is the melting enthalpy of the energy management zone of the PHW, P (W m⁻²) is the solar irradiation intensity, S (m²) represents the area of the top surface of the samples, and Δt (s) is the duration of the phase change under the simulated radiation, the relevant data of this are shown in [Table R3](#).

Table R3. Data for photothermal conversion calculations.

M (g)	ΔH_m (J g⁻¹)	P (W m⁻²)	S (m²)	Δt (s)	η (100%)
2.68	155.51	1000	0.00051	900	90.80

By calculation, the photothermal conversion efficiency of the energy management zone of the PHW is 90.80 %.

Effective Desorption Enthalpy

To examine the water desorption behavior of our sorbent, we measured its heat flow signal using Differential Scanning Calorimetry (DSC) ([Figure R1](#)). The effective desorption enthalpy of the PHW sorbent was found to be 1540 J g⁻¹ by integrating the heat flow over time, representing the energy required for water to transition from the sorbed state to the desorbed state. Prior to this test, the sorbent was exposed to 90% RH for 12 h to reach

sorption equilibrium, achieving a water uptake of approximately 3.03 g g⁻¹. Additionally, we measured the evaporation enthalpy of bulk water, which was found to be 2269 J g⁻¹. The PHW sorbent exhibited a lower desorption enthalpy compared to bulk water, suggesting that water molecule desorption within the PHW is a lower-energy process. This can be attributed to the specific interactions between the sorbent matrix and water.¹⁻³ Subsequently, the measured value of the effective desorption enthalpy was used to calculate the latent heat utilization efficiency, which is a key parameter for evaluating the use of stored latent heat in the photothermal conversion process during desorption under dark conditions.

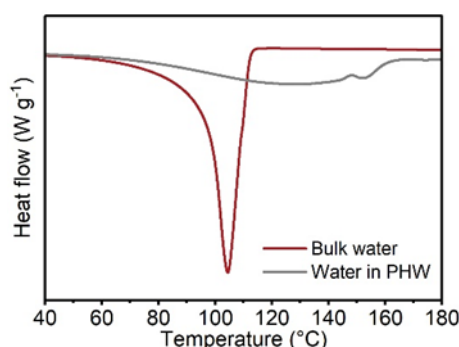


Figure R1. DSC curve of bulk water and water in the sorption zone of PHW.

Stored Latent Heat Utilization Rate

The stored latent heat utilization rate refers to the degree to which the stored latent heat is effectively utilized during desorption. It is calculated using the following formula:

$$\eta = \frac{\Delta H_v M_{\text{water}}}{\Delta H_m M}$$

where ΔH_v (J g⁻¹) is the desorption enthalpy of the water in the sorption zone of the PHW, M_{water} (g) refers to the total amount of water released after the solar simulator (1 kW m⁻²) is turned off (the mass of water desorbed during the process driven by the latent heat of phase change), ΔH_m (J g⁻¹) is the melting enthalpy of the energy management zone of the PHW, M (g) is the mass of the energy management zone of the PHW, the relevant data of this are shown in [Table R4](#).

Table R4. Data for stored latent heat utilization rate calculations.

ΔH_v (J g ⁻¹)	M_{water} (g)	ΔH_m (J g ⁻¹)	M (g)	η (%)
1540	0.2345	155.51	2.68	86.65

By calculation, the stored latent heat utilization rate is 86.65%.

[1] Lu, H. et al. Tailoring the Desorption behavior of hygroscopic gels for atmospheric water harvesting in arid climates. *Adv. Mater.* **34**, 2205344 (2022).

[2] Yang, B. et al. Low-Energy Atmospheric Water Harvesting and Heat Recovery System Based on Vertically Stratified Structural Composite. *Adv. Funct. Mater.*, e14160 (2025).

[3] Lin, B. et al. Biomass-Derived Hydro-Sponge for Ultra-Efficient Atmospheric Water Harvesting. *Adv. Funct. Mater.*, **35**, 2500679 (2025).

Changes made:

We have added a detailed quantitative energy balance analysis of solar-to-thermal conversion efficiency, effective desorption enthalpy, and stored latent heat utilization rate in **Supplementary Notes 4, 5 and 7 of the revised SI (pages 16–18)**.

On page 13 of the revised manuscript, we have added a description of the photothermal conversion efficiency, highlighted in red, as shown below:

“Notably, PHW exhibits a transient temperature plateau, indicating a rapid phase transition and latent-heat energy storage process of the PCM. The photothermal conversion efficiency of PHW reaches as high as 90.80%. (Supplementary Note 4)”

On page 13 of the revised manuscript, we have added a description of the effective desorption enthalpy and latent heat utilization efficiency, highlighted in red, as shown below:

“This improvement is enabled by a high latent-heat utilization efficiency of up to 86.65%, based on the PCM latent enthalpy and the enthalpy required for water desorption (Supplementary Notes 5 and 6)”

4. COMSOL simulations and kinetic analysis (Figs. 3d–e) are discussed qualitatively. Please include governing equations, boundary conditions, and validation with experimental sorption rate data to enhance credibility.

Response:

We would like to thank you for the constructive suggestion. In response to your comments, we have substantially expanded the description of the COMSOL model and the kinetic analysis. The revised manuscript now includes governing equations, detailed boundary conditions, and quantitative validation using experimental sorption kinetics.

Governing equations:

The diffusion process of water vapor across the transverse cross-section of the wood matrix with varying structures is modeled using Fick's law of diffusion:

$$\frac{\partial C}{\partial t} = D \nabla^2 C$$

Where C (kg m^{-3}) is the concentration of water in the sorbent, D ($\text{m}^2 \text{s}^{-1}$) is the diffusivity of vapor in the sorbent, t (s) is the time. We assume that the diffusivity D of the sorbent is constant during the sorption.

Boundary conditions:

The boundary conditions used in COMSOL Multiphysics are specified as follows:

$$C_{in} = C_{ext}$$

$$C_{out} = 0$$

Where C_{in} is the water vapor concentration at the inlet, C_{ext} is the external water vapor concentration, and C_{out} is the water vapor concentration at the outlet.

These boundary conditions are applied to both the ordered and disordered wood microstructures in the simulations, ensuring that the model accurately reflects the physical conditions during vapor transport across the transverse cross-section of the wood.

Kinetic analysis of sorption experiment data.

To quantitatively validate the COMSOL simulation results, we performed a kinetic analysis using the early-time solution of Fickian diffusion in a planar slab. The fitting equations for the ordered and disordered wood structures at 60% RH are as follows:

$$\frac{M_t}{M_e} \approx \frac{4}{\sqrt{\pi}} \left(\frac{D t}{L^2} \right)^{1/2}$$

Where M_t (g g^{-1}) is the water sorption, M_e (g g^{-1}) is the equilibrium water sorption, D ($\text{m}^2 \text{s}^{-1}$) is the effective diffusion coefficient, t (s) is the time during the sorption process. L (m) is the half length of the cross-sectional of the sorbent.

For the ordered structure at 60% RH:

$$M_t/M_e = 0.00679 t_s^{1/2} - 0.04270, R^2 = 0.9529$$

Using the slope $a = 0.00679$, the effective diffusion coefficient D is calculated as:

$$D = 9.05 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$$

For the disordered structure at 60% RH:

$$\frac{M_t}{M_e} = 0.00674 t_s^{1/2} - 0.03729, R^2 = 0.9628$$

Using the slope $a = 0.00674$, the effective diffusion coefficient D is calculated as:

$$D = 8.91 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$$

Simulation data comparison with experimental results:

We compared the simulation results with the experimental sorption kinetics. The simulation data presented in [Figure R2](#) show that the vapor transport rate in the ordered structure is significantly higher than that in the disordered structure, which is consistent with the experimental findings. In the simulation, the water vapor transport rate in the ordered structure ranges from 0 to $1.2 \mu\text{m s}^{-1}$, whereas it is only from 0 to $0.6 \mu\text{m s}^{-1}$ in the disordered structure. These values align with the effective diffusion coefficients obtained experimentally, with the ordered structure having an effective diffusion coefficient of $9.05 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$ which is greater than the $8.91 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$ observed in the disordered structure. The strong agreement between the simulation and experimental data confirms that the COMSOL model accurately predicts the vapor diffusion behavior in both types of

wood structures, reflecting the lower diffusion resistance and higher transport rate in the ordered structure.

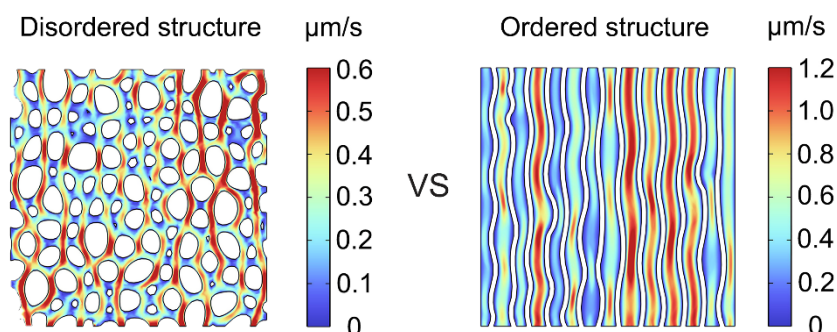


Figure R2. Predict vapor transport in the sorption zone of PHW with different matrix types (disordered structure and ordered structure) during the sorption process.

Changes made:

In Supplementary Note 2 of the revised Supplementary Information (Page 15), we have added a description of the governing equations and boundary conditions used in the COMSOL Multiphysics simulations.

In Supplementary Note 3 of the revised Supplementary Information (Pages 15-16), we have included the section on “Kinetic analysis of sorption rate and validation with COMSOL simulations”.

Additionally, **in the revised manuscript (Page 11)** we have included the following description, highlighted in red:

“We also validated the simulation results by comparing them with experimental sorption kinetics, further confirming the accuracy of the analysis (Supplementary Note 3).”

5. The manuscript mentions compressive strength variation between wet and dry states. However, cyclic fatigue and durability of the material over repeated sorption–desorption cycles need quantification.

Response:

We appreciate your insightful comment. Long-term mechanical stability is essential to ensure the sustained performance of the sorbent. To assess the durability of the sorbent under repeated sorption–desorption cycles, we conducted 50 sorption-desorption cycles, where each cycle consisted of 12 h of sorption at 90% RH followed by 2 h of desorption, which included 1 h under solar intensity of 1 kW m⁻² and then 1 h in darkness.

Specifically, we measured the compressive strength after the 1st, 25th, and 50th sorption cycles, as well as after the corresponding desorption cycles (Figure R3). The compressive strength after the first sorption cycle was 0.20 MPa, which decreased to 0.14 MPa after the 25th cycle and 0.13 MPa after the 50th cycle. The corresponding compressive

strengths after desorption were 3.95 MPa, 3.45 MPa, and 3.37 MPa, respectively. From the 1st to the 50th cycle, the compressive strength during sorption decreased by 35%, while the desorption compressive strength only decreased by 14%. These results demonstrate that the sorbent maintains stable mechanical properties over prolonged cycling, with minimal degradation, thus ensuring its suitability for long-term use in water harvesting.

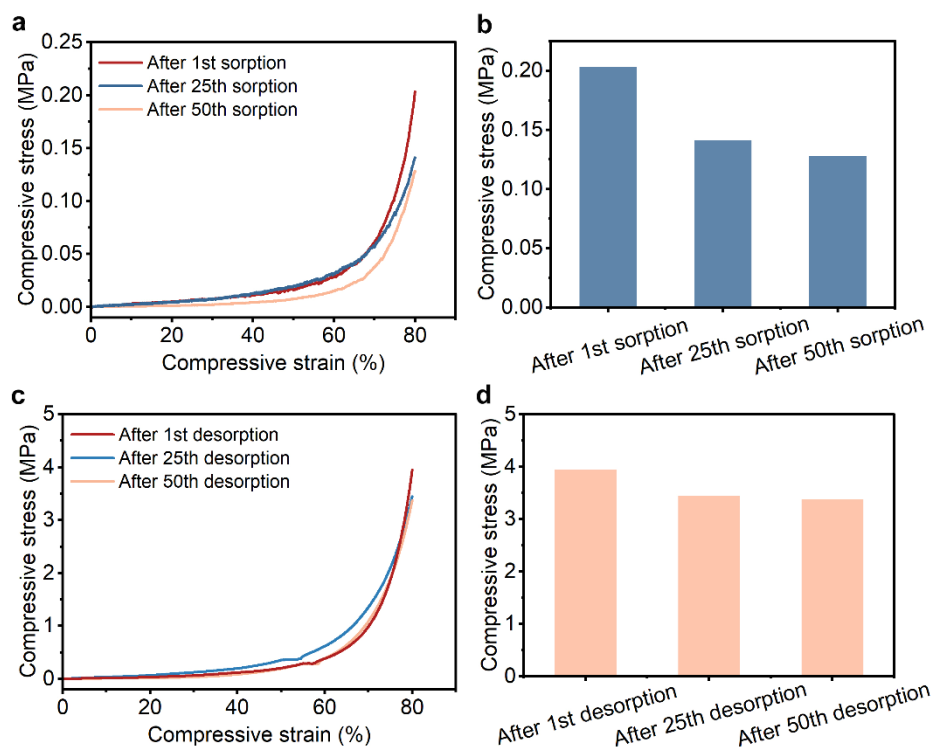


Figure R3. a) Compressive stress–strain curves and b) compressive stress of the sorbent after 1st sorption, 25th sorption and 50th sorption. c) Compressive stress–strain curves and d) compressive stress of the sorbent after 1st desorption, 25th desorption and 50th desorption.

Changes made:

We have modified and added the following description **in the revised manuscript (Page 14)**, highlighted in red, as shown below:

“Structural and material characterization further verifies durability, with the macroscopic morphology, microstructure, chemical composition, and mechanical properties remaining well preserved after cycling tests (Supplementary Figs. 29–32)”

We have added **Supplementary Figure 31 on page 34 of the revised Supplementary Information.**

6. The outdoor prototype data are promising but limited to a single day that clearly doubts over reliability. Authors should include more numbers of day results.

Response:

We sincerely appreciate your valuable suggestion. Indeed, the outdoor experiment conducted for only one day has inherent limitations and uncertainties. To address the reviewer's concerns, we conducted an additional 4-day outdoor experiment, in Xiamen, China, to assess the stability of both the sorbents and the device (Figure R4).

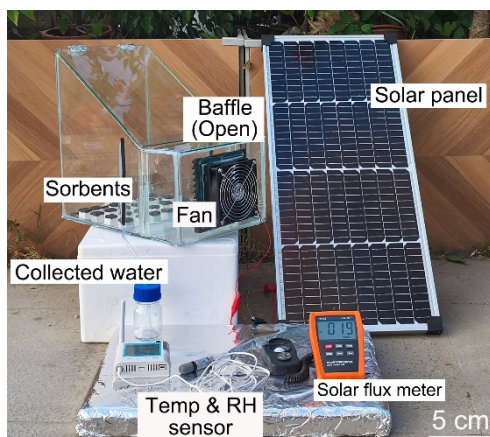


Figure R4. Photograph of the AWH device featuring a large-scale PHW array in operation in the outdoor environment of Xiamen, China.

During these four days, we recorded the outdoor weather conditions (including temperature, RH, and solar intensity), as shown in Figures R5-8. Owing to the winter season and the relatively low latitude of Xiamen, the sunrise time was relatively late. Therefore, during the 4-day outdoor experiments, we adopted a daily operation mode consisting of three sorption-desorption cycles to better match the actual environmental conditions. Specifically, sorption was conducted from 0:00 a.m. to 8:00 a.m., followed by desorption from 8:00 a.m. to 10:00 a.m. Subsequently, two additional cycles were performed, each comprising 2.5 h of sorption and 1 h of desorption.

The experimental results indicate that our device equipped with large-scale array sorbents, demonstrated reliable operational stability over these four days, with daily water production ranging from 0.96 to 1.37 L kg⁻¹ day⁻¹. The water collection efficiency varied between 70.59% and 81.06% (Figure R9). The specific daily water production and collection efficiency were influenced by the daily temperature, humidity conditions and solar intensity. This part has been added to the revised manuscript and the supplementary information, and relevant sections of the revised manuscript have been appropriately modified to further demonstrate the stability of the sorbents and device.

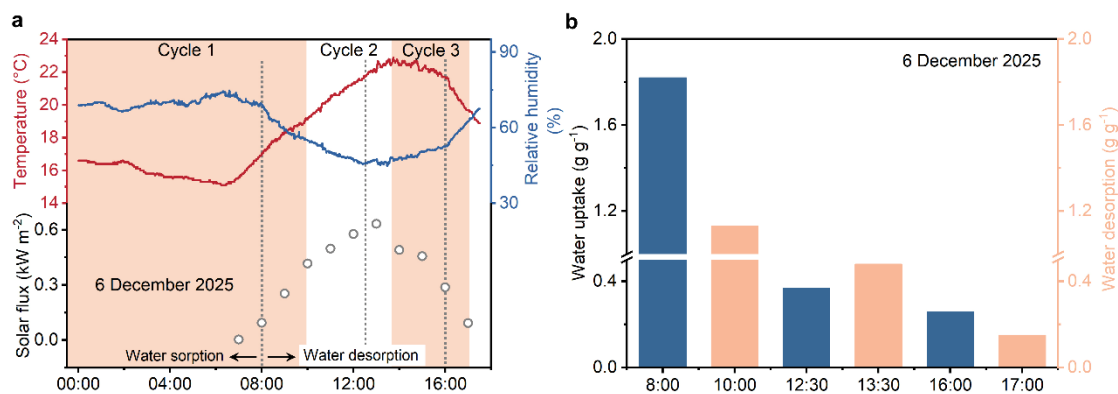


Figure R5. a) Changes in environmental conditions, including RH, temperature, and solar flux, during the first day of outdoor experiments in Xiamen, China. b) Water uptake and water desorption of the sorbents during four sorption-desorption cycles on the first day of outdoor experiments in Xiamen, China.

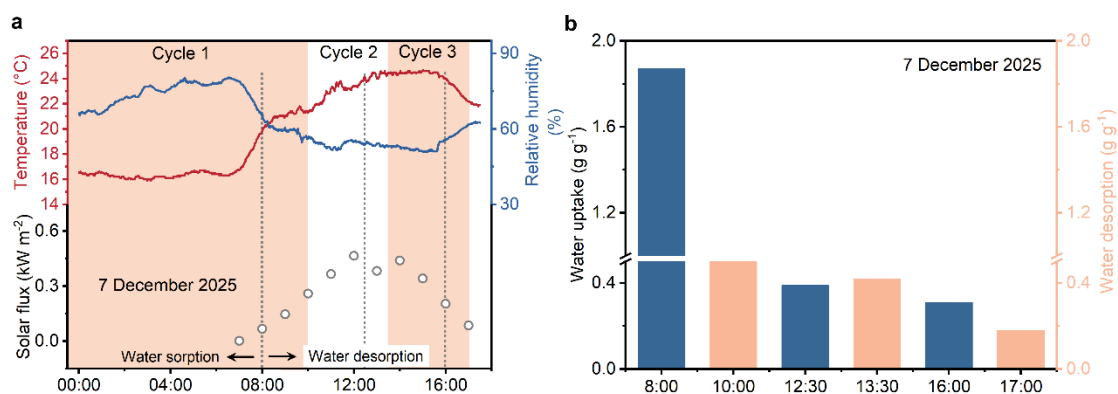


Figure R6. a) Changes in environmental conditions, including RH, temperature, and solar flux, during the second day of outdoor experiments in Xiamen, China. b) Water uptake and water desorption of the sorbents during three sorption-desorption cycles on the second day of experiments in Xiamen, China.

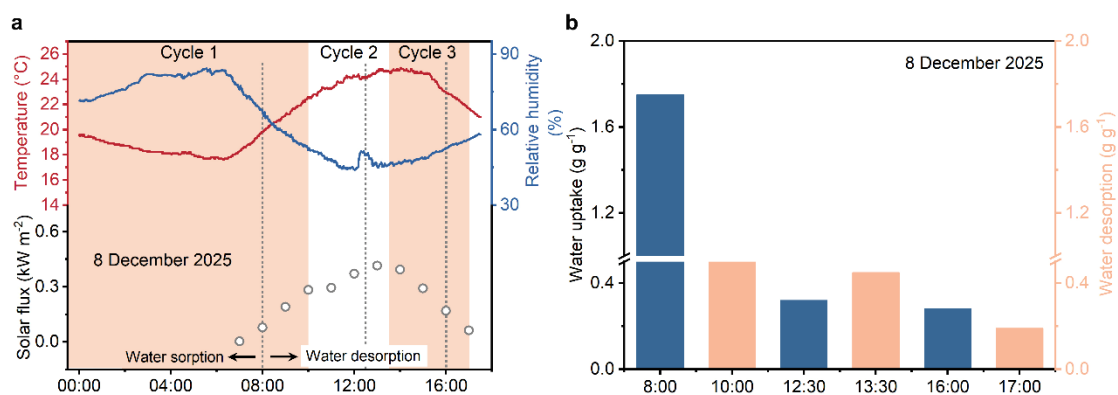


Figure R7. a) Changes in environmental conditions, including RH, temperature, and solar flux, during the third day of outdoor experiments in Xiamen, China. b) Water uptake and

water desorption of the sorbents during three sorption-desorption cycles on the third day of experiments in Xiamen, China.

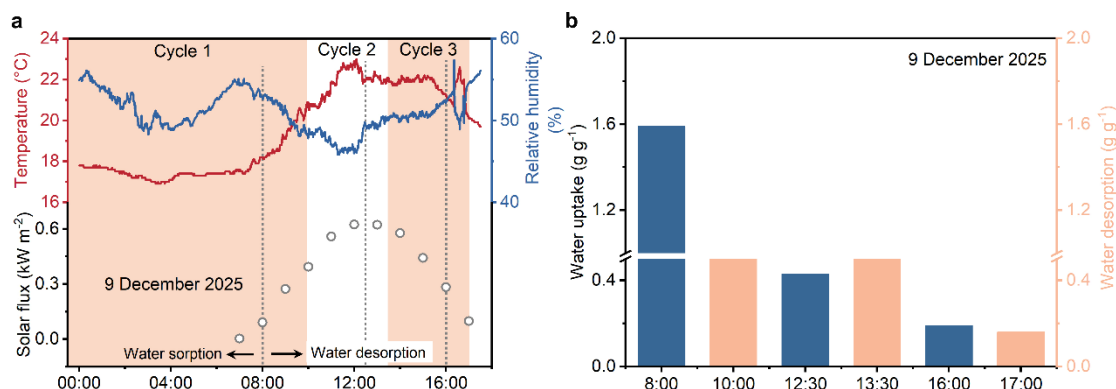


Figure R8. a) Changes in environmental conditions, including RH, temperature, and solar flux, during the fourth day of outdoor experiments in Xiamen, China. b) Water uptake and water desorption of the sorbents during three sorption-desorption cycles on the fourth day of experiments in Xiamen, China.

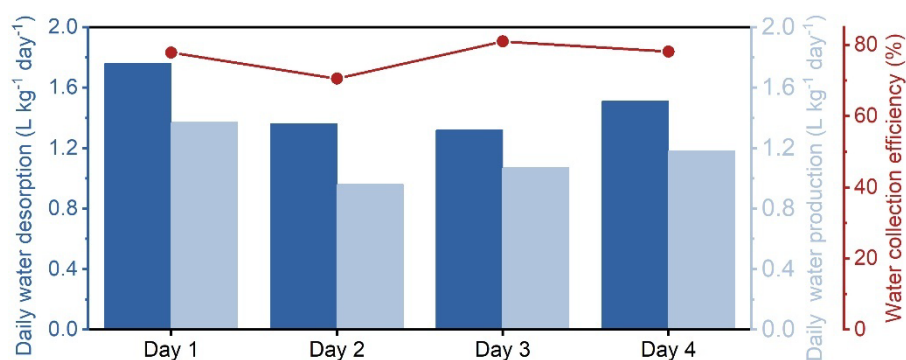


Figure R9. Daily water desorption, daily water production of the sorbents and water collection efficiency during long-term outdoor experiments.

Changes made:

We have redesigned **Figure 6 in the revised manuscript (Page 15)** to include data from the continuous multi-day outdoor experiment.

We have added **Supplementary Figs 38-40** in the **revised Supplementary Information (Page 37)**.

We have added the calculation formula for water collection efficiency **in the revised Supplementary Information (Page 8)**, Outdoor water harvesting measurements, highlighted in red, as shown below:

“In outdoor experiment, the water collection efficiency is calculated using the following formula:

$$\text{Water collection efficiency} = \frac{W_c}{W_d} \times 100\%$$

Where W_c (g) represents the mass of water collected from the AWH device during the outdoor experiment, and W_d (g) represents the mass of water desorbed from the sorbent array during the outdoor experiment.”

We have modified and added the following description **in the revised manuscript (Pages 16-17)**, highlighted in red, as shown below:

“This AWH device was then extended for testing in different seasons and regions. A four-day continuous experiment was conducted in Xiamen during the winter, from 00:00 AM on December 6 to 17:00 PM on December 9, 2025. Given the winter conditions in Xiamen and its relatively low latitude, which leads to delayed sunrise and limited effective daylight hours, a daily operational mode consisting of three sorption–desorption cycles were adopted to maximize water uptake and daily freshwater production (Fig. 5d). Specifically, the first sorption cycle was performed from 00:00 AM to 08:00 AM, followed by solar-driven desorption from 08:00 AM to 10:00 AM. To further capitalize on available daylight, two additional cycles were conducted within the same day, each comprising 2.5 h of sorption and 1 h of desorption. This multi-cycle strategy effectively utilized nocturnal high humidity in combination with daytime solar energy input, thereby enhancing overall water productivity under winter conditions.

Throughout the 4-day experiment, 24 sorbent units were placed in the sorption chamber during each overnight cycle. On the first day in Xiamen, the PHW array achieved a water uptake of 1.82 g g^{-1} under an average RH of 60.63% and an average ambient temperature of 18.67°C . The specific variations in ambient temperature, RH, and solar irradiation during the experiment are shown in Fig. 5e. Following sorption, the chamber was sealed by inserting the baffle, and the desorption process was initiated. During desorption, condensed water droplets were clearly observed on the inner surface of the chamber (Fig. 5c), confirming effective vapor release and collection. Two additional sorption–desorption cycles were subsequently completed on the same day.

As a result of this multi-cycle operation strategy, the sorbents in this AWH device achieved a freshwater production of $1.37 \text{ L kg}^{-1} \text{ day}^{-1}$ with a water collection efficiency of 77.84% on the first day of outdoor testing in Xiamen (Figs. 5f–g). The daily freshwater yield and collection efficiency exhibited moderate variations in response to day-to-day fluctuations in ambient temperature, humidity, and solar irradiation. During the subsequent three days in Xiamen, daily freshwater productions of 0.96, 1.07, and $1.18 \text{ L kg}^{-1} \text{ day}^{-1}$ were obtained, with corresponding collection efficiencies of 70.59%, 81.06%, and 78.15%, respectively (Fig. 5g, Supplementary Table 6 and Supplementary Figs. 38–40), demonstrating stable and reliable freshwater generation under real outdoor conditions.”

7. Additionally, specify the mass of sorbent, collection efficiency, and ambient

RH/temperature variation in tabular format for clarity.

Response:

We sincerely appreciate your valuable suggestion. The data on sorbent mass, collection efficiency, daily water production, as well as the ambient relative humidity (RH) and temperature variations, are indeed crucial for clearly presenting the experimental results. In response to your suggestion, we have organized these data into a table to provide a clearer presentation of the experimental conditions and results (Table R5). Regarding the temperature and humidity variations, we have presented the specific fluctuations in Figures R5-8, while the table shows the average values of temperature and humidity during the experiment. Furthermore, to enhance the credibility of our outdoor experimental data, we have supplemented detailed data from the continuous multi-day outdoor operation, including sorbent mass, collection efficiency, daily water production, and ambient RH and temperature variations, covering the 4-day experimental period.

Table R5. Outdoor Experiment Data on Sorbent Performance and Environmental Conditions

Day	Sorption zone mass (g)	Daily water production ($L_{\text{water}} \text{ kg}_{\text{sorbent}}^{-1}$)	Collection efficiency (%)	Average ambient RH (%)	Average ambient temperature (°C)
One day in Harbin, China	32.49	1.72	79.11	62.95	29.24
Day 1 in Xiamen, China	31.54	1.37	77.84	60.63	18.67
Day 2 in Xiamen, China		0.96	70.59	63.40	19.99
Day 3 in Xiamen, China		1.07	81.06	63.74	21.13
Day 4 in Xiamen, China		1.18	78.15	51.2	20.20

Changes made:

We have added **Supplementary Table 6** in the **Supplementary Information (Page 12)**.

We have added the calculation formula for water collection efficiency **in the revised Supplementary Information (Page 8)**, Outdoor water harvesting measurements, highlighted in red, as shown below:

In this outdoor experiment, the water collection efficiency is calculated using the following formula:

$$\text{Water collection efficiency} = \frac{W_c}{W_d} \times 100\%$$

Where W_c (g) represents the mass of water collected from the AWH device during the outdoor experiment, and W_d (g) represents the mass of water desorbed from the sorbent array during the outdoor experiment.

8. The levelized cost of water (LCOW) analysis lacks assumptions (e.g., sorbent cost, system lifetime, maintenance frequency). Please detail the economic parameters and sensitivity range used in modeling.

Response:

We appreciate your detailed feedback and valuable suggestions. Based on the comments, we have thoroughly revised our Levelized Cost of Water (LCOW) model and clarified the economic assumptions, parameter choices, and sensitivity ranges used. Specifically, we have provided clear definitions for sorbent costs, system lifetime, maintenance frequency, and their associated sensitivity ranges. After detailed calculations, the raw material cost is 13.14 USD, the electricity cost for sorbent preparation is 7.66 USD, and the sorbent lifetime is 2 years. The Capital Recovery Factor (CRF) is calculated using an 8% discount rate and a 2-year lifespan. The annualized sorbent cost is 11.66 USD per year. The outdoor AWH equipment cost is 27.58 USD with a 10-year lifetime, and at an 8% discount rate, the annualized cost is 4.11 USD per year. The maintenance cost is based on 5% of the initial total equipment investment per year, resulting in an annual maintenance cost of 1.38 USD L⁻¹. There is no electricity consumption during system operation, so the operational electricity cost is 0 USD. The annual total cost is 17.15 USD year⁻¹. The annual water production is calculated based on the global water production potential of our sorbent, which ranges from 0.08 to 2.48 L kg⁻¹ day⁻¹.

1. LCOW Definition and Calculation Formula (USD/L)

Levelized Cost of Water (LCOW) is defined as the cost of producing each unit of water, calculated as:

$$LCOW = \frac{\text{Annualized CAPEX} + \text{OPEX}}{\text{Annual Water Yield}}$$

Components of LCOW:

Annualized CAPEX: The annualized capital expenditure (CAPEX) represents the yearly cost of equipment investment after amortizing it over the system's lifetime and discount rate.

OPEX: The operating expenses (OPEX), which include:

- (i) The equivalent annual cost of sorbent re-manufacturing/replacement (combining raw material costs and fabrication electricity),
- (ii) Maintenance costs (calculated as a percentage of the initial device investment),
- (iii) Operational electricity costs (for the outdoor AWH system, this is 0 due to solar-powered operation).

Annual Water Yield: The annual water yield refers to the amount of water produced by the sorbents over the course of one year.

2. Sorbent Cost (Raw Materials + Fabrication Electricity)

The sorbent cost consists of two parts:

Raw Material Cost: The cost of materials required for sorbent production, which is 13.14 USD, is detailed in [Table R6](#).

Table R6. The raw material cost for preparing 24 PHW arrays.

Raw materials	Unit Price (USD/g)	Mass (g)	Cost (USD)
Balsa wood	0.07	30.35 g	2.12
NaClO ₂ (7758-19-2)	0.019	80	1.52
CH ₃ COOH (64-19-7)	0.0081	16.78	0.14
NaOH (1310-73-2)	0.018	120	2.17
LiCl (7447-41-8)	0.2222	18.29 g	4.06
PEG (25322-68-3)	0.015	55.13 g	0.83
AA (79-10-7)	0.0167	8.04 g	0.13
MBAA (110-26-9)	0.0764	0.40 g	0.03
APS (7727-54-0)	0.0136	0.16 g	0.0022
CNT (308068-56-6)	1.6667	0.3312 g	0.55
Deionized water	0.0003	5300 g	1.59

Electricity Cost: The electricity consumed during sorbent preparation, amounting to 7.66 USD for steps such as stirring, heating, and freeze-drying, is detailed in [Table R7](#), based on the electricity price of \$0.10 per kilowatt-hour.

Table R7. Electricity Consumption and Cost for PHW Fabrication (per batch of 24 samples).

Step	Operation description	Estimated power consumption (kWh)	Electricity cost (USD)	Equipment and power ratings
NaClO ₂ , NaOH and LiCl solution preparation	Magnetic stirring for a total of 20 min	0.06	0.006	Magnetic stirrer, 200 W
Wood treatment	80 °C water bath × 24 h (2% NaClO ₂ solution) + 80°C × 10 h (8% NaOH solution)	20.4	2.04	Constant temperature water bath, 600 W

Melting PEG	80°C oven heating for 30 min	0.5	0.05	Oven, 1000 W
Precursor solution Stirring	Magnetic stirring for 10 min	0.033	0.0033	Magnetic stirrer, 200 W
Thermal curing	60 °C thermal curing for 6h	6	0.6	Oven, 1000W
All freezing and freeze-drying	−10°C freezing 8h × 2, −80°C freeze-drying 48h × 2	49.6	4.96	Small refrigerator, 100w; Small freeze dryer, 500W
Total	—	76.6	7.66	—

Fabrication Electricity Cost: The electricity consumed during sorbent preparation, which amounts to 7.66 USD for steps such as stirring, heating, and freeze-drying.

Thus, the total sorbent cost is:

$$C_{\text{sorb},0} = 13.14 + 7.66 = 20.80 \text{ USD}$$

Assuming the sorbent lifetime is 2 years, the annualized sorbent cost is:

$$C_{\text{sorb},\text{ann}} = 20.80 \times CRF(0.08,2) = 20.80 \times 0.5608 = 11.66 \text{ USD year}^{-1}$$

Where CRF (capital recovery factor) is calculated using the 8% discount rate and a 2-year lifespan.

3. Equipment Cost and System Lifetime

The equipment cost refers to the hardware investment for the outdoor AWH system, including the fan, glass chamber, and solar panel, totaling 27.58 USD, as detailed in [Table R8](#).

Table R8. Total capital investment of the device.

Component	Unit Cost (USD)	Quantity	Total Cost (USD)
Fan	3.72	1	3.72
Glass device	12.41	1	12.41
Solar panel	11.45	1	11.45
Total	—	—	27.58

The annualized equipment cost is calculated by amortizing the total cost over the system's lifetime (10 years) at an 8% discount rate:

$$C_{\text{equip},\text{ann}} = 27.58 \times CRF(0.08,10) = 27.58 \times 0.1490 = 4.11 \text{ USD year}^{-1}$$

The system lifetime is assumed to be 10 years, which is typical for outdoor small-scale systems.

4. Maintenance Costs

Maintenance costs are typically calculated as a percentage of the initial equipment investment. In this model, we assume a maintenance rate of **5% per year**, which results in an annual maintenance cost of:

$$C_{\text{maint}} = 0.05 \times 27.58 = 1.38 \text{ USD year}^{-1}$$

This includes periodic equipment checks, part replacements, and other necessary operational maintenance.

5. System Electricity Costs

Since the system is entirely solar-powered, the operational electricity cost is **0 USD**. The electricity consumption only occurs during sorbent preparation, and this cost is already accounted for in the sorbent cost (7.66 USD).

6. Annual Water Yield

The annual water yield depends on the water production potential of the sorbent, which ranges from 0.08 to 2.48 L kg⁻¹ day⁻¹. Assuming there are 365 days in a year, the annual water yield can be calculated as:

$$Y_{\text{water}} = 365 \times L_{\text{day}} \times M_s$$

where Y_{water} (L) is the annual water yield, L_{day} (L kg⁻¹ day⁻¹) the water production potential, and M_s (kg) is the sorbent mass.

7. Final LCOW Calculation Formula

Substituting the economic assumptions into the LCOW formula, we arrive at the final LCOW calculation:

$$LCOW = \frac{17.15}{365 L_{\text{day}} M_s}$$

Where 17.15 USD is the total annual cost, L_{day} (L kg⁻¹ day⁻¹) the water production potential, and M_s (kg) is the sorbent mass.

8. Sensitivity Analysis

To assess the sensitivity of LCOW to changes in the economic parameters, we provide the following ranges (Table R9):

Table R9. Sensitivity Analysis of LCOW Parameters.

Parameter	Base value	Sensitivity range	Reason
Sorbent raw material cost ($C_{\text{sorb},0}$)	20.80 USD	±20% (16.64–24.96)	Market fluctuations
Sorbent lifetime (n_{sorb})	2 years	1–3 years	Sorbent degradation over time
Equipment cost ($C_{\text{equip},0}$)	27.58 USD	±20% (22.06–33.10)	Equipment price fluctuations

System lifetime (n_{sys})	10 years	8–15 years	Durability of outdoor systems
Maintenance rate	5% year ⁻¹	5–10% year ⁻¹	Standard engineering practice
Discount rate (i)	8%	5–12%	Economic environment variations
Annual operating days (N_{day})	365	300–365	Climate-dependent operation frequency
Water production potential (L_{day})	0.08–2.48 L·kg ⁻¹	Fixed	Climate-driven parameter
Operational electricity cost	0	0	Solar-powered operation

By incorporating sorbent costs, equipment costs, maintenance fees, system lifetime, and climate differences, we arrive at the LCOW calculation formula. The sensitivity analysis shows the key parameters affecting LCOW. Finally, LCOW is reported in USD L⁻¹ for easier comparison across different regions.

Changes made:

We have updated and added more details, then recalculated the LCOW in the **Supplementary Note 9 of the SI (Pages 18-22)**

We have also updated **Figure 6c and Figure 6d in the revised manuscript (Page 18)**.

The related description **in the revised manuscript (Page 18)** has been updated and modified, highlighted in red, as shown below:

“We further calculated the levelized cost of water (LCOW) for each country by integrating the annual water yield with the annualized system cost (Fig. 6c). The calculation incorporates key economic parameters, including sorbent and device costs, system lifetime, and maintenance frequency, and further accounts for the sensitivity ranges associated with reasonable variations in these parameters (Supplementary Note 8).”

9. The reported LCOW < 0.01 USD/L in humid regions seems unrealistically low; verify with actual sorbent cost inputs.

Response:

We have carefully reviewed all the cost data involved in the calculations, including **raw material costs, energy consumption for sorbent preparation, maintenance costs, and equipment costs**. These cost data were initially underestimated, particularly when considering the **sorbent costs** and **equipment maintenance**. After re-verification, we have confirmed the following:

Revised Cost Verification and Model Update

(1) Sorbent Costs:

We re-verified the raw material costs and energy costs for sorbent preparation to

ensure the accuracy of all relevant data. The total raw material cost is 13.14 USD, and the energy consumption cost for the preparation process is 7.66 USD, making the total sorbent cost 20.80 USD. We also confirmed that the annualized sorbent cost is 11.66 USD/year, based on a 2-year sorbent lifespan.

(2) Equipment Costs:

We reassessed the initial investment in the equipment, including the procurement costs for the fan, glass device, and solar panel, and confirmed the total initial equipment investment is 27.58 USD. Using an 8% discount rate, we applied amortization and derived the annualized equipment cost of 4.11 USD/year.

(3) Maintenance Costs:

Based on industry standards, we confirmed the annual maintenance cost for the equipment is 1.38 USD/year, which is 5% of the initial equipment investment.

(4) System Operational Costs:

Since the system operates entirely on solar power, we verified that the operational electricity cost is 0 USD/year.

As a result, we confirmed that the annual total cost is 17.15 USD/year. The annual water production is calculated based on the global water production potential of our sorbent, which ranges from 0.08 to 2.48 L kg⁻¹ day⁻¹. Based on this, we recalculated the LCOW, which now provides more reasonable and accurate results.

LCOW Calculation and Regional Differences

Based on the re-verified cost data, we derived the specific LCOW values for different regions. Specifically, the LCOW in humid regions ranges from 0.14–0.22 USD/L, while in other regions, the LCOW varies according to climate and water production potential. The following is an explanation of the LCOW for different regions, with specific examples of countries for each region:

(1) Humid Regions LCOW: 0.14-0.22 USD/L

In humid regions, the atmospheric water vapor content is consistently high, leading to a substantially higher annual water production per unit mass of sorbent (L kg⁻¹). As a result, the LCOW in these regions is relatively low, ranging from 0.14 to 0.22 USD L⁻¹. This demonstrates that, under high-humidity conditions, freshwater can be produced at a comparatively low cost per unit volume, even when conservative and verified cost inputs are applied. Representative countries in this category include Equatorial Guinea (0.158 USD L⁻¹), French Guiana (0.163 USD L⁻¹), Colombia (0.164 USD L⁻¹), Brazil (0.219 USD L⁻¹), and Ghana (0.217 USD L⁻¹).

(2) Arid Regions LCOW: 0.28-1.33 USD/L

In contrast, arid regions are characterized by extremely low atmospheric moisture content, which significantly limits sorbent water uptake and reduces annual water yield. Consequently, the LCOW in these regions is markedly higher, typically ranging from 0.28 to 1.33 USD L⁻¹, despite comparable system configurations and cost structures. Representative countries include Saudi Arabia (0.9 USD L⁻¹), Egypt (0.80

USD L⁻¹), Israel (0.54 USD L⁻¹), Jordan (0.76 USD L⁻¹), Qatar (0.69 USD L⁻¹), and Kuwait (1.33 USD L⁻¹).

After reverifying the data, we find that LCOW values below 0.01 USD L⁻¹ cannot be obtained when realistic costs of equipment, maintenance, sorbent, and water production potential are considered. Even under humid climatic conditions, the LCOW remains well above 0.01 USD L⁻¹. In the revised calculations, the LCOW for humid regions falls within the range of 0.14–0.22 USD L⁻¹.

Changes made:

We have updated and added more details, then recalculated the LCOW in the **Supplementary Note 9 of the SI (Pages 18-22)**

We have also updated **Figure 6c and Figure 6d in the revised manuscript (Page 17)**.

The related description **in the revised manuscript (Page 18)** has been updated and modified, highlighted in red, as shown below:

“The results reveal pronounced regional disparities in water affordability that are primarily governed by climatic water availability. In high-yield, humid regions, the LCOW typically falls within the range of 0.14–0.22 USD L⁻¹, whereas in arid and extremely arid regions, the LCOW increases substantially and can reach values of 0.7–1.3 USD L⁻¹. As illustrated in Fig. 6d, countries such as Malaysia, Colombia, Vietnam, and the Philippines exhibit the lowest LCOW values owing to persistently high relative humidity and favourable temperature conditions that enhance the efficiency of sorption–desorption cycles. In contrast, countries including Iran, Namibia, Jordan, and Saudi Arabia display markedly higher LCOWs, primarily due to limited atmospheric moisture under arid climatic conditions, which constrains the water uptake capacity and overall performance of the sorbents.”

10. Clarify abbreviations on first use (e.g., HW, PHW, PCM, RH, LCOW).

Response:

Many thanks for your helpful comment. We confirm that all abbreviations are defined with their full terms at their first occurrence in the manuscript. For the reviewer’s convenience, the first appearance of each abbreviation has been highlighted in yellow in the revised manuscript to facilitate quick identification, without altering the original structure or wording of the text.

Changes made:

“HW” corresponded to “hygroscopic wood”. **(Page 12 in the revised manuscript)**

“PHW” corresponded to “PCM-integrated hygroscopic wood PHW”. **(Abstract, Page 1 in the revised manuscript)**

“PCM” corresponded to “phase change materials”. **(Abstract, Page 1 in the revised manuscript)**

“RH” corresponded to “relative humidity”. (**Introduction, Page 2 in the revised manuscript**)

“LCOW” corresponded to “levelized Cost of Water”. (**Page 18 in the revised manuscript**)

11. Correct typographical errors such as “confiemd” → “confirmed”, “exhibites” → “exhibits”.

Response:

We appreciate your careful reading. All typographical errors mentioned, including “confirmed” and “exhibits”, have been corrected. We have also conducted a thorough proofreading of the entire manuscript to ensure consistency and accuracy. To facilitate your review, we have highlighted the changes made in the revised manuscript.

Changes made:

We have corrected “confiemd” to “confirmed” (**Page 5 in the revised manuscript**), highlighted in red.

We have corrected “exhibites” to “exhibits” (**Page 7 in the revised manuscript**), highlighted in red.

12. In Figure 3g, specify reference numbers beside data points for easier traceability.

Response:

Thank you for your valuable suggestion. Considering that each study involves multiple water uptake values at different humidity levels, annotating each data point with its corresponding reference may lead to a cluttered figure. Therefore, we propose grouping all data points from the same study using a single color and uniformly indicating the reference for that study, as shown in Figure R10. This approach will enhance clarity and traceability without overcomplicating the figure. We greatly appreciate your insightful feedback and welcome any further suggestions.

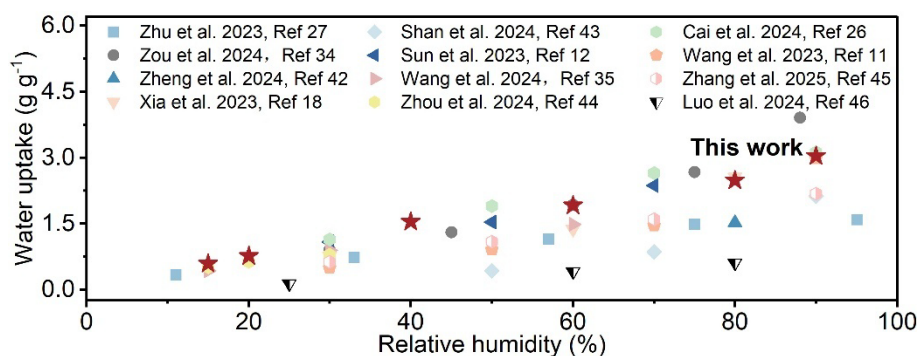


Figure R10. Comparison of water uptake performance between our work and reported sorbents for AWH application.

Changes made:

We have updated the **Figure 3g** in the revised manuscript (Page 9).

13. Mention the uncertainty/error bars in all experimental plots (uptake, desorption rate, etc.).

Response:

Thank you very much for your valuable comment. We have added error bars to several figures using a suitable layout, although in some cases they may affect the visual clarity of the curves. To ensure readability without overcrowding the plots, we will also provide a separate dataset reporting the mean values and the corresponding uncertainty for each data point shown in the curves. This approach preserves figure clarity while making the uncertainty information fully available.

14. Ensure consistent units (e.g., g g^{-1} , kg m^{-2}) and specify whether “ g g^{-1} ” refers to per gram of sorbent or total device.

Response:

Thank you very much for your valuable suggestion. We have standardized the units throughout the manuscript. In our initial experiments, the desorption process was presented using the unit “ kg m^{-2} ”, which could potentially lead to confusion and misinterpretation of our results. To present the data more clearly with consistent units, we have reconducted the desorption experiments and used “ g g^{-1} ” as the unit. For clarification, “ g g^{-1} ” refers to the amount per gram of sorbent.

In order to ensure unit consistency, we have reconducted the PHW (with PCM) and HW (without PCM) desorption experiments. The experiments involved sorption for 12 h at 90% RH, followed by 2 h of desorption, which included 1 h under solar intensity of 1 kW m^{-2} and 1 h in darkness. Using the resulting data, we calculated the mass change and desorption rate during the desorption process, as shown in [Figure R11](#).

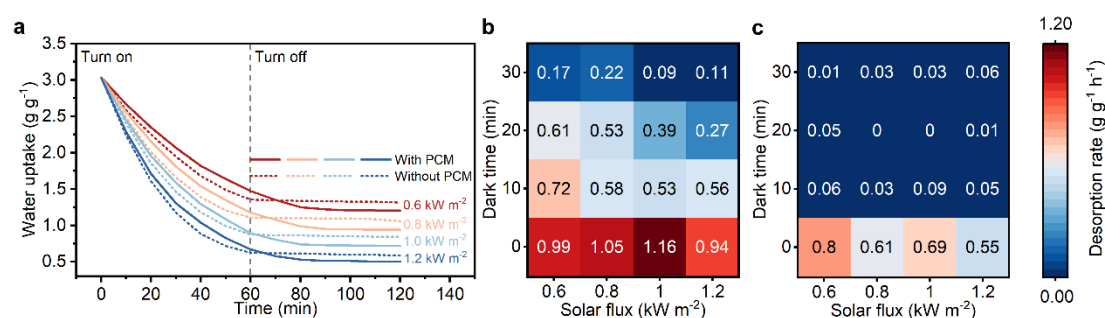


Figure R11. a) Dynamic desorption curves of PHW (with PCM) and HW (without PCM) under different solar intensities ($0.6\text{--}1.2 \text{ kW m}^{-2}$) and then in darkness within 120 min. The desorption rate of g) PHW (with PCM) and h) HW (without PCM) within different dark times

after solar radiation cessation (0.6-1.0 kW m⁻²).

Changes made:

We reconducted the sorption-desorption experiments and standardized the units to “g g⁻¹”.

We have updated **Figures 4f, 4g, and 4h** accordingly (**Page 12 in the revised manuscript**).

In addition, the corresponding descriptions of the desorption experiments have been revised and clarified **on Page 13 of the revised manuscript**, with the relevant text is highlighted in red, as detailed below:

“Under adequate solar radiation, PHW showed slightly lower desorption mass changes (1.56–2.36 g g⁻¹) than those of HW (1.67–2.41 g g⁻¹) under solar intensities of 0.6–1.2 kW m⁻² within 60 min (Fig. 4f). However, the maximum difference was less than 7%, indicating that the incorporation of PCM had a minimal impact on the normal solar-driven desorption performance.

The stored latent heat is subsequently released to sustain water desorption under weak solar input and in darkness. Within the first 10 min after light cessation (0.6–1.2 kW m⁻²), PHW maintains a high desorption rate of 0.56–0.72 g g⁻¹ h⁻¹ (Fig. 4g). In contrast, desorption from HW nearly stops immediately, with rates of only 0.05–0.06 g g⁻¹ h⁻¹ (Fig. 4h), highlighting the effectiveness of latent-heat-driven water release. As the solar intensity increases from 0.6 to 1.2 kW m⁻², the desorption rate of PHW decreases slightly after the light is turned off, likely because less water remains in the sorbent, reducing the driving force for further desorption.

Notably, even 30 min after light cessation, PHW still sustains a desorption rate of 0.11–0.17 g g⁻¹ h⁻¹. Over a full day–night simulation cycle consisting of 60 min irradiation (0.6–1.2 kW m⁻²) followed by 60 min darkness, PHW achieves larger desorption mass changes of 1.83, 2.09, 2.32, and 2.52 g g⁻¹ h⁻¹, compared with 1.72, 1.95, 2.18, and 2.45 g g⁻¹ for HW. This improvement is enabled by a high latent-heat utilization efficiency of up to 86.65%, based on the PCM latent enthalpy and the enthalpy required for water desorption (Supplementary Notes 5 and 6)”

Reviewer #2

This manuscript addresses two core bottlenecks of AWH—slow adsorption due to diffusion resistance and desorption interruptions caused by intermittent solar energy—through material design (wood-derived sponge) and a PAA-CNT-PEG photothermal phase-change hydrogel. However, the manuscript exhibits significant shortcomings in the depth of its mechanism explanation, quantitative comparison of key performance metrics, and rigorous analysis of practical applications. It requires supplementation with critical data and analysis to substantiate its innovation and potential impact.

Response:

We sincerely thank you for your constructive feedback and valuable suggestions. We greatly appreciate your careful evaluation of our work. In response, we have made detailed revisions to improve the depth of the mechanism explanation, provide a quantitative comparison of key performance metrics, and offer a more rigorous analysis of practical applications. Additionally, we have supplemented the manuscript with critical data and relevant analyses. We have carefully addressed each of your points and made the necessary revisions to enhance the clarity and quality of the manuscript. We believe these changes will meet your expectations and contribute to the overall improvement of the paper.

1. Supplement data on the adsorption and desorption performance variations of materials under different ambient temperatures to enhance their practical application scenarios.

Response:

Thank you very much for your constructive suggestion. We fully agree that evaluating the sorption and desorption performances of our sorbent under different ambient temperatures is essential for assessing its suitability in diverse practical application scenarios. In response, we conducted systematic sorption experiments across a wide range of RH at various temperatures, as well as desorption tests under different ambient temperature conditions. These measurements allowed for a comprehensive evaluation of the temperature-dependent sorption behavior of the material and its robustness in realistic operating conditions.

#Sorption performance

To evaluate the sorption performance of the sorbent at different temperatures, sorption tests were conducted within the RH range of 20% to 80%, at temperatures of 15 °C, 25 °C, 35 °C, 45 °C, and 55 °C (Figure R12). The results demonstrate that the sorbent exhibits low sensitivity to temperature variations, while showing a stronger dependence on relative humidity, highlighting its adaptability across varying climatic conditions. Specifically, within the 20% RH to 80% RH range, the equilibrium sorption capacity of the sorbent decreases only slightly as the temperature increases from 15 °C to 55 °C. At 20% RH, the equilibrium water uptake at 55 °C is only reduced by approximately 0.33 g g⁻¹ compared to 15 °C; at 40% RH, 60% RH, and 80% RH, the water uptake decreases by 0.30

g g^{-1} , 0.23 g g^{-1} , and 0.40 g g^{-1} , respectively. These findings indicate that the sorbent demonstrates good stability across a range of temperatures, with significant potential for application, especially in water harvesting systems designed to function under fluctuating temperature and humidity conditions.

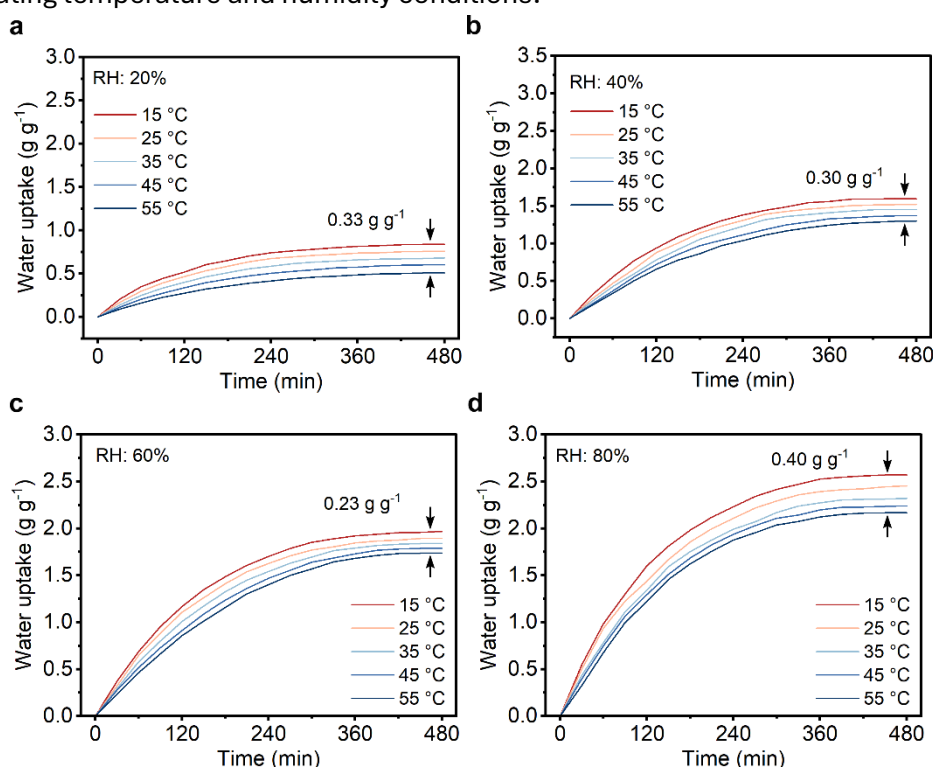


Figure R12. Water uptake of the sorption zone in PHW from 15 °C to 55 °C under RH of a) 20%, b) 40%, c) 60%, and d) 80%.

#Desorption performance

To evaluate the desorption performance of our sorbents under different ambient temperatures, we designed a simulated high-temperature and temperature fluctuation experiment to systematically assess the thermal desorption behavior and thermal buffering capability of PCM-integrated sorbents. Before the desorption experiments, all sorbents had sorbed moisture at 90% RH to reach equilibrium sorption state. In this study, the PCM in the energy management zone of the sorbents is capable of undergoing complete phase transition and storing latent heat at 50 °C. Based on this property, PHW (with PCM) and HW (without PCM) sorbents were placed in ovens set at 50 °C, 70 °C, and 90 °C for 60 min to simulate high-temperature desorption conditions. Subsequently, the sorbents were immediately transferred to a 25 °C environment to examine their sustained desorption behavior under abrupt temperature fluctuations (Figure R13).

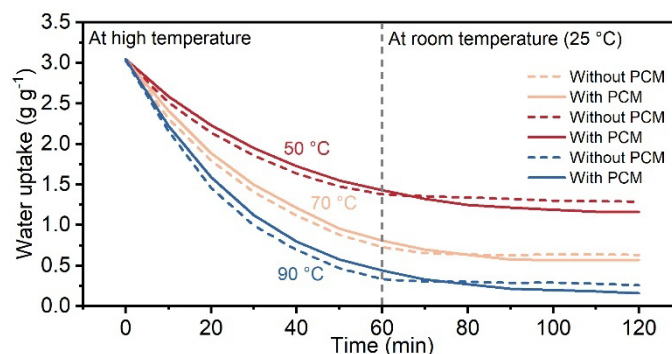


Figure R13. Overall desorption performance of PHW (with PCM) and HW (without PCM), including the initial desorption stage under simulated high-temperature conditions (50 °C, 70 °C, and 90 °C) followed by the subsequent desorption stage at 25 °C.

During the initial high-temperature desorption stage, the mass change of desorption in PHW (with PCM) was slightly lower than that in HW (without PCM). This difference can be attributed to the fact that PHW not only undergoes water desorption but also stores a portion of the supplied heat through the PCM phase transition, thereby reducing the water release. However, once the sorbents were removed from the high-temperature environment and transferred to a lower 25 °C environment, HW rapidly ceased desorption due to the sudden temperature drop. In contrast, PHW continued to release water with a significantly higher release rate. This continued desorption is due to the PCM undergoing phase transition upon entering the lower temperature environment, gradually releasing the stored latent heat, which helps maintain a higher internal temperature in PHW and allows for sustained desorption. This distinct thermal-buffering effect is absent in HW.

Therefore, although PHW exhibited slightly lower desorption during the initial high-temperature stage, its overall water release surpassed that of HW due to its continued desorption during the temperature fluctuation stage. At 50°C, 70°C, and 90°C, followed by a transfer to a 25 °C room temperature environment throughout the process, the total water release of PHW was 1.88, 2.47, and 2.88 g g⁻¹, respectively, all of which were higher than the corresponding values for HW (1.75, 2.40, and 2.77 g g⁻¹). These results clearly demonstrate that PHW not only exhibits effective thermal desorption performance under simulated high-temperature conditions but also displays excellent thermal buffering ability when experiencing sudden temperature changes. This performance indicates that PHW has strong application potential in practical scenarios where environmental temperature fluctuations are significant.

Changes made:

We have added description of the sorption kinetics at different ambient temperatures (**Page 10 in the revised manuscript**), highlighted in red, as shown below:

“In addition, dynamic sorption is highly sensitive to RH but only weakly dependent on temperature, indicating robust sorption kinetics and stable performance across different

climates (Supplementary Fig. 21).”

We have added description of the desorption kinetics at different ambient temperatures (Page 14 in the revised manuscript), highlighted in red, as shown below:

“Moreover, PHW exhibited excellent desorption performance and adaptability to different practical application scenarios, including under fluctuating ambient temperatures and simulated high-temperature conditions (Supplementary Figs. 23-24).”

We have already added **Supplementary Figure 21 on page 31 of the revised SI** and **Supplementary Figure 24 on page 32 of the revised SI**.

2. Explain the suitability of the PCM layer under high-temperature environmental conditions during summer.

Response:

We greatly appreciate your insightful comments. Clarifying the suitability of the PCM layer under high-temperature conditions during the summer is indeed crucial for defining the realistic working scenarios of our sorbent. In the energy management zone of PHW, the PCM absorbs solar energy and stores latent heat through a solid-to-liquid phase transition during solar illumination. Upon cessation of illumination, the PCM releases the stored latent heat, thereby maintaining the desorption of water. However, under high environmental temperatures, particularly during the summer, the indoor temperature may exceed the crystallization temperature of the PCM, preventing it from effectively releasing the stored latent heat and thus impacting the continuity of the desorption process.

To evaluate the desorption performance of our sorbents under different environmental temperatures, we employed a constant temperature and humidity chamber for temperature control, thereby simulating different climatic conditions. Before the desorption experiments, all sorbents had sorbed moisture at 90% RH to reach equilibrium sorption state. During the desorption tests, the ambient temperature was strictly maintained at 18 °C, 24 °C, 30 °C, and 42 °C (Figure R14). Each desorption cycle consisted of a 60-min illumination stage, with solar irradiation intensity set at 1 kW m⁻², followed by a dark stage.

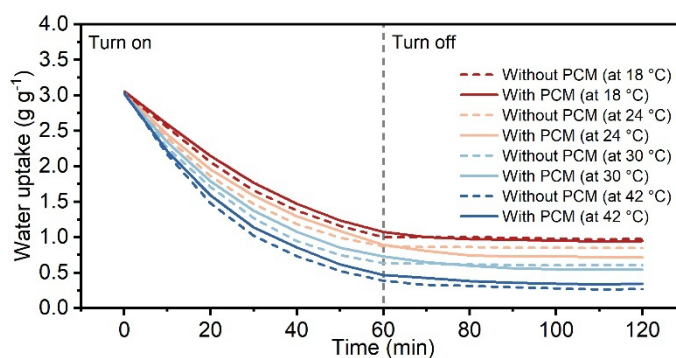


Figure R14. Desorption performance of PHW (with PCM) and HW (without PCM) at ambient temperatures of 18 °C, 24 °C, 30 °C and 42 °C. The desorption process consisted of an initial stage under 1 kW m⁻² solar irradiation, followed by a dark stage.

During the illumination stage, PHW exhibited a lower mass change of water release than HW at all tested temperatures. This phenomenon can be attributed to the PCM in PHW stores a portion of the solar energy as latent heat, thereby reducing the immediate heat available for desorption. Once the illumination ceased, the desorption process of HW decreased rapidly and nearly ceased at ambient temperatures below 30 °C. In contrast, PHW continued to release water by utilizing the stored latent heat, prolonging the desorption process into the dark period.

Despite fluctuations in ambient temperatures ranging from 18 °C to 30 °C and inevitable heat losses, PHW consistently demonstrated superior cumulative water release compared to HW. For instance, under the lowest ambient temperature of 18 °C, where heat loss was most significant, PHW achieved a water desorption capacity of 2.11 g g⁻¹, outperforming HW, which achieved 2.07 g g⁻¹. At 24 °C and 30 °C, the water desorption capacities of PHW were higher than those of HW by 6.91% and 1.64%, respectively. However, at the extremely high ambient temperature of 42 °C, which exceeded the crystallization temperature of the PCM, PHW could not effectively release latent heat. Nevertheless, due to these extreme high-temperature conditions, both PHW and HW continued desorption after illumination stopped, with the final desorption mass of HW slightly higher than that of PHW.

Despite this, PHW demonstrated excellent performance in coping with the intermittency of solar radiation within the more typical ambient temperatures of 18 °C to 30 °C, which are more common in real-world environments. Moreover, such extreme high-temperature conditions are infrequent. These results suggest that PHW can effectively tolerate variations in ambient temperature and intermittent solar radiation, maintaining stable and sustained desorption performance. Its ability to store and release thermal energy allows PHW to provide continuous freshwater production even under realistic outdoor conditions. These results indicate that PHW can effectively tolerate variations in ambient temperature and intermittent solar radiation under certain conditions. Its ability to store and release thermal energy allows PHW to maintain stable and sustained desorption performance, highlighting its strong potential for continuous freshwater production under realistic outdoor conditions.

Changes made:

We have added description of the desorption kinetics at different ambient temperatures (**Page 14 in the revised manuscript**), highlighted in red, as shown below:

“Moreover, PHW exhibited excellent desorption performance and adaptability to different practical application scenarios, including under fluctuating ambient temperatures and simulated high-temperature conditions (Supplementary Figs. 23-24). Notably, when the ambient temperature reaches an extreme high of 42 °C, surpassing the crystallization

point of the incorporated PCM, the PCM loses its ability to effectively regulate and release its stored latent heat. Consequently, under such extreme high-temperature conditions after illumination ceases, both PHW and the control HW continue to desorb, with HW showing a marginally higher final desorption mass. This result highlights a current limitation of the PCM within this extreme high-temperature range. To address this limitation, future applications could involve designing specialized PCM capable of efficiently releasing heat under such extreme conditions for energy management, thus expanding the application range.”

We have already added **Supplementary Figure 23 on page 31 of the revised SI.**

3. Please confirm whether PHW is the sample after loading LiCl. Verify whether PHW in Figure 4f has been loaded with lithium chloride and further confirm the temperature under dark conditions.

Response:

Thank you very much for the reviewer’s question. PHW refers to samples loaded with LiCl to provide hygroscopicity. We understand the reviewer’s curiosity, as our previous manuscript used multiple units and several material names, which may have caused confusion. To improve clarity and consistency, we repeated the relevant experiments and unified the units throughout the manuscript (as noted in our earlier responses). We also re-measured the temperature response under the same conditions using the same samples to ensure overall consistency and transparency.

We conducted a new sorption-desorption experiment and recorded the temperature evolution during desorption, together with infrared thermal imaging. The total desorption duration was 120 min: the first 60 min under simulated sunlight, followed by 60 min in the dark, as shown in Figures R15-17.

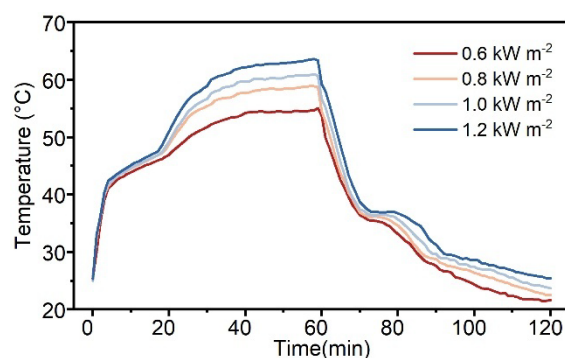


Figure R15. Surface temperature of PHW (with PCM) under different solar intensity (0.6-1.2 kW m⁻²) and then in dark condition within 120 min.

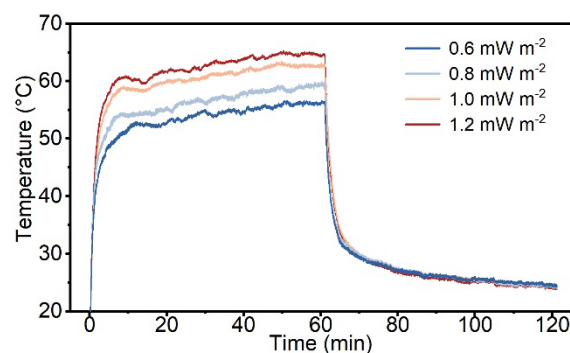


Figure R16. Surface temperature of HW (without PCM) under different solar intensity (0.6–1.2 kW m^{-2}) and then in dark condition within 120 min.

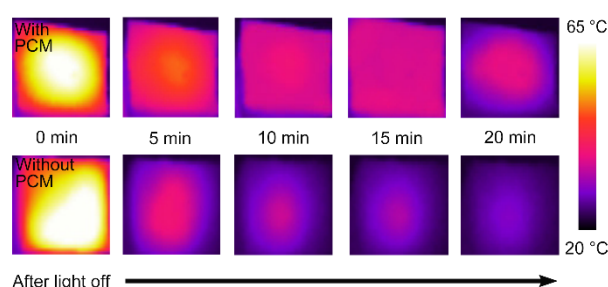


Figure R17. IR images of PHW (with PCM) and HW (without PCM) after the cessation of solar radiation (1.0 kW m^{-2}).

Changes made:

We have already updated and added **Fig. 4d and Fig. 4e on page 12 of the revised manuscript**, as well as **Supplementary Fig. 22 on page 31 of the revised SI**.

We have added a description of the temperatures associated with the desorption of PHW and HW **on page 13 of the revised manuscript**, highlighted in red, as shown below:

“The PHW reached maximum temperatures of 54.64, 58.64, 60.28, and 63.05 °C, respectively, after 60 min of solar illumination at intensities ranging from 0.6 to 1.2 kW m^{-2} , while HW reached maximum temperatures of 56.23, 59.28, 62.69, and 64.48 °C. Notably, PHW exhibits a transient temperature plateau, indicating a rapid phase transition and latent heat energy storage process of PCM.”

“Upon cessation of illumination, the surface temperature of HW rapidly decreased to ambient temperature within 5 min, while the temperature of the PHW dropped more slowly, demonstrating its excellent thermal buffering capability. Specifically, PHW maintained a surface temperature above 35 °C for up to 15 min after the solar simulator was turned off (Figure 4e).”

4. Determine whether PEG (total organic carbon) is present in the water and whether

PEG leakage occurs during the desorption process.

Response:

Thank you very much for your valuable suggestion. We understand the concern regarding possible PCM leakage during desorption. In our design, PEG is immobilized by a hydrogel network and further confined within the wood scaffold, which together maximize retention and interfacial integrity. The leakage-prevention mechanism is multi-scale and synergistic:

1. **Macroscopic confinement (wood scaffold):** The wood sponge has an interconnected, large-pore structure that serves as a rigid framework, providing primary physical support and spatial confinement for the PEG gel composite. This prevents bulk deformation and suppresses macroscopic leakage during phase transition.
2. **Micro/nanoscale spatial limitation (cross-linked PAA network):** A three-dimensional cross-linked poly(acrylic acid) (PAA) network is formed by in situ free-radical polymerization. This network partitions and traps PEG within micro- and nanoscale meshes, creating a physical barrier after melting and reducing PEG mobility through steric hindrance.
3. **Molecular anchoring (hydrogen-bonding network):** Dense hydrogen bonding among cellulose (wood), PAA, and PEG provides strong molecular-level fixation. PEG (ether oxygens and terminal hydroxyl groups) forms hydrogen bonds with PAA carboxyl groups, while PAA chains also hydrogen-bond with hydroxyl groups in the wood matrix, indirectly but firmly anchoring PEG to the stable wood substrate.

Overall, this “macroscopic physical confinement, micro–nano spatial limitation, and molecular anchoring” strategy ensures excellent leakage resistance and structural stability during long-term cycling.

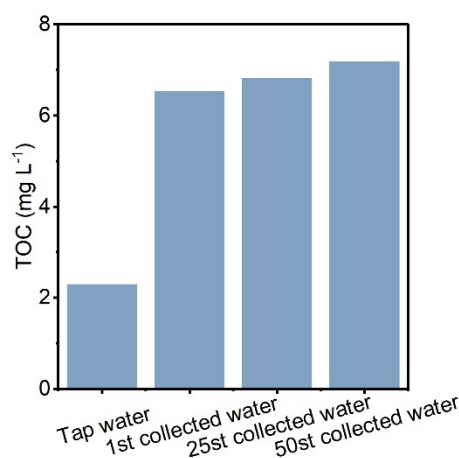


Figure R18. Total Organic Carbon (TOC) concentrations in tap water, the first collected water, 25th collected water, and 50th collected water.

To further clarify potential impacts on water quality, we conducted total organic carbon (TOC) tests on the tap water used during sorption and on the collected water after long-term sorption–desorption cycles (Figure R18). The TOC of laboratory tap water was 2.30 mg L⁻¹. After the 1st cycle, the collected water showed 6.53 mg L⁻¹; after the 25th cycle, 6.82 mg L⁻¹; and after the 50th cycle, 7.18 mg L⁻¹. Although the TOC is slightly higher than that of the tap water, it changes only marginally over repeated cycles, indicating that no significant PEG leakage occurs during long-term operation.

Changes made:

We have already added **Supplementary Fig. 33 on page 35 of the revised SI**.

We have added a description of the organic carbon content in the collected water **on page 14 of the revised manuscript**, highlighted in red, as shown below:

“The safety of collected water with PCM incorporation was assessed by measuring total organic carbon (TOC). The TOC values of the harvested water range from 6.53 to 7.18 mg L⁻¹, indicating no significant PCM leakage during long-term operation (Supplementary Fig. 33).”

5. How stable is the material structure after 20 adsorption-desorption cycles?

Response:

Thank you for raising the concern regarding the structural stability of the material after 20 sorption-desorption cycles. To better assess the material's stability throughout the cycling process, we conducted an additional set of cyclic tests and evaluated the mechanical properties, microscopic morphology, and chemical structure (as assessed by FTIR) of the material before and after 50 sorption-desorption cycles. Each cycle consisted of 12 h at 90% RH, followed by 2 h of desorption, which included 1 h under solar intensity of 1 kW m⁻² and then 1 h in darkness.

Microscopic Structural Stability Evaluation

After 50 sorption-desorption cycles, we observed that the material's microscopic structure remained largely unchanged (Figure R19). The wood sponge matrix, integrated with PCM hydrogel, retained its shape and functional integrity, showing no significant deformation or leakage. Additionally, the sorption zone maintained its vertically aligned, multi-layered ordered structure, which ensures low diffusion resistance and high sorption-desorption kinetics over extended cycles. SEM imaging, along with EDS mapping, confirmed that the hygroscopic salt was uniformly distributed on the matrix surface both before and after cycling, further demonstrating the material's structural stability.

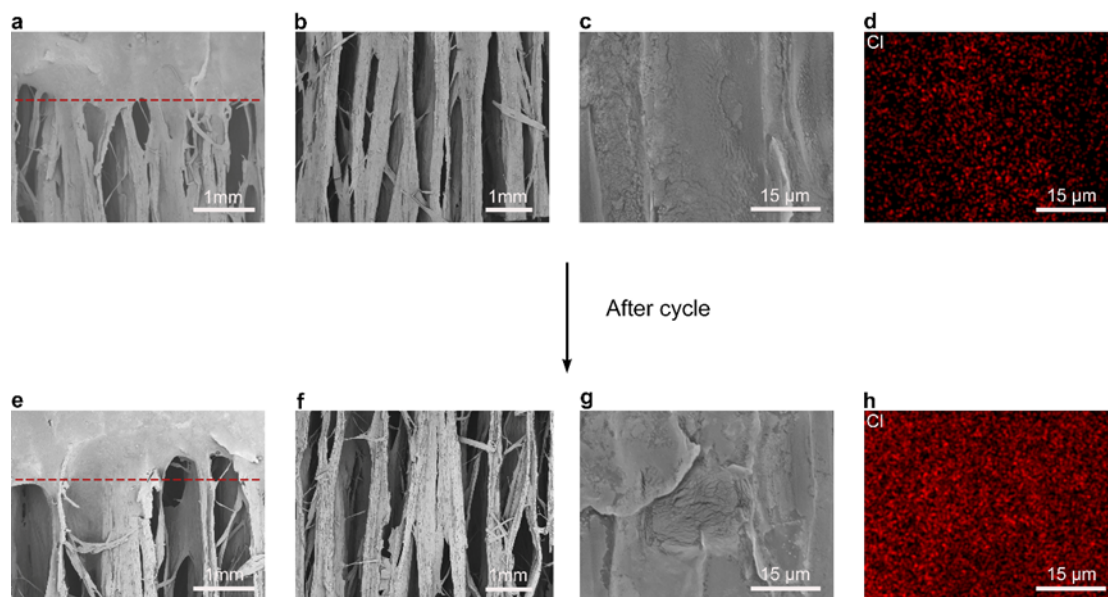


Figure R19. SEM image of the junction area between the upper and lower zones of PHW a) before and e) after the sorption-desorption cycles. The SEM image of the sorption zone of the PHW b) before and f) after the sorption-desorption cycles. High-magnification SEM image of the sorption zone of the PHW c) before and g) after the sorption-desorption cycles. EDS mapping of the sorption zone of the PHW d) before and h) after the sorption-desorption cycles.

Mechanical Performance Stability Evaluation

In terms of mechanical performance, we measured the compression stress-strain response and compressive strength at the 1st, 25th, and 50th sorption-desorption cycles (Figure R20). The compressive strength after the first cycle was 3.95 MPa, which decreased to 3.45 MPa after 25 cycles and 3.37 MPa after 50 cycles. The compressive strength showed only a 14.68% decrease from the first to the 50th cycle, indicating that the sorbent maintains excellent mechanical stability even after long-term cycling. This suggests that the material can support long-term usage without significant loss in mechanical performance.

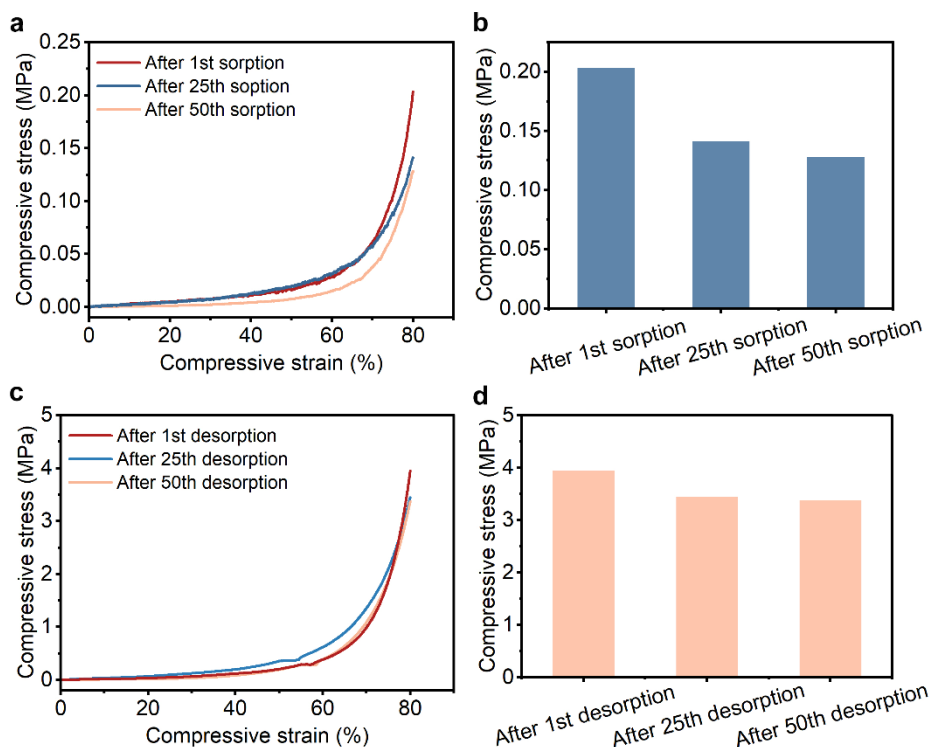


Figure R20. a) Compressive stress–strain curves and b) compressive stress of the sorbent after 1st sorption, 25th sorption and 50th sorption. c) Compressive stress–strain curves and d) compressive stress of the sorbent after 1st desorption, 25th desorption and 50th desorption.

Chemical Structural Stability Evaluation

Additionally, FTIR analysis confirmed the stability of the chemical structure, with no significant changes in the characteristic peak positions or intensities, regardless of whether it was in the lower sorption zone or the upper energy management zone of the PHW after 50 sorption-desorption cycles (Figure R21).

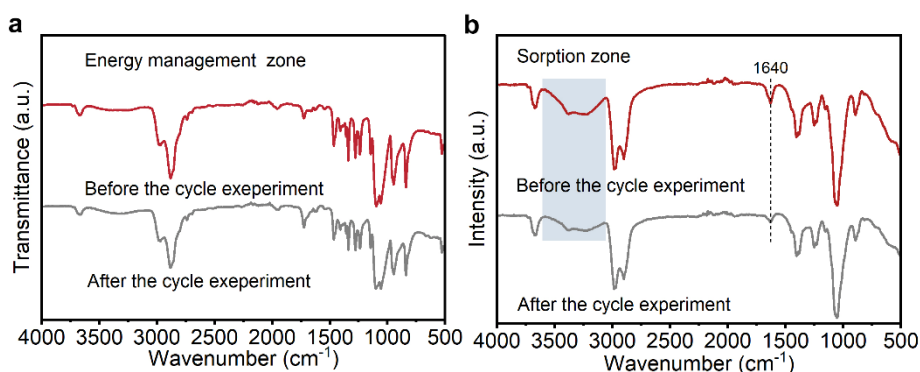


Figure R21. a) FTIR spectra of the energy management zone of the PHW before and after sorption-desorption cycles. b) FTIR spectra of the sorption zone of the PHW before and after sorption-desorption cycles.

Changes made:

We have modified and added the following description **in the revised manuscript (Page 14)**, highlighted in red, as shown below:

“The high stability of water release from PHW is confirmed by 50 sorption–desorption cycles, consisting of 12 h sorption at 90% RH under high humidity, followed by solar-driven release under 1 kW m⁻² irradiation and an additional 1 h release in darkness (Supplementary Figs. 27 and 28). Structural and material characterization further verifies durability, with the macroscopic morphology, microstructure, chemical composition, and mechanical properties remaining well preserved after cycling tests (Supplementary Figs. 29–32)”

We added the **Supplementary Figs. 27-33 in the revised SI (Pages 33-35)**.

6. The manuscript emphasizes the efficient utilization of solar energy; please quantify the solar-to-thermal conversion efficiency.

Response:

The photothermal conversion efficiency (η) refers to the proportion of received light energy converted into heat energy, which is an important parameter for assessing the photothermal performance of the material. It is calculated using the following formula:

$$\eta = \frac{M\Delta H_m}{PS\Delta t}$$

where M (g) is the mass of the energy management zone of the PHW, ΔH_m (J g⁻¹) is the melting enthalpy of the energy management zone of the PHW, P (W m⁻²) is the solar irradiation intensity, S (m²) represents the area of the top surface of the samples, and Δt (s) is the duration of the phase change under the simulated radiation, the relevant data of this are shown in [Table R10](#).

Table R10. Data for photothermal conversion calculations.

	M (g)	ΔH_m (J g ⁻¹)	P (W m ⁻²)	S (m ²)	Δt (s)	η (100%)
PHW	2.68	155.51	1000	0.00051	900	90.80

By calculation, the photothermal conversion efficiency of the energy management zone of the PHW is 90.80 %.

Changes made:

We have added a detailed quantitative energy balance analysis of solar-to-thermal conversion efficiency to **Supplementary Note 4 of the revised SI, page 16**.

On page 13 of the revised manuscript, we have added a description of the photothermal conversion efficiency, highlighted in red, as shown below:

“Notably, PHW exhibits a transient temperature plateau, indicating a rapid phase transition and latent-heat energy storage process of the PCM. The photothermal conversion efficiency of PHW reaches as high as 90.80%. (Supplementary Note 4)”

7. In outdoor experiments, it is recommended to supplement daily water production data and compare it with other studies.

Response:

Thank you very much for the valuable suggestion. The actual outdoor water production performance is indeed of great significance when evaluating the practical application ability of sorbents. We selected a variety of sorbents, including hydrogel-based, aerogel-based, foam-based sorbents, as well as MOF-based materials. Due to differences in sorbent properties, dimensions, thicknesses, the operational modes and durations during testing, as well as local environmental factors such as temperature, humidity, and solar intensity, the actual daily water production varies. The specific comparison results can be found in [Table R11](#).

To ensure the reliability and rigor of our outdoor water production values, in addition to the single-day water production values measured in Harbin, we also conducted several days of continuous outdoor experiments in Xiamen, from which we obtained an average value. When comparing our sorbent's daily water production capacity with those of state-of-the-art sorbents, our materials remain in the top tier.

Table R11. Daily water production of various sorbents in outdoor experiments.

Sorbent	Operation mode	Daily water production
MPH ¹⁷	From 20:00 to 8:00 the next day for sorption, from 8:00 to 14:00 for desorption	1.04 $\text{g}_{\text{water}} \text{g}_{\text{sorbent}}^{-1} \text{day}^{-1}$
DAW-PM ¹⁸	24-h cyclic test	0.32 $\text{g}_{\text{water}} \text{g}_{\text{sorbent}}^{-1} \text{day}^{-1}$
PDMAPS/CNT/LiCl ¹⁹	From 6:00 pm to 7:00 am the next day for sorption, from 9:00 a.m. to 3:00 p.m for desorption.	0.62 $\text{L}_{\text{water}} \text{kg}_{\text{sorbent}}^{-1} \text{day}^{-1}$
PAETA-Ac/CNT hydrogel ²⁰	From 7:00 p.m. to 6:00 a.m. the next day for sorption, from 10:30 a.m. to 2:00 p.m.	0.53 $\text{L}_{\text{water}} \text{kg}_{\text{sorbent}}^{-1} \text{day}^{-1}$
PAM-CNT-CaCl ₂ hydrogel ²¹	From 5 p.m. to 10 a.m. the next day for sorption, from 13:20 p.m. to 15:50 p.m. for desorption	0.57 $\text{L}_{\text{water}} \text{kg}_{\text{sorbent}}^{-1} \text{day}^{-1}$
Our work	From 0:00 a.m. to 6:00 a.m. for sorption, followed by desorption from 6:00 a.m. to 7:00 a.m. Afterward, three cycles of sorption for 2.5 h and desorption for 1 h each. (Harbin, China)	1.72 $\text{L}_{\text{water}} \text{kg}_{\text{sorbent}}^{-1} \text{day}^{-1}$
	From 0:00 a.m. to 8:00 a.m. for sorption, followed by desorption from 8:00 a.m. to 10:00 a.m. Afterward, two cycles of sorption for 2.5 h and desorption for 1 h each. (Xiamen, China)	0.96-1.37 $\text{L}_{\text{water}} \text{kg}_{\text{sorbent}}^{-1} \text{day}^{-1}$

[1] Wang, X. et al. High solar-thermal conversion aerogel for efficient atmospheric water harvesting. *Small* **20**, 2307416 (2023).

- [2] Liang, J. et al. Synergistic structural alignment and interfacial switching enable fast and low-energy water release in solar-driven atmospheric water harvesting. *Adv. Funct. Mater.*, e18960 (2025).
- [3] Aleid, S. et al. Salting-in effect of zwitterionic polymer hydrogel facilitates atmospheric water harvesting. *ACS Materials Lett.* **4**, 511-520 (2022).
- [4] Wu, M. et al. Metal- and halide-free, solid-state polymeric water vapor sorbents for efficient water-sorption-driven cooling and atmospheric water harvesting. *Mater. Horiz.* **8**, 1518–1527 (2021).
- [5] Li, R. et al. Hybrid hydrogel with high water vapor harvesting capacity for deployable solar-driven atmospheric water generator. *Environ. Sci. Technol.* **52**, 11367–11377 (2018).

Changes made:

We have added **Supplementary Table 7 on page 12 of the revised SI**, summarizing a comparison of the daily water production of our sorbent with that of other sorbents, as well as the operation modes.

Additionally, we have included **Figure 5h in the revised manuscript (Page 15)** to clearly highlight the exceptional daily water production performance of our sorbent.

8. Explain the interaction mechanism between PCM and the wood sponge interface, and whether it affects the stability of latent heat storage.

Response:

We appreciate your insightful comment. To elucidate the interaction mechanism between the PCM gel and the wood sponge interface, we conducted Fourier-transform infrared (FTIR) spectroscopy and X-ray diffraction (XRD) analyses on the PCM gel, wood sponge, and the upper energy management zone of the PHW (wood sponge/PCM gel interface). Furthermore, differential scanning calorimetry (DSC) thermal cycling tests were performed on both the PCM gel and the upper energy management zone of the PHW (wood sponge/PCM gel) to assess whether these interactions influence the stability of latent heat storage. The following two sections provide a detailed discussion of these two aspects.

Interaction mechanism

The formation and stabilization mechanism of the upper energy management zone of the PHW (wood sponge/PCM gel) can be precisely attributed to the specific molecular-level interactions between the wood sponge and PCM gel, as evidenced by the FTIR spectra (Figure R22a). The spectral evidence indicates a synergistic integration rather than a simple physical mixture. The most significant change occurs in the -OH stretching region^{1,2}. The cellulose hydroxyl groups³ in the wood sponge compete with the polyethylene glycol hydroxyl groups and the carboxylic acid groups of acrylic acid for intermolecular bonding, forming new, strong and wide hydrogen bonds, thereby forming a stable and robust interface structure^{4,5}.

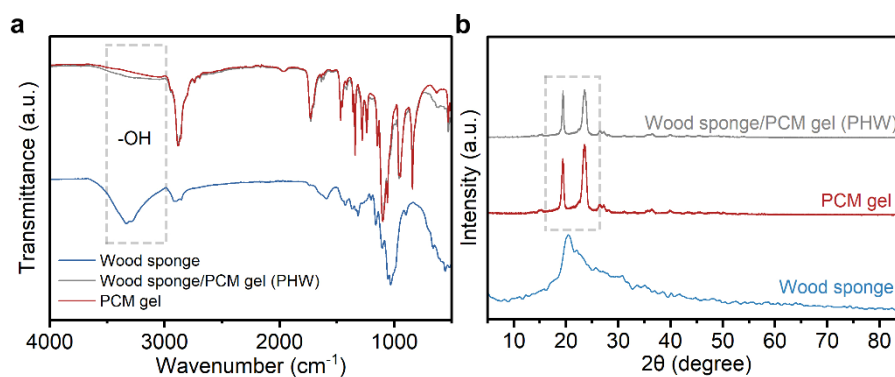


Figure R22. a) FTIR spectra of wood sponge, PCM gel and the upper energy management zone of the PHW (wood sponge/PCM gel). b) XRD curves of wood sponge, PCM gel and the upper energy management zone of the PHW (wood sponge/PCM gel).

To further substantiate this interaction, the XRD patterns (Figure R22b) provide additional structural evidence. The PCM gel exhibits two distinct diffraction peaks at 19.44 ° and 23.53 °, which are typical characteristics of the short-range ordered or microcrystalline regions formed by the internal PEG chain segments within the cross-linked network⁶. In the upper energy management zone of the PHW (wood sponge/PCM gel), these two characteristic peaks shift to 19.54 ° and 23.65 ° respectively^{7,8}. This although minor but systematic shift of the peak positions towards higher angles implies a decrease in the interplanar spacing of the corresponding crystal planes. This strongly suggests that there is a molecular-level close interaction between the cellulose in the wood sponge and the PCM gel. The most likely mechanism is that a large number of -OH on the surface of the wood cellulose form strong hydrogen bonds with the PEG ether bonds (-O-) in the PCM gel and -COOH of acrylic acid⁹. This hydrogen bonding interaction shortens the stacking distance of the molecular chain segments or microcrystalline regions within the gel, thereby causing an increase in the diffraction angle.

Meanwhile, due to the cellulose present in the wood sponge, which has a nanoscale confinement effect on the gel structure, the area of the diffraction peaks decreases and the diffraction intensity reduces. The formation of a strong coupling interface between the gel and the wood framework, and the realization of highly uniform composites and stable structures provide direct evidence for the microscopic foundation of the excellent leak-proof performance and thermal cycling stability of the composite materials.

Stability of latent heat storage

The DSC cycle curves we obtained provide direct evidence for the above mechanism and its positive impact on the stability of latent heat storage. Compared to PCM gel, the peak shapes of the phase change and the changes in latent heat of the composite materials directly reflect the effects of confinement and interfacial nucleation (Figure R23). The most crucial point is that in multiple thermal cycling tests, the wood sponge/PCM gel demonstrated far superior stability compared to PCM gel: the decay rates of their melting enthalpy and crystallization enthalpy were as high as 2.51% and 2.76%, respectively, and the DSC curves of continuous cycling were highly overlapping, with the peak shapes and

positions remaining basically unchanged (Table R12). This fully proves that the physical confinement provided by the wood sponge and the chemical bonding jointly construct a robust and durable encapsulation system. It not only firmly fixes the PCM, resisting the volume change stress during the long-term phase change process, but also effectively reduces the thermal performance degradation of the materials due to volatilization, migration, or phase separation, thereby fundamentally ensuring the long-term cycle stability of the latent heat storage ability.

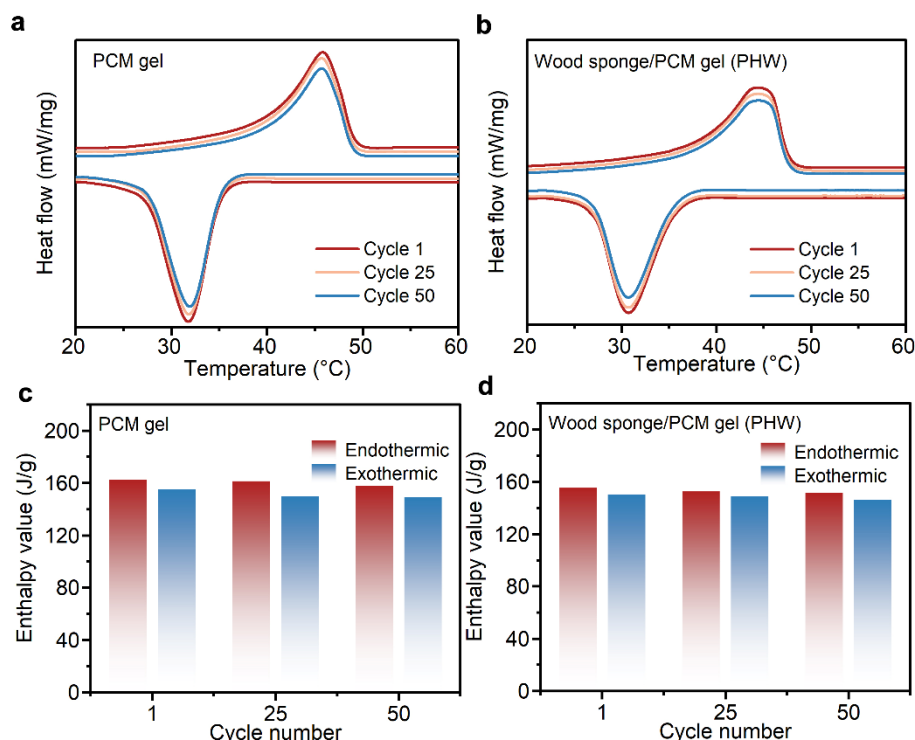


Figure R23. DSC curves of the PCM gel a) and the energy management zone of the PHW (wood sponge/PCM gel) b) measured at cycle 1, cycle 25, and cycle 50, showing the evolution of melting and crystallization behavior under repeated thermal cycling. Endothermic and exothermic enthalpy values of the PCM gel c) and the energy management zone of the PHW (wood sponge/PCM gel) d) over 50 thermal cycles.

Table R12. Evolution of phase change enthalpy of the PCM gel and the energy management zone of the PHW (wood sponge/PCM gel).

Cycle number	Phase change enthalpy of the PCM gel (J g^{-1})		Phase change enthalpy of the energy management zone of the PHW (wood sponge/PCM gel) (J g^{-1})	
	ΔH_m	ΔH_c	ΔH_m	ΔH_c
Cycle 1	162.65	155.10	155.51	150.42
Cycle 25	161.46	150.04	152.66	148.92
Cycle 50	158.17	149.21	151.60	146.27

In conclusion, the FTIR spectra clearly demonstrate that the wood sponge functions as a structural scaffold that tightly binds to the PCM gel through strong hydrogen bond

interactions. This synergistic interfacial bonding effectively locks the PCM gel within the hierarchical channels of the wood sponge, ensuring superior shape stability and completely preventing leakage during repeated solid–liquid phase transitions. This conclusion is further supported by the XRD patterns, where the systematic peak shifts toward higher angles and the reduced diffraction intensity indicate constrained chain packing and strengthened interfacial coupling within the composite, providing structural evidence for the robust integration between the PCM gel and the wood framework. Moreover, the 50-cycle DSC results confirm that this interfacial interaction does not compromise the long-term stability of the latent-heat storage capacity, further validating the robustness and reliability of the PHW sorbent during extended cycling.

- [1] Liu, Y. et al. A Cellulose Salt Gel with Mechanical Transformation and Thermal Control. *Adv. Funct. Mater.* **34**, 2400203 (2024)
- [2] Zhang, X. et al. A flexible, thermal-insulating, and fire-resistant bagasse-derived cellulose aerogel prepared via a refrigerator freezing combined ambient pressure drying technique. *Chem. Eng. J.* **498**, 155466 (2024).
- [3] Liu, C. et al. 3D printing of phase change material-based Pickering emulsion gel for solar-thermal-electric conversion. *Chem. Eng. J.* **499**, 155940 (2024).
- [4] Zhao, X. et al. Phase change mediated mechanically transformative dynamic gel for intelligent control of versatile devices. *Mater. Horiz.* **8**, 1230–1241 (2021).
- [5] Zhao, D. et al. A Stiffness-Switchable, Biomimetic Smart Material Enabled by Supramolecular Reconfiguration. *Adv. Mater.* **34**, 2107857 (2024).
- [6] Song, M. et al. Thermally induced flexible phase change hydrogels for solar thermal storage and human thermal management. *Chem. Eng. J.* **464**, 142682 (2023).

Changes made:

We have added **Supplementary Figures 14–15 (Pages 27-28) and Supplementary Table 4 in the revised SI (Page 9)**, along with corresponding descriptions in the main text to explain the interaction mechanism at the PCM gel–wood sponge interface and to clarify whether this interaction affects the stability of latent-heat storage.

We have newly added the following text, highlighted in red **on page 8 of the revised manuscript**:

“In addition, the strong hydrogen bond interactions identified by FTIR, together with the constrained chain packing evidenced by the systematic peak shifts in the XRD patterns, demonstrate that the PCM hydrogel forms a stable and robust interfacial structure with the wood sponge matrix. This integrated chemical and structural coupling prevents deformation or leakage during phase transitions and preserves the long-term effectiveness of latent heat storage (Supplementary Figs. 14-15 and Table 4). Furthermore, owing to its flexible hydrogel impregnation and the inherent structural adaptability of the wood sponge, the PHW can be readily fabricated into diverse shapes and geometries, greatly enhancing its suitability for various practical application scenarios (Supplementary Fig. 16).”

9. Clarify the distribution of energy between latent heat storage and desorption in PCM

during photothermal conversion.

Response:

We greatly appreciate your insightful comment, which allows us to clarify the relationship between the latent heat storage and the desorption in PCM during photothermal conversion. A detailed explanation of the energy distribution is provided below.

Energy for desorption:

During the 60-min continuous solar irradiation, the sorbent remained in a solar-driven desorption state, releasing a total water of:

$$M_{\text{des,total}} = 2.8029 \text{ g}$$

corresponding to energy for desorption of:

$$Q_{\text{des,total}} = M_{\text{des,total}} \times \Delta H_v = 2.8029 \times 1540 = 4316.47 \text{ J}$$

Where $M_{\text{des,tatol}}$ (g) is the total mass of water desorbed during this process, ΔH_v (J g^{-1}) is the desorption enthalpy of the water in the sorption zone of the PHW. This value was determined through DSC thermal flow tests, where the heat flow over time was integrated, as shown in [Figure R1](#). $Q_{\text{des,total}}$ (J) is the total energy required for desorbing the water from the PHW.

Latent-heat storage of the PCM:

The PCM stores latent heat during the melting process, as it undergoes a phase transition from solid to liquid. The amount of latent heat stored in the PCM is calculated as:

$$Q_{\text{latent}} = M \times \Delta H_m = 2.68 \times 155.51 = 416.77 \text{ J}$$

where M (g) is the mass of the energy management zone of the PHW, ΔH_m (J g^{-1}) is the melting enthalpy of the energy management zone of the PHW

Energy distribution:

The total energy used by the PHW is the sum of the energy involved in desorption and the latent heat stored in the PCM:

$$Q_{\text{total}} = Q_{\text{des,total}} + Q_{\text{latent}}$$

Where Q_{total} (J) is the total energy used by the PHW, $Q_{\text{des,total}}$ (J) is the total energy required for desorbing the water from the PHW, Q_{latent} (J) is the latent heat stored in the PHW.

The percentage of the latent heat stored in the PCM of the PHW is calculated as:

$$\text{Energy percentage stored in PCM} = \frac{Q_{\text{latent}}}{Q_{\text{total}}} \times 100\%$$

Thus, the latent heat stored in the PCM accounts for 8.81% of the total energy used by the PHW.

Changes made:

We have added energy distribution between latent-heat storage and desorption during photothermal conversion to **Supplementary Note 6 of the revised SI, pages 17-18.**

We have newly added the following text, highlighted in red **on page 13 of the revised manuscript:**

“The energy for latent heat storage in the PHW accounts for only 8.81% of the total energy required during the desorption process under a solar radiation intensity of 1 kW m^{-2} (Supplementary Notes 5-6).”

Thank you again for all your insightful comments and suggestions, which have greatly helped us improve the manuscript!

Response to the Reviewer

Reviewer #1

The manuscript can be accepted now for publication.

Response:

We sincerely thank you for your recognition that we have addressed all of your comments, and for your recommendation for publication. We hope that our work will be helpful to researchers in related fields.

Reviewer #3

The authors conducted lots of supplementary experiments and made a comprehensive revision to the manuscript. Two small questions need to be clarified before publication.

Response:

We sincerely thank you for your constructive feedback and valuable suggestions. In response to your comments, we have carefully addressed the two questions you raised, incorporating the necessary revisions to improve the clarity and quality of the manuscript. We hope these changes meet your expectations and contribute to the overall enhancement of the paper.

1. How about the repetitiveness of the water uptake tests. The error bars in Fig. R10, Fig. R18 and Fig. R20 should be included.

Response:

We sincerely appreciate your valuable suggestion. In response to your comments, in response to your comments, we previously conducted repeated water uptake tests, which showed excellent stability. Additionally, we have repeated the experiments for the data presented in Fig. R10, Fig. R18, and Fig. R20, and have included the corresponding error bars as requested.

Water Uptake Tests

The water sorption stability of PHW was evaluated over 20 sorption–desorption cycles from 20 to 80% RH, showing no significant change in equilibrium sorption capacity and indicating stable and reversible water uptake (Figure R1).

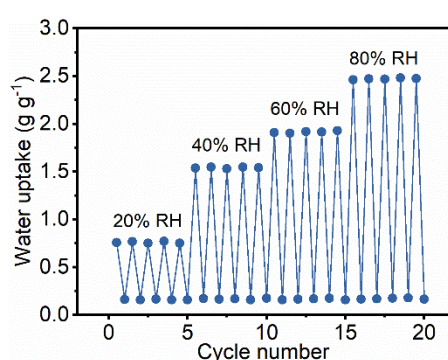


Figure R1. Twenty water sorption cycling tests of sorption zone in the PHW from 20 to 80% RH.

Total Organic Carbon Tests

To guarantee experimental rigor and the accuracy of the data, total organic carbon (TOC) tests were conducted on the tap water used for sorption and on the water collected after long-term sorption–desorption cycles, with each measurement repeated

three times. The TOC of the laboratory tap water was $2.22 \pm 0.18 \text{ mg L}^{-1}$. After the first cycle, the collected water showed $6.49 \pm 0.79 \text{ mg L}^{-1}$; after the 25th cycle, $7.56 \pm 0.81 \text{ mg L}^{-1}$; and after the 50th cycle, $8.23 \pm 1.19 \text{ mg L}^{-1}$. Although slightly higher than that of the tap water, TOC remained largely unchanged over repeated cycles, indicating negligible PEG leakage during long-term operation.

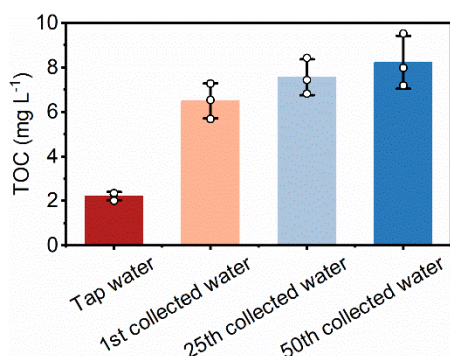


Figure R2. Total Organic Carbon (TOC) concentrations in tap water, the first collected water, 25th collected water, and 50th collected water. All error bars represent the mean $\pm$ standard deviation (SD) from $n = 3$ measurements.

Compression Mechanics Test

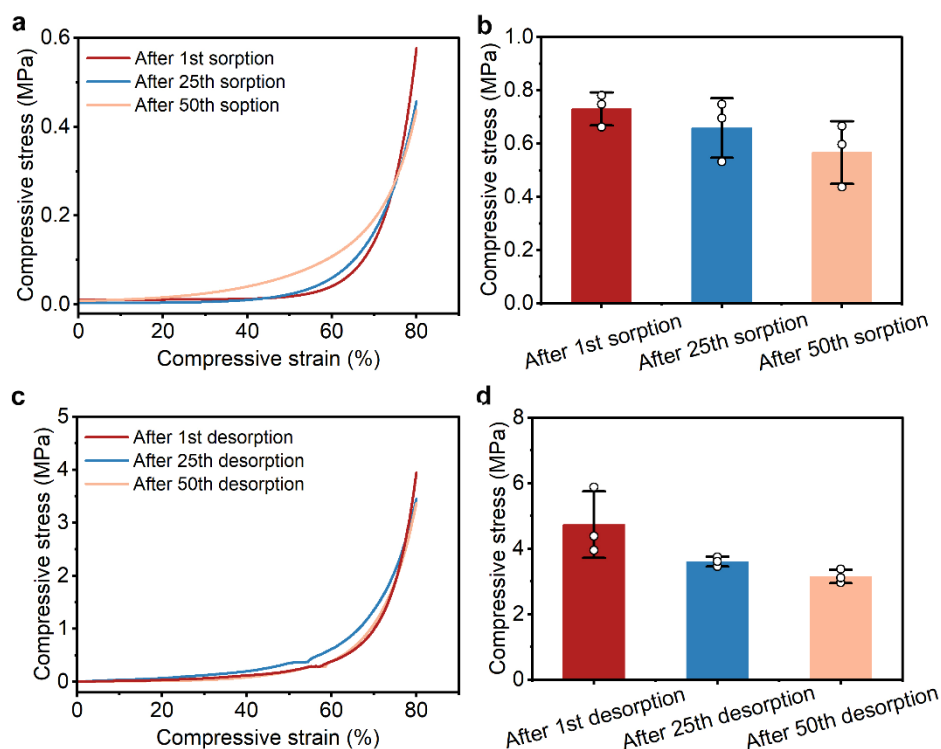


Figure R3. a) Compressive stress–strain curves and b) compressive stress of the sorbent after 1st sorption, 25th sorption and 50th sorption. c) Compressive stress–strain curves and d) compressive stress of the sorbent after 1st desorption, 25th desorption and 50th desorption. All error bars represent the mean $\pm$ standard deviation (SD) from $n = 3$ measurements.

We repeated the measurements of the compression stress–strain response and compressive strength using samples retained after long-term sorption–desorption cycling tests, with evaluations conducted at the 1st, 25th, and 50th cycles and each measurement repeated three times (Figure R3). The compressive strength after the first cycle was 4.74 ± 1.02 MPa, which decreased to 3.60 ± 0.15 MPa after 25 cycles and 3.15 ± 0.21 MPa after 50 cycles. Even after 50 cycles, the compressive strength remained higher than 65% of its initial value, demonstrating the excellent mechanical stability of the sorbent over prolonged cycling. This suggests that the material can support long-term use without significant loss of mechanical performance.

Changes made:

We have revised **Supplementary Fig. 32 (Page 34)** and **Fig. 34 (Page 35)** in the revised Supplementary Information.

We have modified the following description in the revised manuscript (Page 14), highlighted in red, as shown below:

“The TOC values of the water harvested during the cycles range from 6.49 ± 0.79 to 8.23 ± 1.19 mg L⁻¹, indicating no significant PCM leakage during long-term operation (Supplementary Fig. 34).”

2. PEG as the PCM might leak during desorption process as raised by the previous reviewer, the TOC in Figure R18 seems a significant increase in the collected water. The reviewer suggests a simple test with filter paper, to check the state of the composite when heating.

Response:

We sincerely appreciate your valuable suggestion. In the TOC test shown in Figure R18, although the TOC content of the collected water slightly increased compared to tap water during the long-term cycling process, it remained below 10 mg L⁻¹. To address your concern, we conducted an additional leakage test using filter paper (Figure R4). The PHW composite was placed in a vacuum oven at 70 °C for 24 h to evaluate leakage resistance. As shown in Figure R4, throughout the heating process, the PHW maintained its original macroscopic shape without any observable leakage, demonstrating the composite's excellent thermal stability during heating.

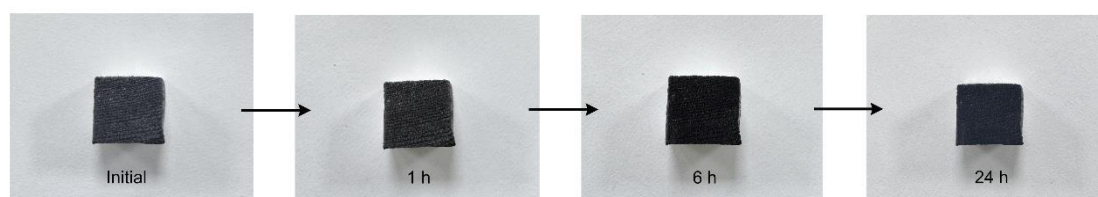


Figure R4. Evaluation of leakproof performance of PHW at 70 °C.

Changes made:

We have already added **Supplementary Fig. 16 on page 28 of the revised SI.**

We have added a description of the leakage resistance test of the PHW **on page 7 of the revised manuscript**, highlighted in red, as shown below:

“This integrated chemical and structural coupling prevent deformation or leakage during phase transitions and preserves the long-term effectiveness of latent heat storage (Supplementary Figs. 15-16 and Table 4).”

Thank you again for all your valuable comments. We hope that we have addressed all of them satisfactorily.

Response to the Reviewer

Reviewer #3

The authors have appropriately addressed the comments. Therefore, the manuscript is suitable for publication.

Response:

We sincerely thank you for your positive evaluation of our revision and for recommending our manuscript for publication. We greatly appreciate your time and valuable comments, and we hope that our work will be of value to researchers in related fields.

This manuscript addresses two core bottlenecks of AWH—slow adsorption due to diffusion resistance and desorption interruptions caused by intermittent solar energy—through material design (wood-derived sponge) and a PAA-CNT-PEG photothermal phase-change hydrogel. However, the manuscript exhibits significant shortcomings in the depth of its mechanism explanation, quantitative comparison of key performance metrics, and rigorous analysis of practical applications. It requires supplementation with critical data and analysis to substantiate its innovation and potential impact.

- 1、 Supplement data on the adsorption and desorption performance variations of materials under different ambient temperatures to enhance their practical application scenarios.
- 2、 Explain the suitability of the PCM layer under high-temperature environmental conditions during summer.
- 3、 Please confirm whether PHW is the sample after loading LiCl. Verify whether PHW in Figure 4f has been loaded with lithium chloride and further confirm the temperature under dark conditions.
- 4、 Determine whether PEG (total organic carbon) is present in the water and whether PEG leakage occurs during the desorption process.
- 5、 How stable is the material structure after 20 adsorption-desorption cycles?
- 6、 The manuscript emphasizes the efficient utilization of solar energy; please quantify the solar-to-thermal conversion efficiency.
- 7、 In outdoor experiments, it is recommended to supplement daily water production data and compare it with other studies.
- 8、 Explain the interaction mechanism between PCM and the wood sponge interface, and whether it affects the stability of latent heat storage.
- 9、 Clarify the distribution of energy between latent heat storage and desorption in PCM during photothermal conversion.