

Towards Cleaner, Greener Future: Energy Efficient Sustainable Hydrogel Composites for Solar Driven Photothermal Water Evaporation and Purification

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Abstract: This comprehensive review presents hydrogel-based water evaporators for solar desalination and purification, focusing on ecological sustainability and mechanism-driven design. Unlike conventional Interfacial Solar-Driven Evaporation (ISDE) systems that prioritize photothermal conversion efficiency, this work emphasizes fundamental thermodynamic and lifecycle considerations for long-term sustainability. We propose a mechanism-driven framework using ecological hydrogel composites derived from renewable, biodegradable, and non-toxic precursors like cellulose, alginate, and starch. A key aspect is regulating water states within hydrogel matrices, particularly forming "intermediate water" that reduces vaporization enthalpy and enhances evaporation rates beyond bulk water limits. We correlate material properties like porosity, thermal conductivity, and optical absorption with thermodynamic parameters including latent heat, temperature gradient, and mass flux. The review also covers molecular hydration engineering, dynamic interfacial effects, and multifunctional systems using waste-derived photothermal materials like biochar. Challenges like fouling, salt accumulation, and scalability are evaluated. By bridging molecular insights with system-scale performance, this work shifts from material-centric to mechanism-oriented design, providing a sustainable roadmap for next-generation desalination technologies that minimize environmental risks.

Key words: Solar-Driven Desalination; Photothermal Evaporation; Biodegradable Polymers; Environmental Sustainability; Carbonaceous Materials; Water Purification

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1. Introduction

Water is an irreplaceable and finite resource pivotal for sustaining life on Earth^{[1][2]}. Drinking water faces an urgent and escalating crisis due to a multitude of pollution sources. **Figure 1(A)** displays the number of water scarcity publications in each year from 1996 to 2016, while the results illustrated in **Figure 1(B)** show that relative efforts devoted to specific knowledge domains across different continents demonstrate a certain degree of consistency, ranging from industrial effluents and agricultural runoff to microplastics and pharmaceutical impurities, pose severe threats to ecosystems and human

health^[4]. Industrial processes, factories, and manufacturing facilities discharge hazardous chemicals^[5] heavy metals^[6], and pollutants indirectly into water bodies, resulting in persistent environmental damage. Harmful algal blooms are made worse by the widespread use of pesticides and fertilizers in agriculture, which causes runoff to carry a range of toxic materials into rivers and lakes^{[7][8]}. The widespread presence of microplastics in water bodies^{[9][10]} originating from sources such as synthetic textiles and the breakdown of larger plastic items, has emerged as a critical concern in aquatic ecosystems^[11]. Therefore, improper disposal of pharmaceuticals and personal care products, coupled with their release through sewage systems, introduces a diverse array of pharmaceutical compounds into waterways^[12]. This poses potential health risks and environmental consequences, as these substances may resist removal by conventional water treatment processes^[13]. The ensuing contamination disrupts aquatic food chains, harms sensitive species, and degrades habitats, including wetlands, lakes, and rivers^[14]. **Figures 1(C)-a** and **1(C)-b** show the top 15 sectors

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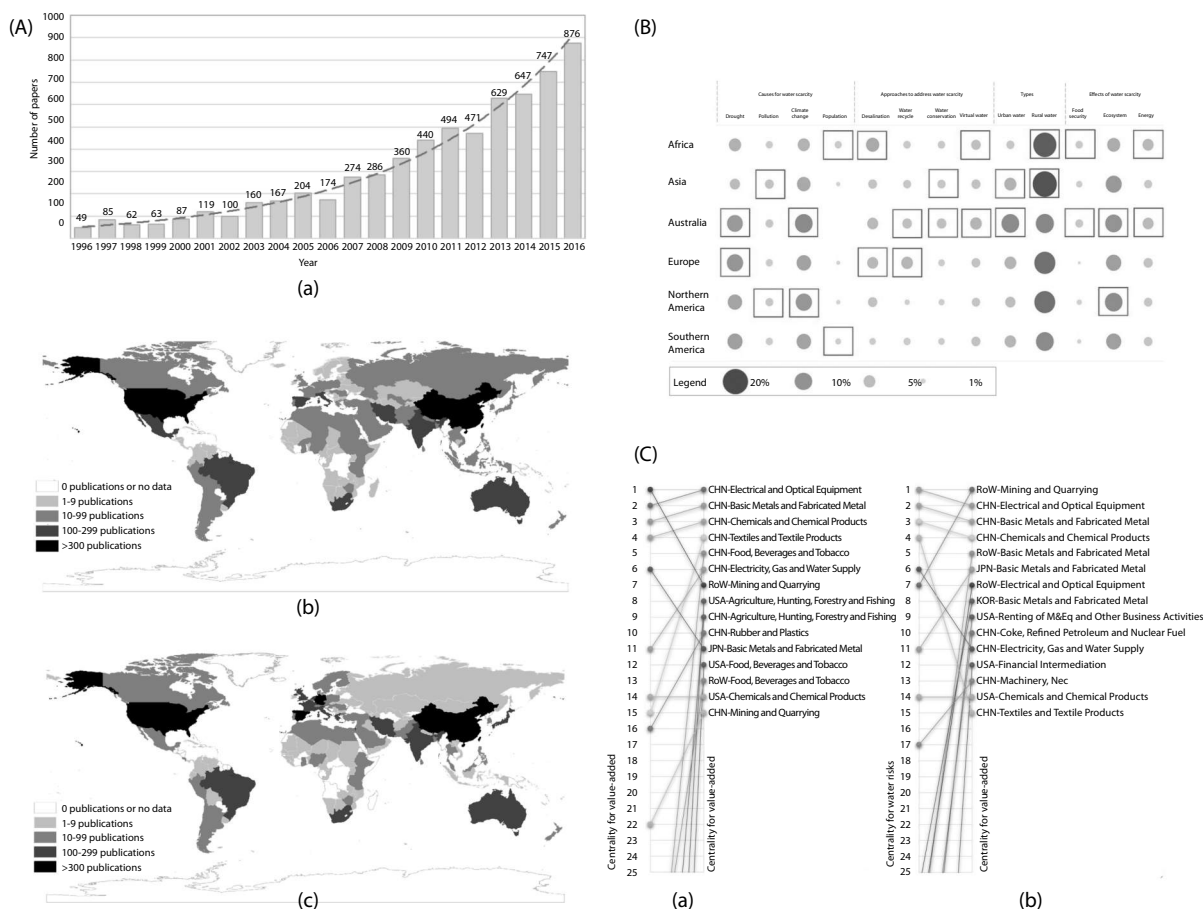


Fig. 1. (A) Bibliometric analysis of global scientific publications. (a) Annual growth trend in the number of published papers from 1996 to 2016. (b) and (c) Global geographical distribution maps illustrating publication outputs by country, color-coded by volume (ranging from 0 to over 300 publications). (B) Ratios of publications in specified research domains to total number of publications in different continents. Red boxes denote two continents with the highest ratios in one research domain. *Reproduced with permission from Springer Nature*^[3]; (C) Changes in rankings of top sectors with respect to two measures of centrality. (a) Top 15 sectors with highest centrality for transmission of water scarcity risks; (b) Top 15 sectors with highest centrality for transmission of value added. *Reproduced with permission from the Taylor & Francis*^[15].

with the highest betweenness centrality in transmitting foreign water scarcity risks and the highest centrality in the transmission of value added, respectively.

Furthermore, the repercussions extend beyond aquatic ecosystems, impacting interconnected terrestrial habitats. For humans, the consequences are significant. Consuming contaminated water not only leads to waterborne diseases but also poses a grave threat to public health, especially in regions lacking adequate water treatment infrastructure^[16]. Persistent exposure to contaminants like heavy metals or endocrine-disrupting compounds can result in enduring health issues, including developmental disorders and specific types of cancer^[17]. The expansion of agriculture and the agro-industrial sector, driven by the imperative to meet the needs of the growing population, exacerbates the strain on water resources. This, in turn, exerts mounting pressure on already fragile ecosystems, depleting water reserves^[18]. The urgency of addressing global water scarcity is underlined by this heightened demand^[19-20]. In response to these challenges, effective water management strategies and advanced treatment methods become imperative. Implementing sustainable water treatment practices is crucial to mitigate the impact of pollution and ensure a stable and secure water supply for both human and environmental needs. The development and implementation of innovative water treatment technologies play a pivotal role in addressing the complexities of contemporary water management.

Traditional water treatment techniques have played a pivotal role in ensuring the safety of water for consumption. However, they come with inherent advantages and disadvantages^[21-22]. Conventional methods, such as chlorination and filtration, often necessitate the use of chemicals, energy, and maintenance, potentially leading to the production of harmful by-products^[23]. The primary, secondary, and tertiary water treatment practices have a proven track record of effectively eliminating pathogens, bacteria, and suspended particles; however, their drawbacks merit careful consideration^[24-25]. The reliance on chemical additives, notably in methods like chlorination, introduces health risks and the generation of disinfection by-products, such as trihalomethanes (THMs)^[26], through reactions with organic matter in water^[27]. While these methods are well-established and widely employed, particularly forming the backbone of municipal water treatment systems, there is a growing recognition of the need for more sustainable alternatives. Many traditional methods incur significant energy consumption in processes like pumping, filtration, and chemical dosing, contributing to greenhouse gas emissions and operational costs^[28-29]. The physical infrastructure of traditional water treatment, encompassing pipelines and treatment plants, requires ongoing maintenance, repair, and replacement to function efficiently^[30]. Additionally, the treatment process itself can inadvertently lead to the formation of harmful by-products, exemplified by the poten-

tial carcinogenicity and health risks associated with chlorinated organic compounds resulting from chlorine use^[31]. In light of these challenges, there is a growing recognition of the need for more sustainable and efficient water treatment methods. Applying advanced techniques, such as photothermal desalination, offers a promising avenue for addressing the limitations of traditional approaches. This method not only holds the potential to enhance efficiency but also to minimize environmental impact and improve overall water treatment processes.

The emergence of innovative approaches, notably solar-based water evaporation, marks a significant advancement in addressing water treatment challenges. Utilizing the sun's energy, this modern approach comes with several benefits^[32]. Solar-based evaporation is inherently sustainable, relying on renewable energy, and proves cost-effective^[33]. Notably, it diminishes the chemical additives, thereby mitigating the risk of harmful by-products. However, the realization of its full potential hinges on the selection of suitable materials. Solar-based water evaporation is profoundly sustainable, drawing upon the virtually boundless energy supplied by the sun. This reliance on renewable energy not only diminishes dependence on fossil fuels but also curtails the carbon footprint associated with energy consumption, aligning with global sustainability objectives^[34]. In addition to its cost-effectiveness, this innovative method offers an environmentally friendly solution, reducing dependence on electricity for conventional treatment processes. This renders it an economical choice, particularly in regions blessed with abundant sunlight^[35]. An inherent advantage of solar-based evaporation is the reduction in reliance on chemical additives, representing a paradigm shift from conventional treatment methods. Reducing the use of chemicals not only preserves resources but also diminishes the risk of generating harmful by-products, thereby improving water safety. Subsequently, the method exhibits substantially reduced environmental impact compared to conventional treatments, resulting in lower energy consumption, fewer emissions, and diminished chemical waste, aligning with the eco-friendly application of biodegradable biopolymer hydrogels^[36] and environmentally responsible practices^[37]. The effectiveness of solar-based water evaporation hinges crucially on the careful choice of materials that can absorb and convert solar energy into heat with high efficiency. Such as Acrylamide^[38], Sodium Alginate^[39-41], PVA, and Chitosan/PVA^[42-47]. The property of the polymer is imperative for maximizing the rate of water evaporation. Nonetheless, materials with elevated thermal conductivity, such as Coconut Shell Biochar^[48], Rice Husk Biochar^[49-50] and Carbon Nanotubes^[51-54], facilitate rapid dispersion and retention of heat^[55]. However, these materials ensure uniform water heating, leading to efficient and consistent evaporation. The selected materials must demonstrate durability, capable of enduring prolonged exposure to sunlight and environmental conditions without substantial degradation. For instance, Chitosan/PVA combined with carbon ink has emerged as a highly promising material, demonstrating the highest evaporation rate at $4.37 \text{ kg m}^{-2} \text{ h}^{-1}$ and an exceptional efficiency of 98.2%^[52]. Aligned with the cumulative focus on sustainable practices, these materials should ideally be environmentally friendly, non-toxic, and easily available^[56-57].

In the world of improving water treatment, ecological hydrogel composites play a key role. In order to attain effective solar-driven water evaporation with a smaller environmental impact, "ecological hydrogel composites" are hydrogel-

based materials made of environmentally safe polymer matrices (such as natural or biodegradable polymers) integrated with sustainable photothermal components, such as bio-derived or waste-derived carbon materials and low-toxicity functional modifiers. Therefore, the three main material categories covered in this review are: (i) hydrogels based on biopolymers (such as cellulose, chitosan, alginate, and polyvinyl alcohol); (ii) carbon-based photothermal fillers made from biomass or polymer waste (such as biochar, carbonized fibers, and graphene-like carbons); and (iii) composite hydrogel architectures designed for improved interfacial evaporation and mechanical robustness. These materials, often integrating sustainable and biocompatible elements, exhibit significant potential in increasing the efficacy of solar-based water evaporation. Beyond facilitating efficient evaporation, these composites fulfill the non-toxicity criterion, rendering them environmentally and human health-friendly^[58]. Navigating this evolving landscape mandates an exploration of both historical precedents and contemporary advancements. By drawing insights from past successes and learning from failures, we can strategically design ecological hydrogel composites that effectively address the challenges posed by water pollution and scarcity^[59]. Examining a diverse array of examples and innovations provides a comprehensive foundation for the development of these materials, enabling us to understand their necessity and envision revolutionary applications in the field of water treatment^[60]. In a significant demonstration, the proposed material exhibited a water evaporation rate and solar photothermal conversion efficiency of $2.44 \text{ kg m}^{-2} \text{ h}^{-1}$ and 153.2%, respectively, under sunlight irradiation^[61]. This emphasizes the capability of ecological hydrogel composites to reshape the field of water treatment, providing solutions that surpass earlier anticipations.

While the global water crisis is exacerbated by industrial effluents and agricultural runoff, traditional centralized treatment systems are increasingly strained by their high energy consumption and the generation of harmful chemical by-products. In this context, solar-driven interfacial water evaporation (ISDE) has emerged as a transformative, decentralized technology that localizes thermal energy at the air-water interface to achieve high efficiency with a minimal carbon footprint.

This review specifically distinguishes itself from previous ISDE literature by focusing on the full lifecycle and synergistic mechanisms of "ecological hydrogel composites". Unlike standard photothermal materials, these ecological composites are defined as three-dimensional (3D) polymeric networks derived from renewable, non-toxic, and biodegradable precursors (e.g., cellulose, alginate, starch, etc.) through green synthesis routes. While prior works often focus on photothermal conversion alone, this review emphasizes the unique ability of hydrogel matrices to regulate the thermodynamic state of water—specifically by generating "intermediate water"—to achieve evaporation rates that exceed theoretical limits for bulk water. By integrating high-impact recent developments^[62-63] this work establishes a coherent narrative from fundamental design principles to multifunctional environmental applications.

2. Photothermal Water Evaporation Design Principles

2.1. Theory

The efficiency of solar-driven photothermal water evapora-

tion system generally depends on how well the design manages three things: light absorption, heat localization, and water transport. This idea is based on materials absorbing sunlight and transforming it into heat, which raises the temperature of water and starts the evaporation process^[64]. Developing a photothermal water evaporator involves selecting high-performance materials, fine-tuning water and thermal pathways, and engineering the system to prevent salt build-up^[65].

To provide a rigorous and internally consistent thermodynamic treatment, the net energy balance at the hydrogel-air interface is given as^[66]:

$$P_{in} = P_{evap} + P_{loss} = P_{evap} + (P_{rad} + P_{conv} + P_{cond}) \quad (1)$$

Where P_{in} is the incident solar power, and P_{rad} , P_{conv} , and P_{cond} represent radiative, convective, and conductive heat losses, respectively. The resulting rate of evaporation (E) can be determined by the energy remaining after accounting for these transport losses:

$$E = \frac{\eta \cdot P_{in} - (P_{rad} + P_{conv} + P_{cond})}{h_{LV,eff}} \quad (2)$$

In addition, the ratio of stored thermal energy to incoming solar flux can be calculated to get the thermal conversion efficiency (η)^[67], as illustrated in the following equation:

$$\eta = \frac{m \cdot h_{LV,eff}}{C_{opt} \cdot P_0} \quad (3)$$

Where m is the mass flux (evaporation rate), $h_{LV,eff}$ is the total liquid-vapor phase-change enthalpy, C_{opt} is the optical concentration, and P_0 represents nominal solar irradiation (1 kW/m²).

While these equations (1), (2) and (3) define the general thermodynamic framework, the specific physical architecture of hydrogel systems significantly modulates these parameters to surpass the limits of bulk water through three different mechanism:

i) Porosity and Effective Surface Area (A): Hydrogels are multi-scale porous network. The network is relatively large in area and has a high surface area (A_{eff}) for liquid-vapor exchange. The rate of evaporation increases with the increase in this interface:

$$E \propto A_{eff} = A_{proj} \times (1 + \phi) \quad (4)$$

Where A_{proj} is the projected area and ϕ is the porosity factor. This network is also a quick-water supply network and is suitable for vapor escape; therefore, the interface does not form a “drying-out” phenomenon^[67].

ii) Thermal Localization and ΔT : Increase the temperature difference ΔT at the surface, and since hydrogels are naturally poor conductors of heat, the conduction loss P_{cond} to the bulk water is reduced. Fourier’s Law is applied here:

$$P_{cond} = k_{eff} \cdot \frac{\Delta T}{d} \quad (5)$$

Where k_{eff} is the thermal conductivity and d is the material thickness. By keeping k_{eff} low, typically, below 0.6 Wm⁻¹K⁻¹, most of the absorbed solar energy is still concentrated at the evaporation front and has not diffused into the body of water^[68].

iii) Water States and Reduced Enthalpy (L or h_{LV}): Hydrogels have a small latent heat of vaporization ($h_{LV,eff}$) and are thus good insulators. Total enthalpy is a weighted average of the states of water in the polymeric network:

$$h_{LV,eff} = (1 - x) h_{free} + x \cdot h_{IW} \frac{\Delta T}{d} \quad (6)$$

Where x is the fraction of intermediate water (IW) and $h_{IW} < h_{free}$. Hydrophilic functional groups (e.g., -OH, -COOH) weaken the hydrogen-bonding of water molecules in the 3D network to produce intermediate water that is much more easily evaporated. Therefore, the evaporation rate of hydrogels is higher than that of bulk water under the same conditions (1.47 kgm⁻²h⁻¹)^[69].

Materials in photothermal technologies for high-efficiency evaporation must have the following required properties:

- **High solar absorption:** They should absorb a relatively large amount of the incident solar radiation.
- **Low thermal emission:** This minimizes heat loss (P_{rad}).
- **Controlled thermal conductivity:** To transfer the absorbed heat to the water efficiently and minimize loss to the bulk.
- **Durability:** Materials should withstand outdoor conditions.

As shown in **Figure 2**, the above thermodynamic parameters are associated with the internal material properties of hydrogels.

2.2. Benchmarking Criteria for Practical Relevance

For evaporation rates and efficiencies to be comparable across different studies, it is essential to evaluate materials against a standardized set of benchmarking criteria. As recent literature shows, high laboratory performance does not always translate to practical viability^[70-71]. The following parameters are proposed as the standard for normalizing reported data:

• **Feed Composition and Salt Concentration:** Distinguishing between pure water, standard seawater (3.5 wt% NaCl), and high-salinity brines (>10 wt% NaCl) is critical, as salt concentration significantly affects chemical potential and evaporation kinetics^[72-73].

• **Illumination Conditions:** Reporting should adhere to standardized 1-sun intensity (1 kW·m⁻²) with a standard AM 1.5 solar-radiation spectrum to allow for universal efficiency comparisons^[71].

• **Test Duration & Cyclic Stability:** Beyond 1-hour laboratory benchmarks, practical relevance requires assessment of 12–24 hour continuous operation and long-term cyclic stability (e.g., >30 cycles) to account for mechanical fatigue and chemical degradation^[68].

• **Environmental Factors:** Reliable data normalization requires explicit reporting of ambient temperature, relative humidity, and wind speed, as these factors often cause a significant laboratory-to-field performance gap^[74].

• **Distillate Quality:** Verification that treated water meets WHO drinking standards through the measurement of ion rejection rates and the removal of emerging organic or biological pollutants is necessary for real-world deployment^{[68][70]}.

2.3. Basic Setup of Interfacial Solar Evaporation System

The foundational solar water evaporation setup com-

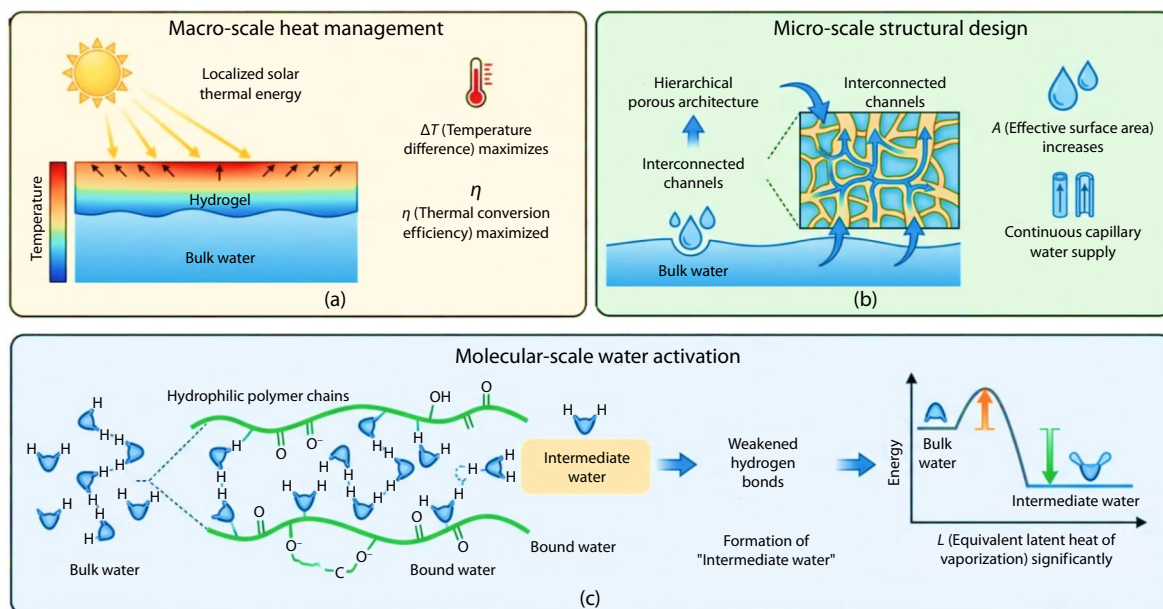


Fig. 2. Conceptual figure linking hydrogel physiochemical properties to thermodynamic evaporation parameters. (a) Schematic of the photothermal hydrogel displaying heat localization at the interface; (b) The hierarchical porous architecture provides an enlarged effective surface area; and (c) Molecular interaction between hydrophilic polymer chains and water, intermediate water state, weakened hydrogen bond, and significantly lowering of equivalent latent heat of vaporization.

prises a solar collector, water reservoir, and condensation system shown in **Figure 3a and 3b**; the materials with high absorp-

tive capacity absorb a large amount of sunlight and transform it into heat. Water supply is absorbed by the insulation and trav-

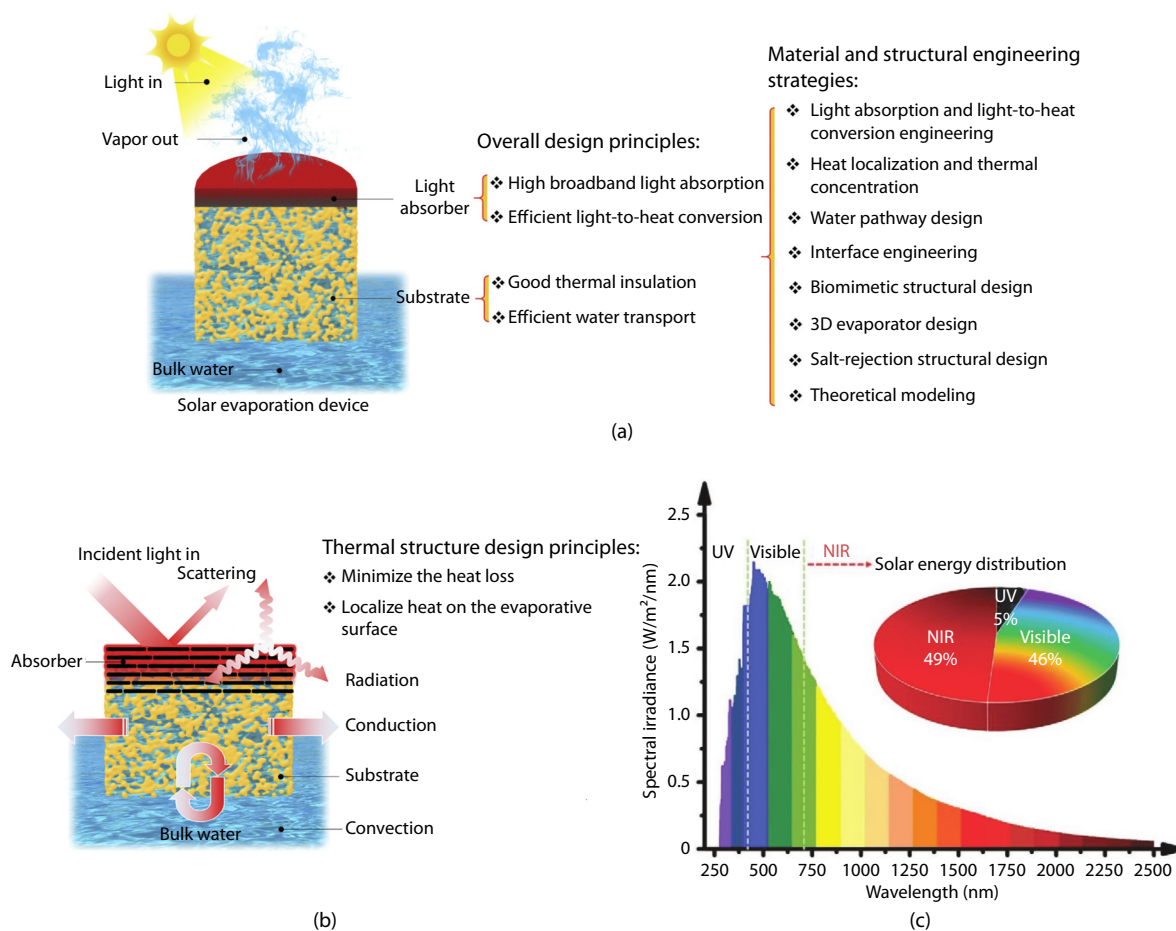


Fig. 3. (a) and (b) The Overall Material/Structural Scheme of a Typical Double-Layered Interfacial Solar Evaporation System, Including Light Absorber, Substrate, Bulk Water, Incident Light, and Vapor. The overall design principles and material and structural engineering strategies are also demonstrated. Reproduced with permission from the Elsevier^[75]; (c) Standard AM 1.5 solar-radiation spectrum and solar-energy distribution. Reproduced with permission from the Wiley-VCH GmbH^[77].

els through capillary forces to the evaporative surface, where it is continuously evaporated due to the heat created. The materials with high absorptive capacity absorb a large amount of sunlight and transform it into heat. However, heat loss occurs through conduction, radiation, and convection, impacting efficiency^[75]. A material's capacity for solar-to-thermal conversion depends heavily on its optical performance, specifically its spectral absorption range and the intensity of that absorption across various wavelengths. The standard energy distribution for sunlight reaching Earth, known as the AM 1.5 spectrum, serves as the benchmark for evaluating these materials, as it details the power available across the ultraviolet, visible, and infrared regions shown in **Figure 3c**^[76-77]. To optimize energy conversion, photothermal materials must behave as 'black bodies' within the solar spectral range, ensuring that incident photons are neither reflected nor transmitted but are instead fully absorbed and converted into heat.

3. Hydrogel Frameworks Design Logic: Connecting Polymer Chemistry to Performance

Hydrogel composites, a highly versatile class of materials, have garnered acclaim for their exceptional capacity to absorb and retain substantial volumes of water. Their inherent quality makes them extremely important for a variety of uses, especially in the field of water evaporation^{[53][78]}. We will examine the polymer backbones used in hydrogel composites

intended for water evaporation in this discussion, emphasizing the use of both natural and synthetic polymers. Selecting the right polymer backbones is crucial for the hydrogel composites' ability to evaporate water.

3.1. Chemical Functionality and Water-State Regulation

The primary role of the polymer backbone is to disrupt the hydrogen-bonding network of bulk water via hydrophilic functional groups ($-\text{OH}$, $-\text{COOH}$, $-\text{NH}_2$). This molecular interaction is the "engine" behind enthalpy reduction.

3.1.1. Natural Biopolymers: High-Density Water Activation

Natural polymers, derived from renewable sources^[79], exemplify a sustainable choice in the development of these composites. Examples include polysaccharides^[80-82] like cellulose^[83] and chitosan^[46-47], known for their biocompatibility and environmentally friendly attributes^[45]. The incorporation of such natural polymers underscores the commitment to sustainable practices in material design for water evaporation applications.

- *Cellulose-Based Hydrogels*: These are derived from plant cell walls^[84-85] (**Figure 4**), cellulose emerges as a sustainable solution that can be finely tuned to optimize thermal conductivity^{[52][86]}. Through precise modulation of porosity and crosslinking^[87-88], these frameworks are customized to enhance salt toler-

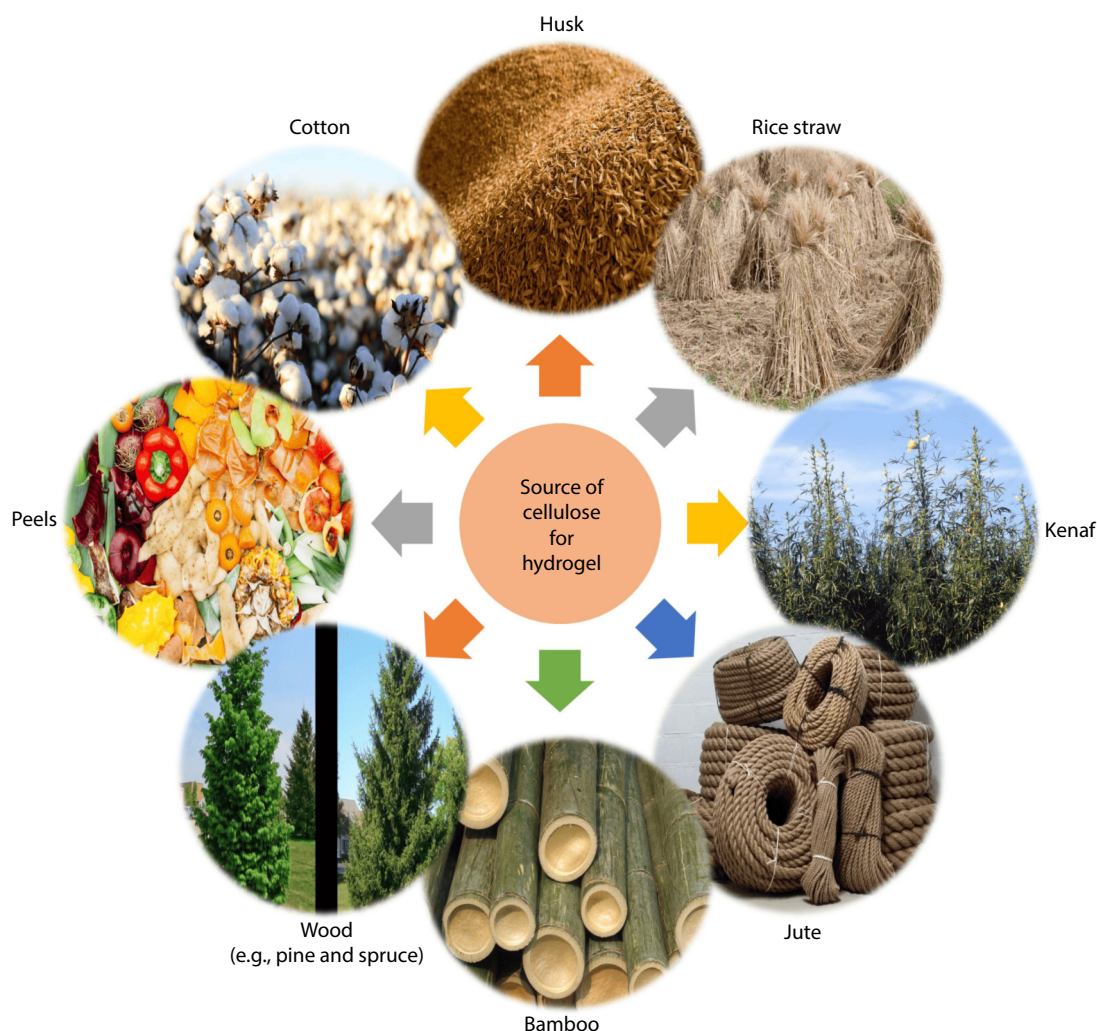


Fig. 4. Source of cellulose for hydrogel.

ance^[89] and ensure longevity under prolonged thermal stress. Furthermore, customization for solar responsiveness allows cellulose-based hydrogels to integrate efficiently into solar-driven desalination processes. Optimizing the hydrogel to absorb and convert solar energy, contributes to the sustainability of thermal desalination methods^[90].

- **Starch-Based Hydrogels:** As shown in **Figure 5**, starch hydrogels^[91] offer non-toxic and biocompatible characteristics^[92]. They are specifically selected for scenarios where maintaining water purity is critical, such as healthcare or food processing^[93-94].

- **Alginate-based Hydrogels:** These are mainly obtained from brown algae^[95-96] specifically phaeophyceae^[41], and alginate has a very large water-absorbing capacity. Laminaria and Sargassum^[97] can be used to obtain a stable source for high-flux applications.

3.1.2. Synthetic Frameworks: Structural Tunability and Resilience

Synthetic polymers can be used to prepare more varied and adjustable hydrogel composites^[98-99]. Polyvinyl alcohol (PVA)^{[51][100]} and polyacrylamide^[101-102] are synthetic polymers that can be used to control the porosity and water absorption capacity of materials precisely^[103].

- **Poly(vinyl alcohol) (PVA) -based composite:** PVA is a synthetic polymer that is widely used in various areas, and it has been applied to desalination^[104]. PVA has good cross-linking

characteristics and is therefore a suitable raw material for preparing three-dimensional networks of hydrophilic polymers that can hold a large amount of substantial water and maintain their structure^[105]. The formation of PVA hydrogels includes crosslinking PVA chains, and thus a gel-like structure is obtained. This crosslinking process can be achieved through physical and chemical means^[106]. Its mechanical strength and elasticity can be modified for resilient performance in solar-driven systems^{[57][107]}.

- **Polyacrylamide (PAAm) -Based composite:** It is highly water-absorbing and frequently used in places that require high water retention and specific control of irrigation-style flux^[108-109].

- **Poly(N-isopropyl acrylamide) (PNIPAM) -Based composite:** PNIPAM has a Lower Critical Solution Temperature (LCST)^[110-113] and is therefore selected for this reason. Thus, a "temperature-sensitive" structure can be designed that collapses and opens when the temperature reaches a certain value.

3.2. Structural Logic: Pore Architecture and Capillary Transport

Chemical Composition of the hydrogel network directly affects the size and the pore architecture for salt resistance and water supply.

- **Pore Size vs. Transport Flux:** Large macropores (> 100 μm) in virgin cellulose have a low resistance to vapor escape but

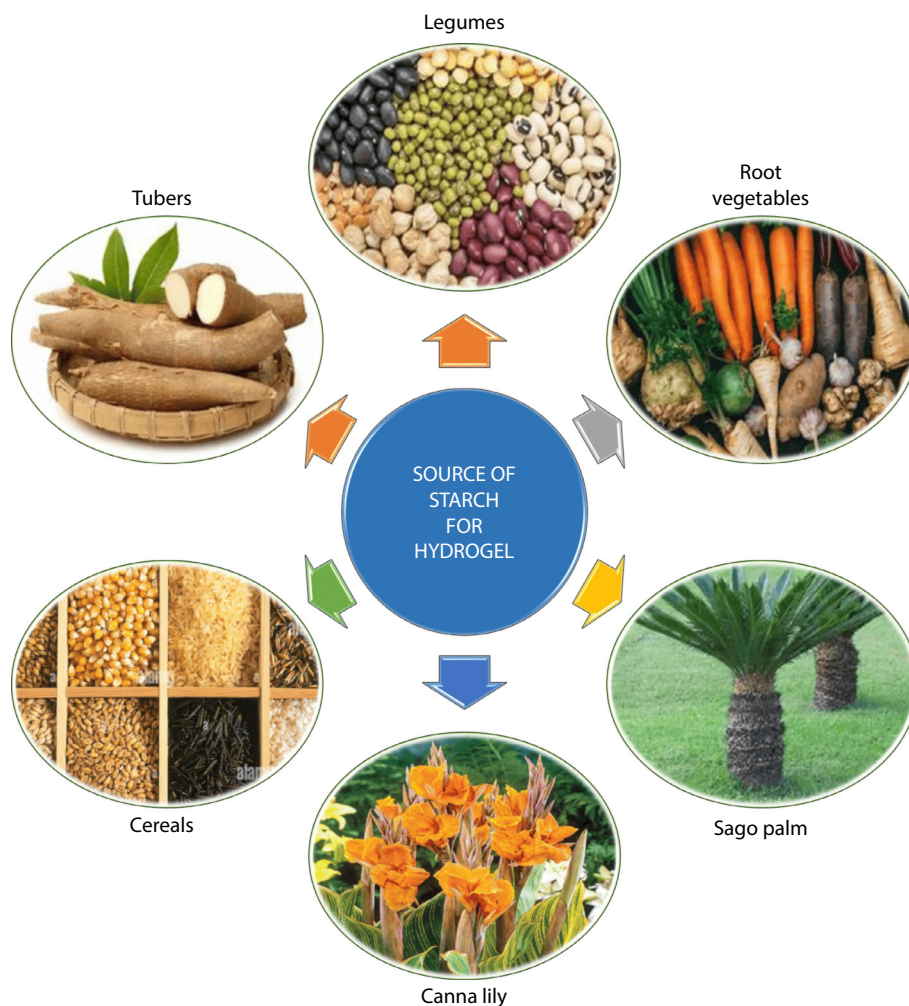


Fig. 5. Source of starch for hydrogel.

may exhibit weaker capillary forces. A fine-grained composite system can be used to form an "intermediate" pore, and thus capillary suction is generated to draw water out of the bulk phase and prevent "dry-out".

- **Decision Rule for Operating Environments:** In high-salinity environments (e.g., 20 wt% NaCl), the framework should be used for salt back-diffusion. As shown in the CSG systems, a framework that keeps a continuous water skin can be used to quickly transport salt ions back to the bulk water and prevent surface crystallization.

3.3. Lifecycle Sustainability: From Feedstock to End-of-Life

The selection of a hydrogel framework is not only a matter of performance but also of environmental accountability. As the field moves toward ecological solutions, a decision-oriented design must consider the functional benefits of a material against its lifecycle footprint. To address these sustainability concerns, we systematically compare the material classes used in ecological hydrogel composites in terms of feedstock origin, biodegradability, and end-of-life risks.

Table 1 highlights, there is a clear distinction in the environmental legacy of these systems. While natural biopolymer matrices are inherently biodegradable and align with circular economy principles, common synthetic alternatives like PAAm or PVA often persist in wastewater or break down into microplastic fragments. Furthermore, the inclusion of high-performance photothermal additives, such as CNTs and carbon black, introduces a specific challenge; while the polymer matrix may degrade, these carbonaceous fillers often persist as residual "mats" with potential adverse effect on terrestrial environments.

4. Synergistic Integration of Photothermal Filler Materials

The combination of carbon materials and hydrogel matrices creates a synergistic environment for ISDE. The integra-

tion of additive materials and fillers emerges as a pivotal strategy in advancing the capabilities of hydrogels across diverse applications, with a particular focus on their role in photothermal processes^[117]. This augmentation plays a crucial role in elevating mechanical properties, enhancing structural integrity, and optimizing overall performance, thereby positioning hydrogels as efficient tools in solar-driven evaporation and desalination^[118].

4.1. Filler Selection Based on Thermal Localization

A critical design rule in interfacial solar desalination system is the deliberate addition of fillers serves to augment light absorption and heat generation capabilities^[119]. This strategic enhancement amplifies the overall efficiency of the process, enabling hydrogels to play a more effective role in harnessing solar energy for desalination purposes^[120].

4.1.1. High-Conductivity Fillers (Graphene and CNTs):

Renowned for exceptional photothermal conversion properties, graphene and carbon nanotubes play a pivotal role in ensuring efficient heat transfer during photothermal evaporation^[54]. As detailed in Section 6.1, these material are the standard for broadband solar absorption due to their π -electron systems. However, a key decision factor is their high intrinsic thermal conductivity.

- **Design Rule:** While Graphene/CNTs offer superior conversion, their high conductivity can inadvertently promote heat dissipation into the bulk hydrogel. To prevent this parasitic heat loss, they should be integrated into low-conductivity frameworks or refined using tunable additives like silica nanoparticles^[121].

4.1.2. Low-Conductivity & Sustainable Fillers

The integration of eco-friendly options such as wood fibres and rice husk ash^[122] stands out for their characteristically low thermal conductivity, effectively minimizing heat loss during evaporation^[123].

- **Selection Logic:** These materials, alongside waste-derived forms like Biochar, should be prioritized for large-scale, sustain-

Table 1. Sustainability and life-cycle comparison of hydrogel composites across material classes.

Material Class	Feedstock Origin	Photothermal Component	Biodegradability	Potential Leachables	Realistic End-of-Life Risk	Ref.
Natural Biopolymers (e.g. Chitosan, Cellulose, Alginate)	Renewable (Biomass/Waste)	Biochar, polydopamine	High: Fully compostable under soil/ marine conditions. LCA shows low terrestrial ecotoxicity and supports circular economy principles	Minimal (organic metabolites, CO ₂ , biomass)	Biodegradation to non-toxic end products: aligns with circular economy with minimal ecotoxicity burden	[114]
Synthetic Polymers (e.g. PVA, PAAm, PAMPS)	Petroleum-based	Metallic (Au/ Ag/ TiN) or MXenes	Low to Partial: Slow hydrolysis; PVA often persists in wastewater; PAAm not biodegradable under normal conditions	Unreacted monomers (acrylamide); microplastic fragments	LCA identifies terrestrial ecotoxicity and human non-carcinogenic toxicity as dominant impacts. End-of-life routes are incineration or landfill with risk of persistent microsolids and soluble oligomers	[115]
Hybrid/ Composite Systems (Biopolymer + Fillers)	Mixed (Renewable + Industrial)	Carbonaceous (CNTs, Graphene, Black Carbon)	Selective: Matrix degrades; filler persists as residual CNT "mat"	Nanofillers embedded in microplastics during weathering; CNTs/ graphene inhibit matrix biodegradation	CNTs/graphene are rarely released as free particles but remain embedded within microplastics after matrix breakdown, complicating disposal. CNT accumulation can be noxious/ toxic ecosystems if unmanaged. Enzymatic/ oxidative degradation of CNTs requires specific peroxidase conditions not found in landfills	[116]

able applications where thermal insulation is as critical as light absorption.

4.2. Matching Filler Chemistry to the Operating Environment

The versatility of additive materials empowers a high degree of customization for hydrogel properties, enabling them to meet specific application requirements^[124]. Through the incorporation of diverse fillers in varying amounts, properties such as swelling behaviour, porosity, and mechanical strength can be precisely tailored^[125].

4.2.1. Semiconductor Fillers (TiO_2 , ZnO):

These emerge as robust selections for photothermal applications^[126]. They should be selected when the application requires multifunctionality, such as simultaneous evaporation and organic pollutant degradation. As shown in **Figure 6**, these metal oxides exhibit superior stability under heat and oxidative stress compared to carbon-based inks in industrial wastewaters.

4.2.2. Adaptability through Additives:

Silica nanoparticles and polymer microspheres offer a high degree of customization in the realm of photothermal evaporation^[121]. Their tunable porosity and thermal properties allow hydrogels to be precisely adjusted to meet the unique demands of specific water-treatment applications^[127-128].

To bridge the gap between material properties and system-level performance, **Table 2** presents a selection guide for photothermal fillers. This matrix acts as a decision-orient tool, allowing researchers to match specific design challenges—such as thermal localization or chemical stability—with the most appropriate filler category based on their inherent struc-

ture-property relationship.

5. Fabrication, Characterization, and Mechanical Robustness

5.1. Fabrication

High-performance evaporators are synthesized via chemical (covalent) or physical (reversible) crosslinking. A critical preparation technique is "freeze-thaw cycling", which creates physical junctions and hierarchical macropores without chemical additives. Hydrogels, intricate three-dimensional networks of water-absorbing polymers, exhibit diverse applications, particularly in desalination^[129]. The synthesis and preparation of hydrogel-based water evaporators involve tailored methodologies, ensuring precise control for various applications, including environmental remediation.

Chemical reactions form robust bonds between polymer chains. Utilizes techniques such as free radical polymerization and enzymatic crosslinking, ensuring the creation of resilient hydrogels^[130-131]. Suitable for a range of applications, including water purification and environmental remediation. Relies on forces like hydrogen bonding or hydrophobic interactions for hydrogel formation. Includes thermal gelation and photo-crosslinking, ensuring the creation of structurally sound hydrogels^[132-133]. Diverse applications, especially in water purification and agriculture. Furthermore, precise control of various parameters such as size, shape, structure, composition, cross-linking degree, and functional moieties is possible and similarly, precise encapsulation of drugs and active molecules can be performed during the synthesis. The chemical crosslinking shown in **Figure 7 (a)** and physical crosslinking shown in **Figure 7 (b)**, are reactions for the synthesis of nanogels^[134]. In situ

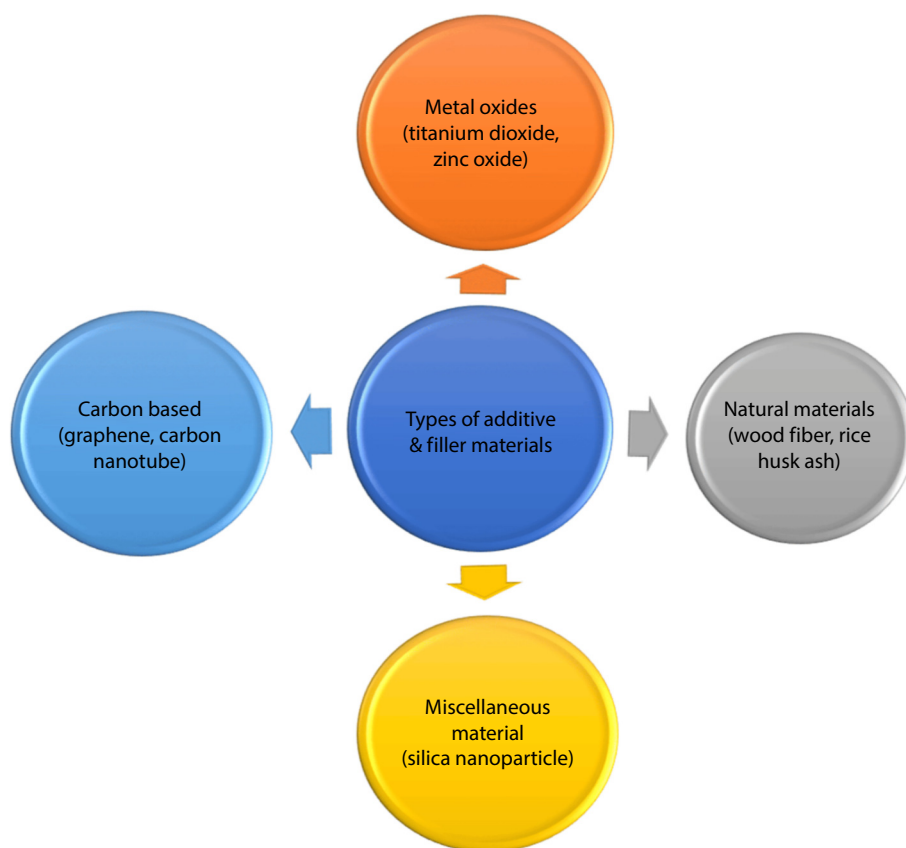


Fig. 6. Types of additive and filler materials.

Table 2. Selection Guide for Photothermal Fillers in Hydrogel Composites

Design Challenge	Preferred Filler Category	Physical Justification	Ref.
Maximizing Solar Harvest	Carbon Nanotubes/Graphene	Near-unity broadband absorption	[54]
Minimizing Heat Loss	Wood Fibres/Rice Husk Ash	Low thermal conductivity + Porous insulation	[122]
Industrial Purification	Metal Oxides (TiO ₂ , ZnO)	Chemical stability + Photocatalytic activity	[126]
Sustainable/Low Cost	Waste-derived Carbon (Biochar)	Circular economy + High availability	[123]

Polymerization shown in **Figure 7 (c)** occurs within the hydrogel matrix, offering meticulous control over structure and properties. Particularly advantageous for applications like environmental remediation, where precise control is essential. The polymerization can be initiated either by the addition of a curing agent or by an increase in temperature^[135]. The inexpensive method involving cycling between freezing and thawing is shown in **Figure 7 (d)**. Practical for agriculture and water purification applications, providing a cost-effective solution. The polymer mixture is subjected to the conditions of F/T cycles (PVA-XG diffraction pattern). This change in signal is supported by the formation of crystallites^[136].

5.2. Characterisation

Characterization techniques play a crucial role in gaining a deep understanding of composite materials^[137]. These materials are typically made up of two or more constituent materials with distinct properties. Here are some common characterization techniques used to analyze and understand composite materials shown in **Figure 8**.

The meticulous characterization of the cellulose hydrogel composite CNT/CH, depicted in **Figure 9A (a)** and validated through scanning/transmission electron microscope observations, plays a pivotal role in elucidating the enhanced mechanical properties and structure of the resultant material. The gelation process unfolds within a specially designed Mold, as illustrated in **Figure 9A (b)**. CNTs play a pivotal role in reinforcing the hydrogel structure, leading to significant improvements in mechanical properties. The incorporation of CNTs enhances ten-

sile strength, modulus, and overall structural stability, contributing to the superior performance of the composite hydrogel^[52].

5.2.1. Chemical Properties (FT-IR confirms network integrity post-stress)

The examination of chemical composition and functional groups in the hydrogel through Fourier Transform Infrared Spectroscopy (FT-IR) is a crucial aspect that confirms the presence of specific components and captures changes during the evaporation process^[44]. The infrared spectra, as depicted in **Figure 9A (c)**, serve to confirm the high purity of the corncob cellulose specifically extracted for this purpose. Characteristic absorption bands corresponding to cellulose, such as the β -1, 4 glycosidic bond at 898 cm⁻¹, the -C-O- group at 1030 cm⁻¹, and the hydroxyl group features around 3300 cm⁻¹, exhibit consistency between the homemade cellulose and commercial cellulose samples^[138-139]. The peak strength ratio at 1030 cm⁻¹ to 898 cm⁻¹ is notably higher for the CH sample, indicating an increased presence of -C-O- groups. This underscores the success of the cross-linking reaction and the synthesis of cellulose hydrogel. The visual appearance of the as-made CH hydrogel is brown-yellow, exhibiting commendable viscoelastic properties. Upon the addition of CNTs, the resulting CNT/CH composite transforms to black color, accompanied by a notable enhancement in mechanical properties^[52].

5.2.2. Morphological Properties (SEM shows pore preservation after cycling)

Scanning Electron Microscopy (SEM) serves as a powerful tool for visualizing the surface morphology and structure of

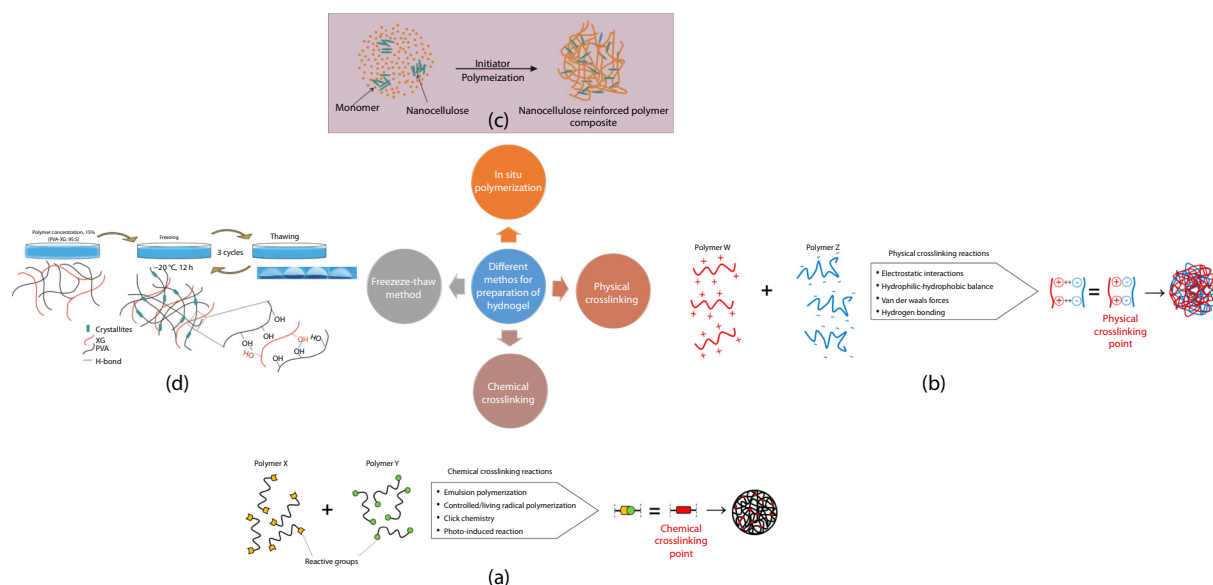


Fig. 7. Illustration of chemical (a) and physical (b) crosslinking reactions for the synthesis of nanogels. Reproduced with permission^[134] followed by the Creative Commons Attribution 4.0 International License; (c) Process for preparation of nanocomposite through in situ polymerization Reproduced with permission from the Taylor & Francis^[135]; (d) Diagram of PVA-XG hydrogel formation through freeze-thaw method Cross-Linked Polyvinyl Alcohol-Xanthan Gum Hydrogel Fabricated by Freeze/Thaw Technique for Potential Application in Soft Tissue Engineering. Reproduced with permission^[136] followed by the Creative Commons Attribution 3.0 International License.

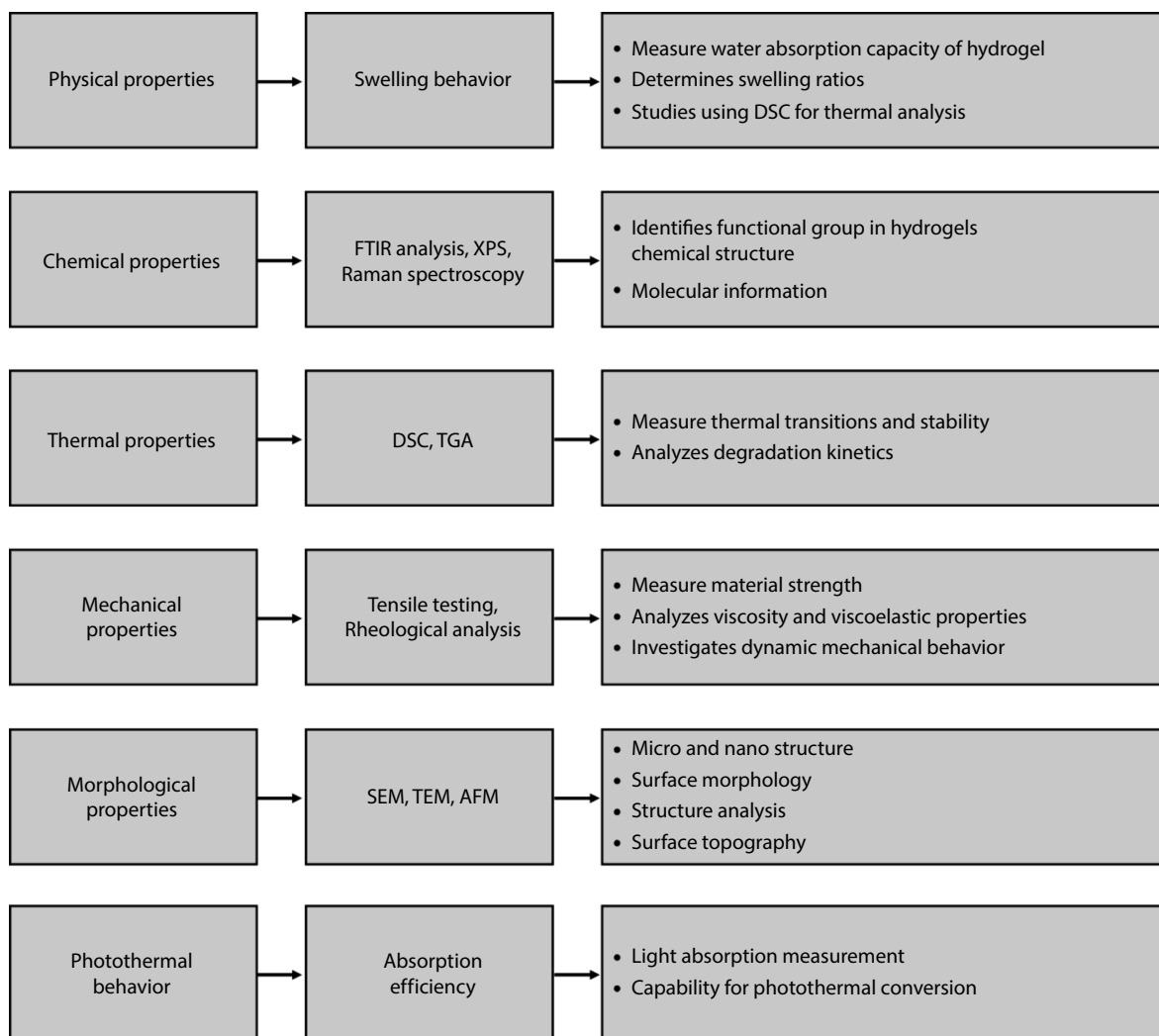


Fig. 8. Characterization techniques in photothermal evaporation.

hydrogels, providing crucial insights into microstructure and porosity^[140]. As depicted in **Figure 9B (a, b, c, d)** images, captured under the same magnification (scale bar: 200 μm) in post-swelling and freeze-drying, offer essential revelations. The analysis reveals that the pore size of the cellulose hydrogel (CH) alone can reach approximately 120 μm . Notably, with an increase in CNT content, the inner pore size of the composite hydrogel gradually diminishes. The pore size of 0.2% CNT/CH, for instance, reduces to about 30 μm to 20 μm shown in **Figure 9B (e, f)**. This dual role of CNTs is instrumental, dispersing within the 3D cross-linked network of the hydrogel while enhancing densification, resulting in a smaller pore structure with strong capillary forces. This structural enhancement facilitates efficient water transport during solar water evaporation in CNT/CH. Transmission Electron Microscopy (TEM) provides high-resolution imaging of the hydrogel's nanostructure, offering insights into its morphology and nanoscale porosity^[141]. TEM observations confirm the even dispersion of CNTs within the cellulose hydrogel as shown in **Figure 9B (g)**. High-resolution TEM (HR-TEM)^[142] analysis reveals the presence of the (002) crystal plane of CNTs with a spacing of 0.343 nm, as depicted in **Figure 9B (h)**. Optimally, in the case of the CNT/CH composite, CNTs are evenly dispersed within the cellulose cross-linked network, resulting in enhanced mechanical properties and preserved hydrogel hydrophilicity^[52].

5.2.3. Thermal Properties (DSC reveals "intermediate water" enabling flexibility) (Thermal Properties and Thermodynamic Water States in Hydrogels)

Differential Scanning Calorimetry (DSC) is a standard analytical technique for studying the thermal behaviour of hydrogel systems, especially their phase transitions and the heat flow during solar-driven water evaporation processes^[141]. In cellulose-based hydrogels, the high density of hydroxyl ($-\text{OH}$) groups allows extensive hydrogen bonding with water molecules. This leads to distinct thermodynamic water states that strongly influence evaporation performance. Unlike conventional evaporators, where water is mostly in a bulk state, hydrogel matrices regulate water into three categories: bound water, intermediate water, and free water. Bound water consists of molecules tightly associated with polymer chains through stable hydrogen bonding; they are energetically restricted and contribute little to evaporation. In contrast, free water behaves like bulk water, with standard thermodynamic properties and a need for full latent heat to undergo phase transition. Between these extremes lies intermediate water (IW), a weakly bound state characterized by partially disrupted hydrogen-bonding networks^[143]. This intermediate state is particularly significant because it has a lower vaporization enthalpy than bulk water, which enhances evaporation efficiency.

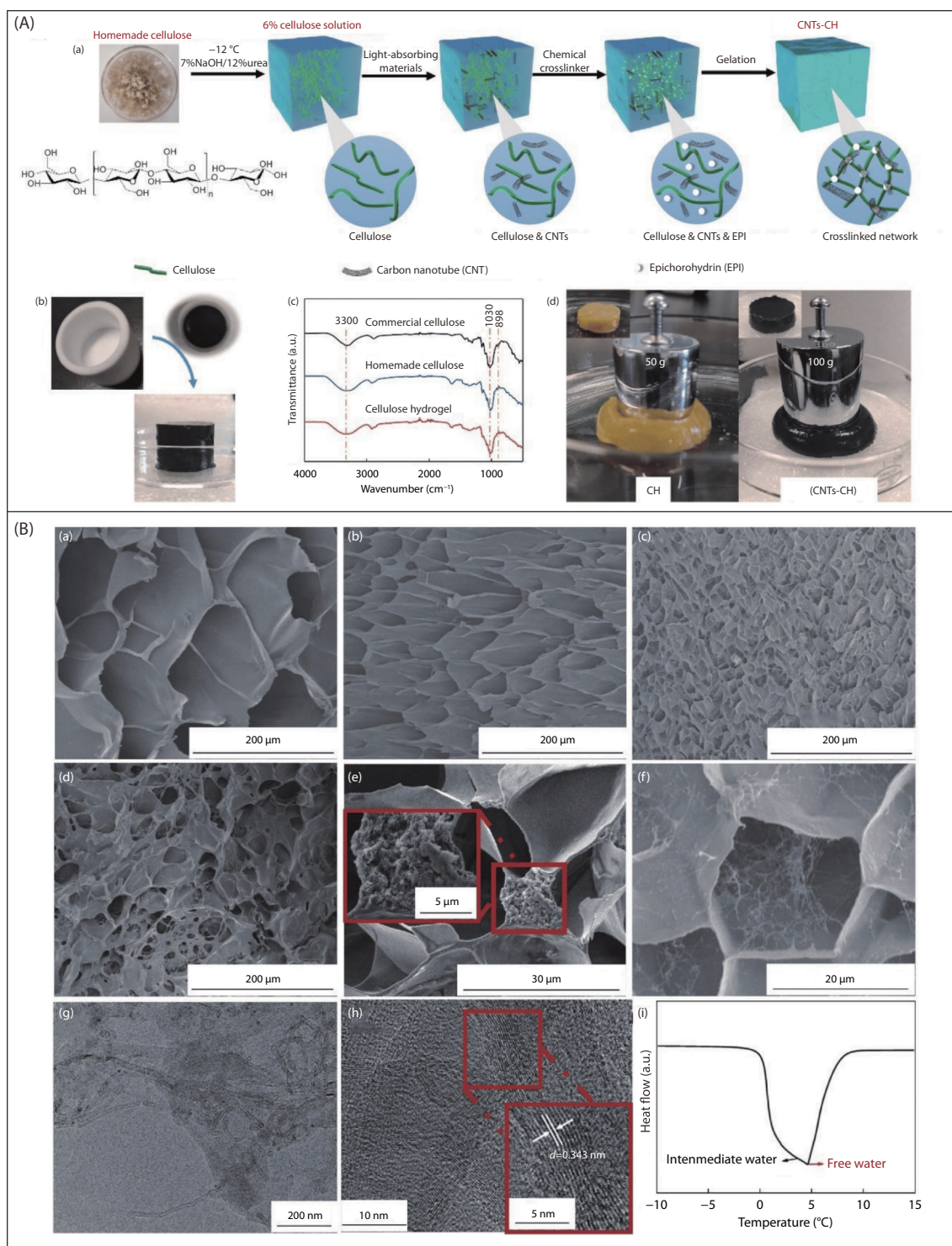


Fig. 9. **Figure 9A.** Preparation, appearance and FT-IR characterization of cellulose hydrogels. (a) Flow chart of the preparation of cellulose hydrogel composite (CNT/CH); (b) Digital photos of CNT/CH after demoulding; (c) Infrared spectra of commercial cellulose, homemade cellulose and cellulose hydrogel (CH); (d) Load-bearing diagrams of CH and CNT/CH (the inset is the recovery diagram of the sample after load-bearing); **Figure 9B.** Electron microscopic images and DSC curve of cellulose hydrogels. (a–d) SEM images of (a) CH, (b) 0.02% CNT/CH, (c) 0.2% CNT/CH, (d) 1.0% CNT/CH hydrogels swollen and freeze-dried at 200 μm size, respectively, (e, f) Local enlarged SEM images of typical 0.2% CNT/CH at 30 μm and 20 μm size (the inset shows the local enlarged view of the crosslinked network structure), (g) TEM and (h) HR-TEM images of 0.2% CNT/CH respectively, and (i) DSC curve of 0.2% CNT/CH hydrogel after swelling equilibrium. *Reproduced with permission from the Elsevier*^[52].

Researchers now view IW as a defining feature of hydrogel-based evaporators, as it reduces the overall liquid–vapor phase-change enthalpy, (h_{LV}) allowing evaporation rates that may exceed the theoretical limits predicted for bulk water systems.

DSC experiments have shown that the two different water

states are indeed separate. For example, in a typical 0.2% CNT/CH hydrogel system, the DSC thermograms show an asymmetric and broadened phase transition peak at an extended temperature range^[144]. Unlike the sharp melting peak characteristic of bulk water, these broadened transitions indicate the coexistence of multiple water populations, particularly free

water and intermediate water confined within the polymer network. Such thermal signals indicate irregular hydrogen-bonding environments and restricted molecular motion; therefore, it can be inferred that the hydrogel matrix has changed the local water structure to form reduced-energy evaporation pathways^[145].

DSC alone cannot reveal the full extent of water–polymer interactions, so a multi-technique approach is required for all-encompassing characterization^[146]. Fourier Transform Infrared (FT-IR) spectroscopy indicated that hydrogen bonding occurs at a wave number of about $\sim 3300\text{ cm}^{-1}$ for the –OH stretching vibrations, and thus a polymer–water interaction network is formed to stabilize the intermediate water^[147]. To study how and where water moves at the molecular level and how it is distributed in the water phase, high-end spectroscopic techniques are also being applied, such as Nuclear Magnetic Resonance (NMR) and Raman spectroscopy^[148]. At the same time, surface wettability tests near a contact angle of zero show that the hydrogel surface is very hydrophilic; therefore, water can be continuously transported from the bulk to the evaporation interface, and a dynamic equilibrium of thermodynamic water states is reached. Morphological analyses using scanning and transmission electron microscopy (SEM/TEM) reveal hierarchical porous architectures and interconnected three-dimensional networks that physically confine water molecules, thereby contributing to the altered phase transition behaviour observed in DSC^[149]. Collectively, these findings establish that both chemical interactions (hydrogen bonding) and physical confinement (micro-/nano-porous structure) synergistically govern the formation and stabilization of intermediate water.

In addition to thermal properties, hydrogels' structural design is important in deciding their mechanical robustness and functional stability. Carbon nanotubes (CNTs) have been added to cellulose hydrogels to form a dense, interconnected three-dimensional cross-linked network that substantially improves the mechanical properties of the hydrogel^[150]. Mechanical tests have shown that although the pristine cellulose hydrogels can be deformed and subsequently recovered under a 50 g load, the CNT/CH composites has sustained a load of up to 100 g and maintained structural integrity and full recovery after unloading, as shown in **figure 9a–d**. Reinforcement will increase the flexibility and extend the service life of the structure, help to stabilize the internal distribution and flow of water, and thus maintain a relatively high level of evaporation rate in operation.

Generally speaking, the whole-system efficiency of hydrogel-based evaporators is also limited by the photothermal conversion mechanisms of the added functional materials. Carbon-based materials, such as CNTs and graphene, are broadband absorbers of sunlight in the AM 1.5 spectrum and have near-blackbody characteristics. Their high thermal conductivity will cause a rapid and localized increase in heat at the water–solid interface, thus promoting efficient interfacial evaporation^[151]. TiO_2 and ZnO are semiconductor materials that generate electronic excitation in the bandgap upon photon absorption, and then non-radiative decay processes lead to the generation of heat. The above materials can promote photothermal–photocatalytic coupling in high-end systems to drive water evaporation and other chemical conversions, such as hydrogen evolution. Plasmonic systems that employ metallic nanostructures (e.g., Ag-based composites) utilize localized surface

plasmon resonance (LSPR) to generate highly localized thermal hotspots. Add a conductive polymer, such as polyaniline, to improve the performance of solar-to-heat conversion, and an efficiency of about 98% can be achieved in some cases.

The following are some still-to-be-solved problems. The quantitative differences between bound, intermediate, and free water still vary greatly by method, and without a standard analytical framework, the data from different studies cannot be directly compared^[152]. The stability of the intermediate water in a cycle of evaporation over an extended period under actual environmental changes has not yet been verified. Finally, the coupling between photothermal conversion efficiency and water-state modulation has not been well-studied at the mechanistic level, and more research is needed to connect the dynamics of molecular-scale hydration with the macroscopic performance of heat conversion. Addressing these problems will help to promote the development of hydrogel-based evaporators from laboratory research to large-scale application.

5.2.4. Surface Wettability and Contact Angle

In ISDE characterization, the contact angle is a vital metric for verifying surface wettability. A zero-contact angle indicates superior hydrophilicity, ensuring that water molecules are rapidly transported from the bulk to the evaporation front. This prevents the localized drying of the absorber surface, which would otherwise lead to salt crystallization and thermal degradation. The solar absorptance of ATHEs is angle-independent from 0° to 60° , making it suitable for various sunlight incident angles due to seasonal and time changes. The hydrogel demonstrates rapid water absorption, with a zero-contact angle. The water saturation content and transportation rate can be tuned by altering the agar/water ratio^[153].

5.2.5. Photothermal Performance

The performance of the Cotton-Thread Solar Gel (CSG) was rigorously evaluated in practical applications, focusing on its application in a solar-driven water purification device^[154], the device, illustrated in **Figure 10a** was designed for efficient water desalination. An evaporation test was conducted in saline water to assess the CSG's desalination capabilities. Notably, the evaporation rates were examined in pure water and NaCl solutions of varying concentrations (3.5, 10, and 20 wt%). While a slight decrease in the evaporation rate was observed with increasing salt concentration, the three evaporation processes remained stable. The mass change rate was essentially constant, as demonstrated in **Figure 10b**. The desalination process resulted in a significant reduction in the salinity of the desalinated water to below 3.3 ppm, achieving over 99.7% removal of Na^+ as depicted in **Figure 10c**. The long-term stability of the CSG was assessed through a 15-day evaporation rate test using a 3.5 wt% NaCl solution. **Figure 10d** illustrates that the evaporation rate exhibited minimal change, with a trend line indicating consistent performance. The average evaporation rate over 15 days was calculated as $1.565\text{ kg m}^{-2}\text{ h}^{-1}$, suggesting the device's reliability for sustained use in brine evaporation cycles. **Figure 10e** shows that even under varying light intensities, the evaporator maintained a high evaporation rate exceeding $1\text{ kg m}^{-2}\text{ h}^{-1}$ ^[153]. The total Dissolved Solids (TDS) measurement shows the activated carbon with an activating agent of NaOH after filtering the polluted water meets the standard requirements for drinking water^[144]. The TDS in the seawater post-evaporation reduced significantly from 8780 to 32 ppm. The CSG effectively removed primary

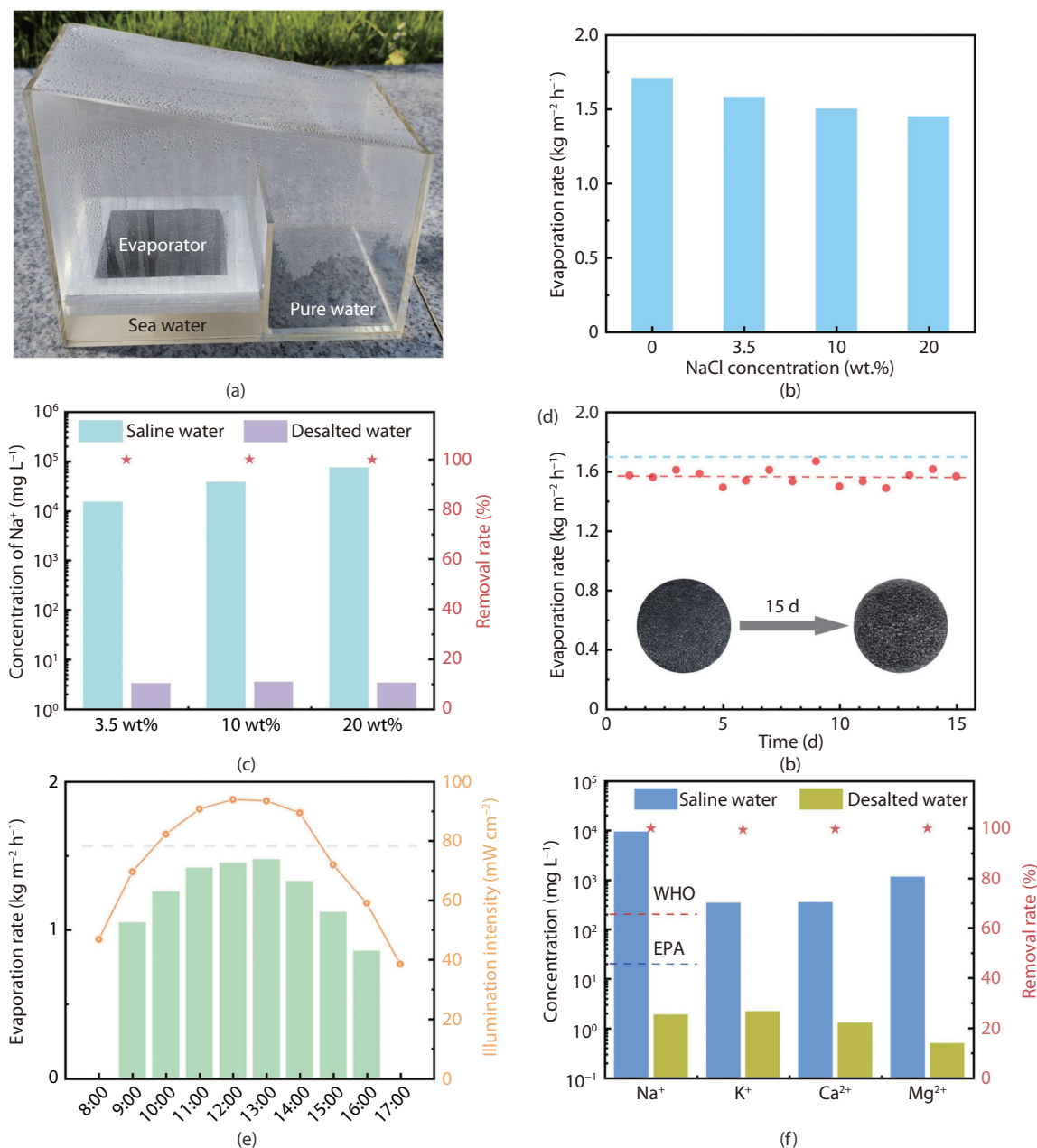


Fig. 10. (a) Schematic of evaporation device; (b) evaporation rate of CSG (with 3.5, 10, 20 wt% NaCl solution, under 1.0 sun); (c) Na^+ concentration and removal rate (with 3.5, 10, 20 wt% NaCl solution before and after treatment, under 1.0 sun); (d) evaporation rate of CSG in 15 day cycle (with 3.5 wt% NaCl); trend line of 15 day evaporation rate (red line), evaporation rate of pure water (blue line); (e) sunlight intensity (0.38–0.93 sun, 08:00 to 17:00) and evaporation rate of CSG from 09:00 to 16:00; (f) concentrations and removal rates of Na^+ , K^+ , Ca^{2+} and Mg^{2+} before and after seawater desalination by CSG evaporation. Reproduced with permission from the Elsevier^[53].

cations (Na^+ , K^+ , Ca^{2+} , and Mg^{2+}) with a removal efficiency exceeding 99.3% shown in Figure 10f^[53].

Since intermediate water is defined operationally by the characterization method used, Table 3 critically compares five characterization techniques—DSC, FT-IR/Raman, NMR, wettability, and cryo-SEM in terms of their operational definitions, key indicators, and known limitations. As highlighted in recent reviews, DSC signatures alone are insufficient due to peak overlap and heating-rate dependence, making multi-method validation essential for reliable quantification of water states.

5.3. Mechanical Robustness

Beyond optical absorption and thermal management, the mechanical resilience and shock resistance of hydrogel-based photothermal systems are crucial for practical implementa-

tion^[68]. These systems are subjected to various mechanical stresses, including handling stress, hydraulic turbulence, wave-induced impact, and recurrent mechanical deformation during real-world operation. Such stresses can compromise the structural integrity and performance of the hydrogel, leading to reduced efficiency and lifespan. Therefore, shock-resistant hydrogels are essential for ensuring the long-term stability, durability, and performance retention of photothermal and water-energy conversion devices^[160].

6. Applications of Hydrogel Composites as a Water Evaporator

6.1. Hydrogel Composite of Carbonaceous Materials

Materials characterized by a high proportion of carbon

Table 3. Critical comparison of characterization methods for classifying water states in hydrogel evaporators

Method	Operational Definition of Water States	Key Indicators	Critical Limitations and Sources of Disagreement	Ref.
DSC	Thermal Transition: Based on presence/absence of melting endotherm during heating	Free: Endotherm at $\sim 0^{\circ}\text{C}$; Intermediate: Endotherm between -40°C to -10°C ; Bound: Non-freezing	Peaks often overlap; enthalpy values depend on heating rate and polymer concentration, leading to hidden intermediate water	[155]
FT-IR/ Raman	Hydrogen Bonding: Based on O–H stretching frequency	Free: Sharp band $\sim 3490\text{ cm}^{-1}$; Bound: Broad band $< 3200\text{ cm}^{-1}$; Intermediate: $3200\text{--}3400\text{ cm}^{-1}$ shoulder	Polymer backbone overlap; qualitative; sensitive to hydration level and film thickness	[156]
NMR	Molecular Mobility: Based on reorientation speed (T_2 relaxation) or self-diffusion coefficient D	Bound: $T_2 < 10\text{ ms}$; Intermediate/Free: $T_2 > 100\text{ ms}$, separated by D : $D_{\text{free}} \approx 2.3 \times 10^{-9}\text{ m}^2/\text{s}$, $D_{\text{intermediate}}$ lower	Rapid exchange between sites blurs states; requires multi-component fitting with model-dependent assumptions	[157]
Wettability	Surface Energy: Indirect measure of polymer–water affinity	High bound/intermediate fraction correlates with contact angle $< 10^{\circ}$ (super-hydrophilicity)	Indirect; cannot quantify water types; dominated by surface roughness/chemistry	[158]
Cryo-SEM	Pore Morphology: Based on ice crystal footprint after sublimation	Free: Large hexagonal crystals $> 1\text{ }\mu\text{m}$; Intermediate/Bound: Suppressed or no visible crystals	Qualitative only; freezing artifacts from cooling rate can mimic or mask true water states	[159]

atoms in their structure^[161] are termed carbonaceous. The versatile applications of carbonaceous materials^[162] Spanning energy, the environment, electronics, and healthcare, have garnered significant attention in recent years. Among their various applications and properties shown in Figure 11, the use of carbonaceous materials in evaporation processes, where liquid water is transformed into vapor using thermal or solar energy, holds considerable importance. Evaporation finds applications in power generation, cooling systems, desalination, and water purification. The effectiveness of water evaporation is influenced by the properties of the materials employed to absorb and transport heat to the water. This overview out-

lines the main categories of carbonaceous materials utilized for evaporation, their characteristics, and discusses the advantages and limitations of each material^[163]. Additionally, recent research examples highlighting the applications of these materials are presented.

6.2. Hydrogel Composite of Biodegradable Materials

Polymers, both organic and synthetic^[164], are gaining prominence due to their ability to break down in the environment and their derivation from renewable sources. These eco-friendly, biodegradable, and biocompatible materials have found applications across diverse sectors, including food, pharmaceuticals, agriculture, industry, and energy. The thermal sta-

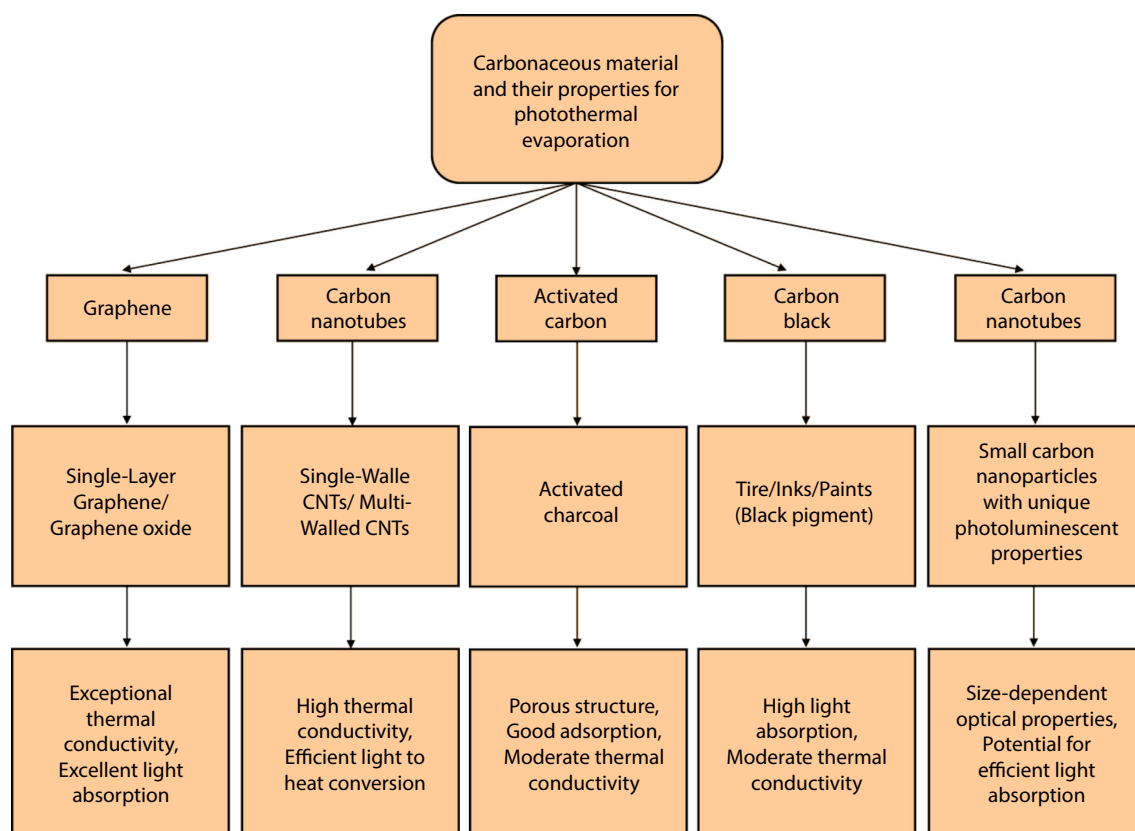


Fig. 11. Carbonaceous material and their properties for photothermal evaporation.

bility of biopolymers, influenced by factors such as chemical structure, molecular weight, crystallinity, and crosslinking, profoundly impacts their performance and applications^[165-166].

In the realm of biodegradable materials, natural biopolymers and synthetic counterparts exhibit the capacity to undergo degradation in the ecosystem or soil. As illustrated in **Figure 12** natural biopolymers including cellulose, starch, chitin, proteins, pectin, and lignin, originate from plant cell walls, carbohydrates, crustacean/insect shells, and various biological structures. These materials biodegrade over time, thanks to the activity of microorganisms and enzymes in the surrounding environment. Synthetic biodegradable polymers such as Polylactic Acid (PLA), Polyhydroxyalkanoates (PHA), Polybutylene Succinate (PBS), Polycaprolactone (PCL), Polyglycolic Acid (PGA), and Polyvinyl Alcohol (PVA) are engineered from renewable resources or produced by bacteria. They demonstrate the ability to biodegrade under specific conditions, contributing to sustainability^[167].

Furthermore, synthetic polymers like PCL and PGA are utilized in medical applications^[168], such as sutures and tissue engineering, and can biodegrade in the body through hydrolysis. While PVA's biodegradability is less pronounced in natural ecosystems, it offers environmentally responsible alternatives in various applications. These biodegradable polymers present sustainable alternatives across industries, contributing to reduced environmental impact and aligning with eco-conscious practices.

6.3. Hydrogel Composite of Waste-Derived Materials

In the pursuit of sustainability and efficient resource utilization, the integration of diverse waste streams holds immense potential to enhance the properties and versatility of hydrogels. As illustrated in **Figure 13**, agricultural waste, encompassing crop residues, fruit and vegetable peels, emerges as a valuable source of natural polymers and organic matter, offering substantial improvements to hydrogel performance across vari-

ous applications. This innovative approach not only addresses waste reduction but also aligns with eco-friendly agricultural practices^[169].

Simultaneously, repurposing industrial waste streams, such as scrap plastics and rubber, pencil waste^[170] provides an opportunity to impart structural support or introduce unique properties to hydrogels. Harnessing these discarded materials contributes not only to waste reduction but also fosters a circular economy ethos, where resources are repurposed for prolonged use^[171].

Exploration of organic kitchen waste^[172] in fruit and vegetable peels for obtaining a source of natural polymers is an eco-friendly way to improve the performance of hydrogels^[173-174]. Add natural foaming agents, such as plant-derived saponins, to produce a foamy texture to the hydrogel and extend its versatility and application range of green products^[175]. These new modes will reduce waste and promote ecological civilization; at the same time, they will also offer new development paths for high-performance materials with diverse functions.

The waste-derived hydrogel reduced waste by 30% compared to the traditional hydrogel production methods and had a relatively small impact on the environment^[176]. Resource re-use was also employed to reduce the amount of raw material required by 25% and enhance the sustainability of resource use [78].

6.4. Recent Developments: From Material Selection to Mechanistic Optimization

Recently, the research on hydrogel-based photothermal evaporators has shown a trend of moving away from material-centric design towards mechanism-driven optimization. Although previous studies have mainly focused on the incorporation of high-performance photothermal materials, research in recent years has begun to investigate how the evaporation

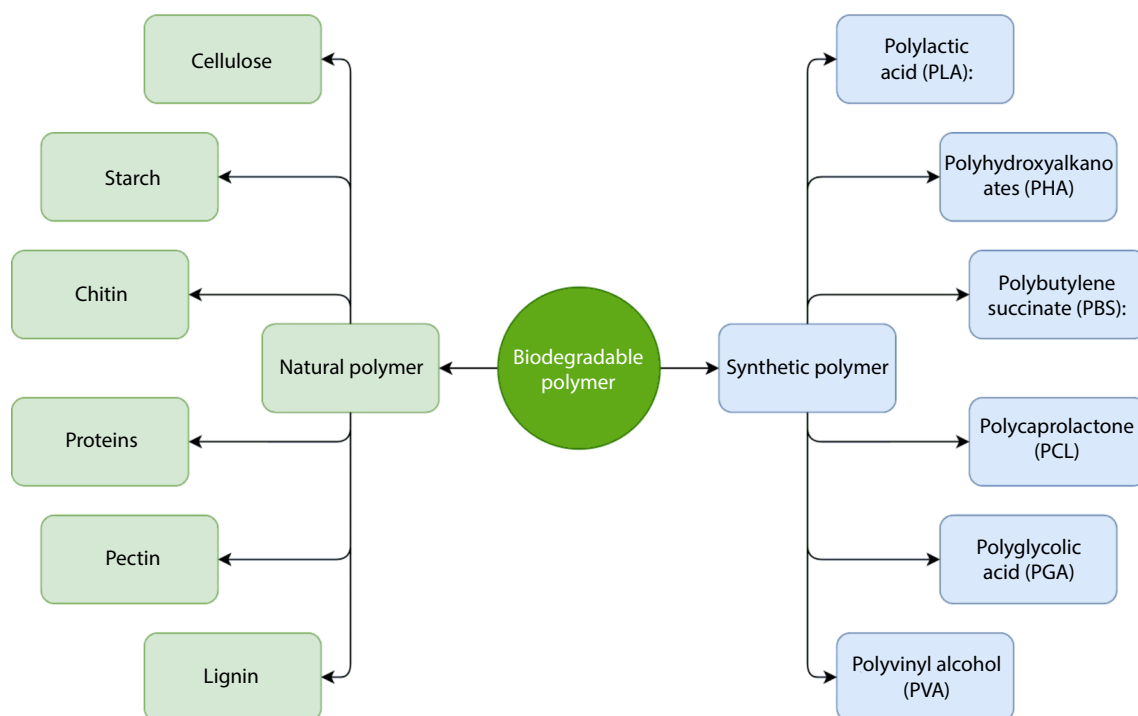


Fig. 12. Different natural and synthetic biodegradable polymers.



Fig. 13. Types of waste derived material for hydrogel composites.

performance of such systems is affected by interfacial heat management, water transport dynamics and hydration thermodynamics^[90]. This shift reflects how the field has evolved. Now, understanding and controlling coupled transport and thermodynamic matters more than simply going for high-absorption materials^[90].

A notable discrepancy exists between evaporation efficiency and evaporation rate across different hydrogel systems, highlighting an important limitation in conventional performance evaluation. For instance, CNT-based hydrogel composites, such as cellulose/CNT systems, demonstrate relatively high efficiencies (~92%) but exhibit only moderate evaporation rates ($\sim 1.52 \text{ kg m}^{-2} \text{ h}^{-1}$), whereas the Chitosan/PVA-carbon ink composite achieves both a significantly higher evaporation rate ($4.37 \text{ kg m}^{-2} \text{ h}^{-1}$) and an exceptional efficiency of 98.2%^{[52][177]}. This contrast indicates that high efficiency alone is not a sufficient indicator of overall performance, as sustained high evaporation flux depends on the synergistic optimization of heat localization, mass transport, and water-state regulation, which are not uniformly achieved across different material systems.

This disparity can be further understood by examining the role of thermal transport within these systems. Carbon nanotubes (CNTs), although widely used due to their high thermal conductivity and excellent photothermal conversion capability, can inadvertently promote heat dissipation into the bulk water, thereby reducing the effective temperature at the evapo-

ration interface^[178-179]. In contrast, high-performance systems such as Chitosan/PVA-carbon ink composites emphasize interfacial heat localization by confining thermal energy at the air-water interface, minimizing conductive losses and maximizing energy utilization for evaporation. This represents a fundamental shift from prioritizing heat generation to optimizing heat management, where spatial control of thermal energy becomes a critical determinant of system efficiency and evaporation rate.

In addition to thermal considerations, water transport dynamics play a crucial role in sustaining high evaporation performance. The three-dimensional polymeric network of hydrogels enables tunable porosity, which directly influences capillary-driven water supply to the evaporation interface^[180]. Optimized systems with pore sizes in the tens of micrometers range (e.g., 20–30 μm) generate strong capillary forces that ensure continuous and rapid water replenishment, thereby maintaining high evaporation flux^[181]. At the same time, the homogeneous dispersion of carbonaceous materials within the hydrogel matrix creates a synergistic structure that balances water retention and transport; however, excessive porosity may compromise thermal confinement and lead to heat loss. Consequently, high-performance hydrogels must achieve a delicate balance between capillary transport and thermal insulation, highlighting the importance of integrated structural and transport design^[182].

Beyond structural and thermal optimization, recent stud-

ies increasingly emphasize the role of water thermodynamics within hydrogel systems. In polymer networks such as Chitosan/PVA, water exists in multiple states, including bound water, free water, and intermediate water, with the latter playing a particularly important role in enhancing evaporation performance^[69]. Intermediate water, characterized by weak hydrogen-bond interactions with the polymer matrix, exhibits a reduced effective vaporization enthalpy compared to bulk water, thereby lowering the energy barrier for phase transition and enabling more efficient evaporation under solar irradiation^[183-184]. This insight represents a significant advancement over conventional photothermal models that primarily consider heat input while neglecting the influence of water-state thermodynamics, demonstrating that molecular-level control of hydrogen-bonding environments can directly influence macroscopic evaporation behaviour.

Collectively, these findings indicate that the superior performance of recent hydrogel systems arises from the synergistic integration of interfacial heat localization, optimized capillary-driven water transport, and molecular-level control of water states, particularly intermediate water. However, despite these advances, many high-performance systems still rely on empirical optimization without quantitatively resolving the relative contributions of these mechanisms, and important trade-offs between thermal management, water transport, and structural stability remain insufficiently understood. Addressing these challenges will require a shift toward mechanistically guided design frameworks that integrate thermodynamics, transport phenomena, and interfacial science, thereby enabling the development of scalable, efficient, and robust hydrogel-based evaporation systems for real-world applications.

6.5. Emerging and Advanced Applications of Hydrogel-Based Photothermal Systems

6.5.1. Photothermal–Photocatalytic Energy and Water Conversion (Integrated Perspective)

Hydrogel-based photothermal systems are rapidly evolving from single-function solar evaporators into integrated photothermal–photocatalytic platforms capable of simultaneous energy and water conversion.

Recently, floatable photocatalytic hydrogel nanocomposites have been developed that operate at the air–water interface to improve light harvesting and gas separation efficiency and increase mass transport speed, thus achieving a high-rate of solar hydrogen production under saline and turbid conditions with good stability. The hydrogel matrix is important in stabilizing photocatalysts, preventing nanoparticle migration, increasing the efficiency of the interface reaction, and thus addressing the deficiencies of traditional slurry-based photocatalytic systems^[162].

As shown in **Table 4** of the previous designs (2018–2022), a passive porous network was used for water transport; however, these next-generation hydrogels actively regulate the hydrogen-bonding environment to promote the generation of intermediate water^[198-199]. Therefore, this control reduces the effective heat of vaporization and improves the accessibility of reactants at photocatalytic sites, enhancing evaporation and catalytic efficiency. Recent studies (2024–2026) has been made in molecularly engineered hydration structures that have fundamentally changed the functionality of hydrogel matrices in such systems, as shown in **Table 5**.

At the same time, the dynamic water microskin effect (2025) offers a new mechanism for interfacial processes. A self-

Table 4. Recent development of Biopolymer and carbonaceous material-based composites.

Sr. No.	Bio-degradable material	Carbonaceous material	Method	Types of Water	Evaporation rate (kg m ⁻² h ⁻¹)	Efficiency (%)	Ref.
1	PVA	Coconut shell Biochar	In Situ chemical Cross-Linking	Salt water (brine)	1.89	85.2	[107]
2	Acrylamide	Rice Husk Biochar	facile foaming-polymerization	Salinity seawater	1.77 ± 0.05	NA	[185]
3	Sodium Alginate	Carbon Nano Tubes	Simple Method	Salt water	1.699	86	[53]
4	PVA	Carbon containing Diesel Soot Particles	Synthesis Method	Seawater	3.01	NA	[186]
5	Chitosan/PVA	Carbon Nano Particles	Cross-Linking Method	Steam generation	2.28	NA	[57]
6	Cellulose Nanofiber	Carbon Black	Simple Method/Fabrication Process	Seawater	1.08	NA	[87]
7	Cellulose from Corncob	Carbon Nano Tube	Chemically cross-linking	Industrial waste water	1.52	92	[52]
8	Cellulose	Carbon Dots	NA	NA	1.30	NA	[187]
9	PVA	Carbon Nano Tubes	In Situ chemical cross-linking co-gelation method	Lake water, dye-containing solution, seawater and brine water	2.09	NA	[188]
10	Chitosan/PVA	Carbon in form of Chinese ink	NA	Artificial seawater with different salt concentration, sewage	4.37	98.2	[189]
11	Carboxymethyl Cellulose	Carbon dots	Chemically cross-linking	Saline water, water contaminate with heavy metal ions, detergent, organic dye	1.4	89	[190]
12	PVA	Graphite Powder & Flakes	Synthesis Method	NA	2.5	NA	[191]

Table 4. (Continued)

Sr. No.	Bio-degradable material	Carbonaceous material	Method	Types of Water	Evaporation rate (kg m ⁻² h ⁻¹)	Efficiency (%)	Ref.
13	poly (vinyl alcohol)	tannic acid and iron ions to change from yellow to black	NA	Sewage treatment and desalination in outdoor condition	1.897±0.11	89.7±2.73	[192]
14	polyvinyl alcohol (PVA)	carbonized cattail (CC)	photothermal synergy	Wastewater recovery and seawater desalination	1.66	88.0	[193]
15	Polyaniline (PANI)	Carbon cloth	Facile electrochemical deposition	Seawater desalination and wastewater purification	2.21	98.0	[194]
16	Polystyrene foam	candle soot coated cotton	NA	Sewage water purification	NA	80	[195]
17	Urea	Carbon dots	microwave irradiation technique	Low-cost water purification kits.	1.11	70	[196]
18	PVA	Carbon nanotubes	NA	Industrial waste purification	2.43	95.90	[197]

Table 5. Emerging Hydrogel Design Strategies for Enhanced Solar Evaporation: From Molecular Hydration Engineering to Multifunctional Energy Systems (2024–2026)

Sr. No.	Polymer Matrix	Functional Material	Method	Water Type	Evaporation Rate (kg m ⁻² h ⁻¹)	Efficiency (%)	Key Mechanism/Innovation	Ref.
1	Engineered hydrogel	Molecular hydration networks	Molecular design	Seawater	~2.5–3.5	> 90	Intermediate water engineering (enthalpy reduction)	[200]
2	Hydrogel interface	Dynamic water microskin	Interfacial engineering	Seawater	~3.0+	> 90	Self-adaptive evaporation interface	[63]
3	Polymer hydrogel	Liquid metal (Ga-based)	Mechanochemical synthesis	Seawater	>3.0	NA + electricity	Coupled water–electricity generation	[201]
4	Hybrid hydrogel	Photocatalyst + carbon	Integrated system	Saline/turbid water	~2.5	NA	Photothermal–photocatalytic coupling (H ₂ production)	[202]

regulating, ultra-thin water layer at the evaporation interface continuously reconfigures in response to solar irradiation to enhance the rate of water replenishment and improve charge and mass transport in photocatalysis. This is a clear advancement over traditional capillary-driven transport models, which dominate earlier literature.

Together, these changes show that the development of recent hydrogel systems is no longer limited to the selection of photothermal materials, but rather includes the synergistic combination of hydration thermodynamics, interfacial water dynamics, and catalytic function, and hydrogel architectures are emerging as scalable platforms for integrated solar hydrogen production and water purification.

6.5.2. Solar-Driven Desalination Coupled with Electricity Generation (Multifunctional Systems)

Research has recently shown that photothermal hydrogels can produce freshwater and generate electricity in situ at the same time to build multifunctional water–energy systems. In particular, liquid-metal-embedded hydrogels (2025) have been synthesized through polyphenolic mechanochemistry, demonstrate strong broadband light absorption, improved thermal conductivity and efficient interfacial evaporation, and have reached evaporation rates higher than those of most previous liquid-metal systems under one-sun irradiation [163].

In addition to the above thermal characteristics, these systems are also capable of generating a relatively stable power source by evaporation-induced ion transport in the hydrogel network for dual functions of water purification and energy harvesting. In this way, a new type that has not been seen in the

previous carbon-based photothermal systems (2018–2022) has been introduced; these systems have only been used for heat generation and lack intrinsic electrochemical functions.

Molecularly engineered hydration networks have also been introduced into these systems to enhance both the mobility of ions and water-transporting paths, and thus regulate the hydration-state to achieve a dual effect of evaporation and electrokinetic processes. Compared to traditional CNT- or graphene-based hydrogels, liquid-metal systems provide:

- Coupled thermal–electrical transport pathways
- Strengthened interfacial charge separation
- Optimized system-level energy utilization

Critically, the above developments show that liquid-metal hydrogels should not be regarded only as new materials but rather as next-generation platforms that can break the limitations of traditional photothermal designs in terms of multifunctionality and energy integration. Therefore, hydrogel-based systems are expected to be applied in resource-scarce areas.

6.5.3. Resource and Energy Recovery from Wastewater Systems (Toward Circular Integration)

Beyond solar-driven desalination, hydrogel-inspired, water-rich interfacial systems are increasingly being explored for resource and energy recovery from wastewater, particularly in bio electrochemical and hybrid treatment platforms. Pilot-scale bioelectrochemical reactors demonstrate that porous, hydrated architectures significantly enhance mass transfer, microbial activity, and hydrogen recovery from organic-rich waste streams, underscoring their scalability and industrial relevance [164].

Recent advances (2024–2026) further extend these capabilities by incorporating dynamic water structuring and molecular hydration control into wastewater treatment systems^[203–204]. The integration of the microskin effect enables improved interfacial reaction kinetics, while engineered hydration networks regulate ion diffusion and substrate accessibility, addressing key limitations of earlier static porous materials^[205].

In addition, emerging hydrogel systems exhibit mechanically adaptive, self-healing, and shock-resistant properties, overcoming durability challenges associated with earlier designs (2018–2022) that were prone to structural degradation under prolonged operation. These features are critical for real-world wastewater environments, where mechanical stability and long-term performance are essential.

When integrated with photothermal and photocatalytic functionalities, these systems enable a closed-loop framework combining:

- Solar-driven desalination
- Hydrogen production
- Wastewater treatment
- Resource recovery (e.g., nutrients and energy)

This convergence aligns with circular-economy principles, transforming hydrogel-based systems from passive treatment technologies into active platforms for energy and resource valorisation.

Despite these promising developments, challenges remain in scaling such systems, particularly in maintaining hydration-state stability, preventing fouling, and ensuring long-term operational reliability. Future research should focus on bridging molecular-level hydration design with system-scale engineering, enabling practical deployment in complex real-world environments.

7. Challenges and Future Directions

While the laboratory-scale success of hydrogel photothermal evaporators—often measured against the benchmarking criteria introduced in Section 2.2—is well-documented, bridging the gap between experimental prototypes and real-world industrial deployment involves addressing several critical technical and economic barriers. The record-breaking evaporation rates reported in literature frequently overlook the long-term degradation of these metrics when exposed to real-world stressors. Addressing the following technical and economic barriers is critical for moving beyond idealized performance:

i) Long-term stability and fouling in real wastewater environments:

A major limitation in current literature is the reliance on short-term testing using purified or simple saline solutions. In practical wastewater treatment, the presence of oils, surfactants, and microorganisms leads to rapid biofouling. This organic accumulation obstructs the internal porous architecture and passivates the photothermal surface, causing a steady decline in evaporation efficiency. Integrating self-cleaning mechanisms, such as antibacterial polymers or zwitterionic coatings, is essential for maintaining performance in contaminated water sources^{[90][191]}.

ii) Salt accumulation and crystallization in high-salinity brines:

High-salinity brine treatment remains a significant hurdle

due to inevitable salt crystallization. Although advanced designs leveraging Marangoni flow or directional macro-channels have improved salt rejection, a performance gap often exists between salt-resistance and optimal thermal localization. Over time, salt buildup not only interferes with solar harvesting but can also induce localized mechanical stress, leading to the structural failure of the polymer matrix^[206–207].

iii) Mechanical degradation under continuous operation:

Due to their high water content, hydrogels are naturally soft and prone to mechanical fatigue caused by repetitive swelling and deswelling cycles. Ensuring that these materials can withstand harsh outdoor conditions—including intense UV radiation, wind, and fluctuating temperatures—is a primary engineering concern. Utilizing double-network architectures or hybridizing with inorganic reinforcements may provide the necessary durability for long-term outdoor use^[208–209].

iv) Scalability and cost of fabrication:

The commercial viability of many high-efficiency hydrogels is restricted by expensive precursors like noble metal nanoparticles or specialized carbon nanotubes. To compete with established desalination technologies, research must pivot toward low-cost, biomass-derived substrates and scalable manufacturing processes, such as 3D printing or continuous roll-to-roll fabrication^[210–211].

v) Disposal and end-of-life management of composite materials:

The ecological impact of disposing of spent composite hydrogels, particularly those containing non-degradable additives, is often overlooked. To fulfill the promise of a circular economy, future designs must adopt a "cradle-to-grave" philosophy, prioritizing the use of fully biodegradable scaffolds and eco-friendly photothermal agents to prevent secondary environmental contamination^{[57][212]}.

vi) Integration of multifunctional systems and system-level complexity:

Recent developments have demonstrated hydrogel platforms capable of integrating multiple functions, including solar desalination, hydrogen production, electricity generation, and wastewater treatment. However, combining these processes into a single system introduces significant complexity due to competing transport phenomena involving heat, mass, ions, and electrons. Additionally, maintaining stability and efficiency under fluctuating environmental conditions remains challenging. Future research must move beyond material-level innovation toward system-level design and optimization, ensuring that multifunctional systems can operate reliably and efficiently in real-world scenarios^[201].

vii) Limited understanding of water thermodynamics and interfacial mechanisms:

Although concepts such as intermediate water, molecular hydration engineering, and the dynamic micro-skin effect have emerged as key breakthroughs, their underlying mechanisms are not yet fully understood. Current studies lack comprehensive characterization of water states beyond basic techniques such as DSC, and the relationships between hydrogen bonding, evaporation enthalpy, and transport kinetics remain unclear. Furthermore, the stability and dynamics of interfacial water layers under realistic conditions are poorly understood.

Addressing these gaps requires advanced experimental techniques and theoretical modelling to establish robust structure–property–performance relationships^[213–214].

viii) Bridging molecular design with real-world deployment:

A critical challenge lies in translating recent molecular-level innovations into practical technologies. While engineered hydration networks and interfacial phenomena have demonstrated significant performance improvements in laboratory settings, their scalability, robustness, and reproducibility under real-world conditions remain uncertain. Future research should focus on linking molecular design principles to macroscopic system performance, developing hierarchical structures that integrate micro- and macro-scale transport processes, and validating system performance under realistic environmental conditions. Bridging this gap is essential for enabling the practical implementation of next-generation hydrogel-based water–energy systems^[215].

8. Conclusion

Ecological hydrogel composites have redefined sustainable water purification by bridging the gap between high-performance thermodynamics and environmental responsibility. By leveraging intermediate water states and hierarchical bio-architectures, these materials achieve efficiencies previously unattainable. Future strategic efforts should focus on the scalability of green fabrication methods (e.g., 3D printing) and the development of fully biomass-derived photothermal agents to ensure a circular economy for global water security.

In the face of water purification and resource scarcity, research has started to apply sustainable, natural and biodegradable materials in solar-driven evaporation systems that reduce environmental pollution. These materials are economically feasible, environmentally friendly and all-round use. Some serious problems have still occurred in this area recently. New materials such as CNT-enhanced cellulose hydrogels (CSG) and CMH-HgSx have been developed that are promising for solar-driven water purification and desalination. CSG can handle tough conditions, and CMH-HgSx is a versatile solar absorber. They both show real potential for sustainable freshwater production. CMH-CDs-SH is suitable for removing heavy metal ions, and PSCH-20 can be used to purify water efficiently from all kinds of sources. Research on PSCH-x hydrogel absorbers has provided support for the design and performance improvement of water treatment, such as water purification and desalination. The above high-tech materials can help to solve the problem of water pollution, and more research will be carried out to promote their applications in practice. Management of the secondary waste produced during the application of these materials must also be implemented properly. Reducing waste generation and handling it reasonably is crucial to keep the ecological benefits of these solutions.

In conclusion, the field of biopolymer and carbonaceous-based hydrogels and their applications has gone much further than what has been achieved so far. There are opportunities to enhance their properties of the above materials for water treatment by adding new functional materials through existing and new methods. Research on new hydrogel technologies for environments and the economy has shown good prospects.

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Conflicts of Interest

The authors declare no conflict of interest.

Abbreviations

PVA: Poly (vinyl alcohol)
 PHAs: Polyhydroxyalkanoates
 PBS: Polybutylene Succinate
 PCL: Polycaprolactone
 PLA: Polylactic Acid
 PGA: Polyglycolic Acid
 PANI: Polyaniline
 PNIPAM: Poly(N-isopropylacrylamide)
 SA: Sodium alginate
 CC: carbonized cattail
 CNTs: Carbon Nano Particles
 CDs: Carbon Dots
 CNT: Carbon Nano Tube
 THMs: Trihalomethanes
 FTIR: Fourier-Transform Infrared Spectroscopy
 SEM: Scanning Electron Microscopy
 TEM: Transmission Electron Microscopy
 DSC: Differential Scanning Calorimetry
 HR-TEM: High Resolution-Transmission Electron Microscopy

Author Contributions

Conceptualization, B.N.P., P.R.D., S.P.P. and A.H.G.; methodology, B.N.P., P.V.D., P.R.D., P.S.B., C.S.P., A.L.P., S.P.P. and A.H.G.; formal analysis, A.H.G., A.L.P. and S.P.P.; investigation, B.N.P., S.P.P., A.H.G.; resources and data curation, B.N.P., S.P.P. and A.H.G.; writing-original draft preparation, B.N.P., P.R.D., P.V.D.; writing-review and editing, B.N.P., P.R.D., S.P.P. and A.H.G.; visualization, B.N.P., P.R.D., S.P.P. and A.H.G.; supervision, S.P.P. and A.H.G.; project administration, B.N.P., P.R.D., S.P.P. and A.H.G. All authors have read and agreed to the published version of the manuscript.

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