A Review on Multi-Scale Structural Design of Graphene-based Photothermal Materials for Solar Interfacial Evaporation

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Abstract: With global water scarcity and environmental pollution becoming increasingly serious, solar interface evaporation technology has become a key path to solve the freshwater crisis due to its advantages such as renewability and zero carbon emissions. Owing to their broad-spectrum light absorption, graphene and its derivatives exhibit remarkable potential in photothermal conversion applications, high thermal conductivity, as well as readily tailored surface characteristics. However, a single component is difficult to meet the comprehensive needs including fast evaporation kinetics, resistance to salt crystal formation, and long-lasting structural reliability. Through multi-scale structural design such as macroscopic three-dimensional configuration (aerogel, hydrogel, membrane structure), microscopic pore engineering (graded pores, vertical orientation pores), and surface chemical modifications (affinity/hydrophobic modification, functional group introduction, and charge regulation), the synergistic regulation of light, heat and quality can be realized, and the evaporation efficiency and environmental adaptability can be significantly improved. The present review comprehensively discusses the structural design strategies and performance regulation mechanisms for graphene-derived photothermal materials, discusses the structure-activity relationship and the principle of multi-scale synergy, and aims to lay theoretical groundwork and supply technical references for high-performance solar interfacial evaporation material development.

Keywords: Graphene; Photothermal Materials; Solar Interface Evaporation; Multi-scale Structural Design; Water Purification

1. Introduction

In solar-driven interfacial evaporation, thermal energy is confined to the water–air interface, which effectively improves energy utilization and offers a practical route for freshwater production. Graphene and related materials are considered promising photothermal candidates owing to their excellent optical absorption, thermal conductivity, and chemical tunability. Nevertheless, pure graphene materials often struggle to balance high evaporation performance, salt resistance, and operational stabilityIn recent studies, simply improving intrinsic photothermal properties has approached its limit. Instead, multi-scale structural design has become an effective approach. By adjusting macroscopic shapes, microchannel structures, and surface characteristics, researchers can optimize light absorption, heat management, water supply, and anti-fouling ability simultaneously. The present work examines developments in graphene-derived photothermal materials across three dimensions: macroscopic configuration, microscopic pore structure, and surface chemistry.

2. Research Background and Value Proposition of Graphene-based Photothermal Materials

2.1 Energy and Environmental Requirements of Solar Interface Evaporation Technology

With the increasing global water scarcity and alongside growing environmental pollution, there is an urgent demand for advanced, eco-friendly water purification solutions. Solar interface evaporation technology is deemed among the key paths to mitigate the freshwater crisis and treat wastewater with high salt/organic pollution by virtue of its advantage in energy efficiency, zero carbon emissions, and distributed water supply due to its use of

renewable solar energy sources to directly drive water phase transitions. [1]In recent years, a variety of photothermal materials have been used to construct solar evaporators, and the evaporation rate generally exceeds 2.0 $kg \cdot m^{-2} \cdot h^{-1}$ under 1 kW/m^2 (i.e., 1 sun) standard light, and even reaches 4.06 $kg \cdot m^{-2} \cdot h^{-1}$[2] or higher in some systems[3], significantly exceeding the theoretical limit of traditional evaporators (1.47 $kg \cdot m^{-2} \cdot h^{-1}$)[4]. In addition, this technology has shown multifunctional integration potential in the fields of seawater desalination and heavy metal elimination, microplastic retention, as well as organic compounds pollutant degradation[1,2,5,6]. For example, the self-floating evaporator based on MXene/silver nanowires has a microplastic removal rate of 99% and antibacterial properties[2]. The nitrogen-doped honeycomb carbon nanosheet evaporator can simultaneously achieve efficient degradation of methyl blue and phenol[6]. These advances highlight the comprehensive value of solar interface evaporation technology in addressing complex water quality challenges, and also put forward higher requirements for its core photothermal materials, they are required to possess high photothermal performance, along with good structural stability, pollution resistance, and environmental adaptability.

2.2 Advantages of Intrinsic Photothermal Characteristics of Graphene Materials

Graphene-based materials, including graphene oxide and reduced graphene oxide show significant advantages in the domain of solar interface evaporation on account of their unique electronic band structure and optical properties. Its planar two-dimensional conjugated π-electron network provides excellent broadband spectral absorption performance, especially across the visible-near-infrared spectral region, with a high extinction coefficient, which can achieve a solar absorption rate across the full spectrum of up to 99.0%[7]. Experiments have shown that a composite evaporator containing only about 10.0% non-oxide graphene sheets can achieve more than 98% sunlight absorption[8]. In addition, Graphene is endowed with extraordinary thermal conduction performance (theoretical value of 5000 $W \cdot m^{-1} \cdot K^{-1}$), which is conducive to the rapid conduction within the material, with heat localized at the evaporation interface, thereby improving energy utilization efficiency[9]. What's more, graphene can be chemically modified, doped or compounded with other functional components to further optimize its photothermal properties. For example, modifying polydopamine on LIG paper substrates can increase the conversion efficiency of solar energy to 125.1%[10]. The PPy-VO-GO hybrid structure achieves a photothermal conversion efficiency of 92% and an evaporation rate of 1.9 $kg \cdot m^{-2} \cdot h^{-1}$ at 1 sun [11]. Compared with 2D films, 3D vertically oriented graphene arrays not only enhance light absorption, but also increase the evaporation rate by 3 times, and effectively promote the crystallization of high-salt solutions[12]. These properties make graphene an ideal platform for building high-performance photothermal materials.

However, when compared versus other representative photothermal materials like MXene, carbon nanotubes, and plasmonic materials, graphene still shows obvious limitations in practical applications, especially in terms of cost, structural stability, and large-scale production.High-quality monolayer graphene is usually obtained through chemical vapor deposition or mechanical exfoliation, which leads to high preparation cost and limits its large-scale use.Although reduced graphene oxide is easier to produce, it contains abundant structural defects that weaken its intrinsic photothermal and thermal conductive properties.By contrast, MXene exhibits strong localized surface plasmon resonance and favorable hydrophilicity, but it is prone to oxidation and structural degradation under long-term light irradiation or high-salt conditions, resulting in poor long-term stability.Carbon nanotubes have high thermal conductivity, but they tend to agglomerate severely during processing, and their purification and dispersion costs are relatively high.Plasmonic materials like gold and silver show intense light absorption at specific wavelengths, but their high cost, easy thermal sintering, and narrow absorption band greatly restrict their durable and universal application.Among these candidates, graphene still presents a more balanced performance with broad-spectrum light absorption, robust chemical stability, and mature fabrication routes.Nevertheless, its inherent drawbacks cannot be thoroughly solved by simply adjusting intrinsic properties.Instead, multi-scale structural design, including macroscopic 3D assembly, microscopic pore engineering, and surface

chemical regulation, has become an indispensable strategy to compensate for these weaknesses and further upgrade the comprehensive photothermal evaporation performance.

2.3 The Scientific Significance of Multi-Scale Structural Design for Performance Regulation

Therefore, the synergistic regulation of light, heat and mass through multi-scale structure design has become the core strategy to improve the performance of graphene-based photothermal materials. At the macroscopic scale, aerogels, hydrogels, membranes, and bionic 3D configurations can expand the light capture area, enhance the thermal local effect, and optimize the water transport path. For example, the evaporation flux of the three-dimensional graphene oxide structure reaches 34.7 $kg \cdot m^{-2} \cdot h^{-1}$ by increasing evaporation surface area index (EAI=70), which achieves nearly twenty times the value of conventional two-dimensional evaporators[13]. The pyramidal array structure imitating durian skin still maintains high evaporation stability under oblique light and wind conditions [14]. At the microscopic scale, the regulation of hierarchical pores, vertical orientation channels and pore size distribution directly affects the steam escape resistance and salt resistance. A 3D-printed graded porous rGO/CB evaporator achieved a high evaporative flux of 10.5 $kg \cdot m^{-2} \cdot h^{-1}$ for 10 wt% NaCl saline water under 8 $kW \cdot m^{-2}$ light and had excellent salt resistance[3]. The vertically arranged graphene/cellulose nanofiber structures achieve efficient transport through directional water channels[8]. At the surface chemical level, hydrophili/hydrophobic modification, functional group introduction and charge regulation can adjust the evaporative energy barrier and anti-fouling performance at the interface. For example, graphene/TiO_2 aerogel can operate stably in 25 wt% NaCl for 100 hours by constructing an internal and external bizone heterogeneous wetting structure[15]. The honeycomb structure fabric induces the Marangoni effect, achieving local crystallization and self-dissolution of salts[16]. In conclusion, multi-scale structural design provides a systematic solution to break through the bottleneck in the field of solar-driven interfacial evaporation performance by coordinating the regulation of light harvesting, thermal control, water transport, and interfacial behavior[9,17,18].

3. Regulation of Photothermal Performance by Macroscopic Three-Dimensional Configuration Design

3.1 Broadband Light Absorption and Thermal Local Effects of Aerogel Structures

As a typical three-dimensional porous macrostructure, aerogel shows excellent broadband light absorption capacity and thermal localization effect in graphene-based photothermal materials. For example, 3D porous graphene films (forest-like laser-induced graphene) based on forest-like structures have a full solar spectral absorption rate of up to 99.0%, which can heat up to 87.7 °C in just 30 seconds under simulated sunlight irradiation and finally reach an equilibrium temperature of 90.7 ± 0.4 °C. In addition, the composite aerogel constructed by bidirectional freeze-casting using non-oxide graphene sheets (NOGF) and cellulose nanofibers (CNF) achieved a sunlight absorption rate of more than 98% when the NOGF content was only 10.0%, and the solar vapor production rate reached 2.39 $kg \cdot m^{-2} \cdot h^{-1}$, and the energy conversion ratio reached 93.7%[8]. Similarly, the polyelectrolyte/polypyrrole (PPC) photothermal aerogel achieved an evaporation velocity of 1.41 $kg \cdot m^{-2} \cdot h^{-1}$ and a solar-driven steam conversion efficiency of 96.9% in vertical operation mode under low pyrrole concentration (PPC10) due to its high light absorption rate, thermal localization effect, and open porous structure[19]. These studies show that the 3D network of aerogels not only provides rich light scattering paths to enhance light capture, but also achieves efficient thermal localization by limiting the heat conduction path, thereby significantly improving the overall photothermal conversion performance.

3.2 Water Supply-evaporation Synergy Mechanism of Hydrogel Networks

The hydrogel network structure effectively coordinates the water supply and interface evaporation process through its hydrophilic three-dimensional skeleton and continuous water channel, which has become one of the key strategies to improve the efficiency of photothermal evaporation. For example, the multi-scale ordered hydrogel photothermal evaporator (MHH) with a gill structure is

supported by ultra-long hydroxyapatite nanowires, combined with the MXene absorbing layer, and the evaporative flux of pure water reaches up to 6.278 kg/m^2·h and 4.931 kg/m^2·h for actual seawater under 1 kW/m^2 of light, while reducing the water evaporation enthalpy to 1113 J/g, which is markedly better than the traditional evaporation system[20]. Another study developed a PCR hydrogel (composed of PVA, cellulose nanocrystalline CNC, and reduced graphene oxide RGO) with a three-dimensional porous structure, achieving 95% light absorption in the 250–2500 nm band, an evaporative flux of 1.83 kg·m^{-2}·h^{-1} when processing 3.5 wt% NaCl solution, and a stable performance of 1.88 kg·m^{-2}·h^{-1} in industrial black liquor, showing excellent water supply capacity and salt resistance[21]. In addition, the bio-inspired unidirectional silk fibroin (SF)-AgS/AgPO three-dimensional evaporator increased the water evaporation rate to 2.86 kg·m^{-2}·h^{-1} (1 sun) through a directed water transport mechanism and achieved a VOCs removal rate of 91.9% in phenol-contaminated water[22]. The above results show that the hydrogel network can effectively alleviate the water supply bottleneck at the evaporation interface by constructing a continuous and rapid water transport channel, and realize the dynamic coordination of water supply and evaporation.

3.3 Regulation Strategies for Interfacial Wettability of Membrane Structures

The interfacial wettability of the membrane structure directly affects the distribution, evaporation kinetics and salt crystallization resistance of water molecules at the evaporation interface, so it has become a key regulatory dimension in macrostructure design. For example, graphene/titanium dioxide aerogel (GTA) has been prepared by freeze-drying and thermal reduction to achieve an evaporative flux of 1.52 kg·m^{-2}·h^{-1} and a power conversion efficiency of 91.02% under 1 sun of light, and can dynamically regulate the crystallization-dissolution process of salts, and can run stably in 25 wt% NaCl high salt solution for 100 hours[15]. On the other hand, the broccoli-like biomimetic multi-level structure nickel black@graphene (Ni@Gr) film significantly reduced the evaporation enthalpy to 1343.6 kJ/kg, and the evaporation rate was as high as 2.05 kg·m^{-2}·h^{-1} at 1 sun, breaking the theoretical threshold (1.47 kg·m^{-2}·h^{-1})[4]. In addition, the Janus solar evaporator based on recycled waste paper and tea residue forms an asymmetric wetting structure by introducing MXene and carbon nanotubes, achieving an evaporative flux of 1.52 kg·m^{-2}·h^{-1} and a transformation efficiency of 90.79%, and has excellent salt repulsion[23]. It is worth noting that although forest-like LIG films are superhydrophobic, they can still achieve efficient desalination, indicating that the regulation of wettability needs to be optimized in combination with specific application scenarios[7]. In conclusion, by accurately designing the hydrophili/hydrophobic region distribution or gradient wetting structure of the membrane surface, the interfacial water behavior can be effectively regulated, the evaporation efficiency can be improved, and the long-term operation stability can be enhanced.

3.4 Structure-Effect Relationship between Three-dimensional Configuration and Evaporation Efficiency

A large number of studies have shown that the design of the three-dimensional macroscopic configuration directly determines the evaporation efficiency of photothermal materials, and its structure-activity relationship is reflected in multiple dimensions such as light capture ability, thermal management performance, water transport efficiency and steam escape kinetics. For example, compared with 2D graphene films (2D-G), 3D vertically oriented graphene films (3D-G) have better light harvesting and heat transformation properties, resulting in a three-fold increase in water evaporation rate[12]. Similarly, the three-dimensional conical structure formed by folding paper-based LIG/PDA materials not only elevates the effective evaporative surface area, but also optimizes the light absorption and heat localization, achieving an evaporative flux of 2.02 kg·m^{-2}·h^{-1} and a utilization level of 125.1% in one sun, which is significantly better than its two-dimensional counterpart[10]. Furthermore, the evaporation rate of the heterostructured graphene-based evaporator constructed by 3D printing technology is as high as 17.9 kg·m^{-2}·h^{-1} in seawater at a wind speed of 2 m/s, far exceeding that of freshwater systems, proving that the complex three-dimensional configuration can further amplify the evaporation advantage of environmental factors such as wind fields[24]. In addition, the

biomimetic mushroom-type evaporator (BMSSG) integrates self-flotation gel and wood moisture transport channels, achieving an outstanding evaporative performance of 1.67 $kg \cdot m^{-2} \cdot h^{-1}$ without additional insulation, demonstrating the dual value of structural integration for system simplification and performance improvement [25]. Taken together, the coupling efficiency of light-heat-mass transfer is determined by multi-scale synergy, including macrogeometry, pore orientation, and interface wetting gradient, which is the core design principle for achieving high-performance solar interface evaporation[9,26].

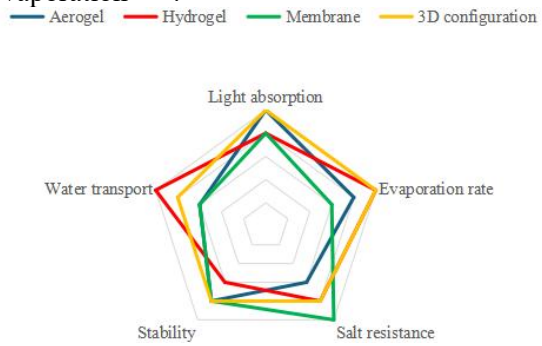

Figure 1. Performance Benchmark Radar Chart of Typical Macroscopic Structures for Graphene-based Photothermal Materials

To intuitively compare the comprehensive properties of different macroscopic configurations, a performance benchmark radar chart was established, as shown in Figure 1. Aerogels exhibit outstanding advantages in light absorption and thermal localization. Hydrogels are superior in water transport and evaporation rate due to their continuous hydrophilic network. Membrane structures present the best salt resistance through wettability regulation. 3D configurations show balanced and excellent performance in all dimensions, indicating strong comprehensive application potential. The radar chart clearly reflects the structure–activity relationship and provides a reference for structural selection and optimization in practical applications.

4. Microscopic Channel Engineering and Mass Transfer Performance Optimization

4.1 Influence of Graded Hole Structure Design on Light Scattering and Vapor Escape

The graded pore structure plays a key role in regulating light absorption and vapor transport in graphene-based photothermal materials. By constructing micro-nanometer multi-scale interconnect channels, light reflection can be effectively reduced and light capture capabilities can be enhanced, thereby improving the overall photothermal conversion efficiency. For example, forest-like LIG has achieved a full-band sunlight absorption rate of up to 99.0% due to its graded pore structure and vertically oriented channels, and heats up to 87.7 °C within 30 seconds, significantly improving the photothermal response speed and evaporation performance[7]. Similarly, three-dimensional graphene aerogels construct hierarchical pore structures through the synergistic interaction of bubbles and nanoparticle templates, which not only increase the specific surface area and interface area, but also optimize the vapor escape path, thereby improving the mass transfer efficiency[27]. In addition, nickel black@graphene (Ni@Gr) membrane with broccoli-like morphology uses a biomimetic hierarchy structure to elevate the solar irradiation absorption level to 90.36%, and breaks the theoretical vaporization rate limit by mitigating the evaporation latent heat (1343.6 kJ/kg), delivering an excellent interfacial evaporation performance of 2.05 $kg \cdot m^{-2} \cdot h^{-1}$ under one sun light[4]. These studies show that the graded pore structure effectively synergizes the photothermal energy transformation efficiency and vapor release by regulating diffuse light scattering behavior and vapor diffusion resistance.

4.2 Fast Water Transport Mechanism of Vertically Oriented Holes

The vertical orientation channel provides a low-resistance channel for fast water diffusion from the bulk phase reaching the evaporation region interface, which is the core structural feature of efficient solar interface evaporation. Studies have shown that the tree-like vertical channel structure of non-oxide graphene/nanocellulose (NOGF/CNF) composite aerogels constructed by bidirectional freeze-casting significantly promotes the longitudinal transport of water, achieving a distillation rate of 2.39 $kg \cdot m^{-2} \cdot h^{-1}$ and an energy conversion efficiency of 93.7% with only 10.0% NOGF[8]. Similarly, compared with traditional 2D graphene films (2D-G), 3D vertically oriented graphene arrays (3D-G) not only show improved photothermal transformation capability, but also increase the water evaporation rate by up to 3 times, which is attributed to the optimization of the mass transfer process by its

directional pores[12]. In addition, gelatin/cellulose nanofiber-based aerogels allow for efficient exchange of ions between channels by modulating the sidewall pore structure of vertical channels, thereby maintaining a high evaporation rate of 2.20 $kg \cdot m^{-2} \cdot h^{-1}$ in 3.5 wt% brine and achieving long-term operational stability[28]. These results show that the vertical orientation channel significantly reduces the water transport resistance by constructing a continuous and penetrating water transport network, and provides a structural guarantee for stable operation in high evaporation rate and high salinity environment.

4.3 Correlation Between Pore Size Distribution and Salt Crystallization Resistance

Proper distribution of pore size is crucial for inhibiting salt crystallization and maintaining long-term evaporation stability. Studies have shown that the 3D printed hierarchical porous rGO/CB (3DP-HP rGO/CB) evaporator containing numerous low-resistance channels for salt ion delivery not only realizes a remarkable evaporative flux of 10.5 $kg \cdot m^{-2} \cdot h^{-1}$ under 8 $kW \cdot m^{-2}$ light, but also can operate in 10 wt% NaCl solution for a long time without salt clogging, and its excellent salt resistance comes from the effective promotion of salt ion diffusion by the graded pores[3]. Similarly, the multilayer graphene oxide-based evaporator is designed with a biomimetic gradient pore structure to continuously work for salt-free crystallization for more than 9 hours while maintaining an evaporation rate of 3.28–4.51 kg m^{-2} h^{-1} and an energy use efficiency of up to 80%[29]. In addition, CNF/PVA/PEI@ polypyrrole (CPP) aerogels maintained an evaporation performance of 1.50 $kg \cdot m^{-2} \cdot h^{-1}$ in 20 wt% high-salinity brine due to their layered porous hierarchical structure and vertical transport channels, further verifying the key role of pore size distribution in salt resistance[30]. In conclusion, by regulating the size distribution of pores, especially the introduction of macropores as rapid diffusion channels for salt ions and microporous/mesoporous as buffer areas, salt crystallization can be effectively delayed or avoided, so as to ensure the long-term efficient operation of graphene-based photothermal materials in high-salt environments.

5. Surface Chemical Modification and Interface Behavior Regulation

5.1 Regulation of the Energy Barrier at the Evaporation Interface by affinity/Hydrophobic Modification

The surface wettability of graphene-based photothermal materials has a decisive impact on their properties during evaporation at the solar interface. By regulating the hydrophilic/hydrophobic properties of the surface of the material, the adsorption, diffusion and vaporization behavior of water molecules at the evaporation interface can be effectively adjusted, so as to reduce the energy barrier during the phase transition process. The results show that the hydrophilic surface is conducive to rapid replenishment of water to the photothermal region and maintain continuous evaporation. The moderate hydrophobic region helps the steam escape quickly and avoids heat loss due to local supersaturation. For example, the introduction of hydrophilic polymers such as gelatin methacrylate (GelMA) on the surface of graphene oxide (GO) besides optimizing material hydrophilicity, this structure greatly promotes photothermal conversion performance (from 11.8% to 47.6% of pure GO)[31]. In addition, by constructing composite structures with gradient wettability, such as xylan or alginate-modified copper sulfide nanoparticles into chitosan matrix, water transport and interfacial evaporation kinetics can be optimized at the same time, and the photothermal performance can be improved by 1.4 times compared with unmodified systems [32]. These results show that precise regulation of surface hydrophilicity/hydrophobicity is a key strategy to reduce the energy barrier at the evaporation interface and improve the overall evaporation efficiency.

5.2 Functional Groups Introduce Enhanced Photothermal Conversion Efficiency Mechanisms

The type and density of surface functional groups of graphene materials directly affect their light absorption capacity and non-radiative relaxation path, which in turn regulates the photothermal conversion efficiency (PCE). Reduced graphene oxide (rGO) restores its conjugated structure by removing some oxygenated functional groups, and its photothermal conversion efficiency is twice that of graphene oxide (GO)[33]. Further studies have

shown that the introduction of specific functional groups can enhance PCE by enhancing the non-radiative recombination of photogenerated carriers or promoting local surface plasmon resonance effects. For example, after compounding octadecyl alcohol in conjugated microporous polymer hollow microspheres with graphene coating (GCMPs-HS), the material maintains a light absorption rate of about 90% in the visible light region, and the photothermal conversion efficiency is as high as 87.15%[34]. In addition, the covalent conjugation of the photosensitizer IR780 to the surface of rGO and coated with hyaluronic acid (HA) not only improves the absorption of the material in the near-infrared region, but also significantly enhances the therapeutic effect through synergistic photothermal/photodynamic effects[33]. It is worth noting that some functional groups can also participate in the reduction process through photochemical pathways, such as GO under visible light, and its oxygen-containing functional group can be reduced through photochemical dissociation, which can occur at lower than the traditional thermal reduction temperature, indicating that the functional group itself can participate in energy conversion as a light response unit[35]. Therefore, appropriate tailoring of surface functional groups constitutes a practical method to elevate the photothermal traits of graphene-structured materials

5.3 Correlation between Surface Charge Regulation and Antifouling Performance

The surface charge state not only affects the dispersion stability of graphene-based materials in the aqueous environment, but also directly correlates with their resistance to biological pollution and inorganic salt crystallization. Negatively charged surfaces can inhibit the attachment of negatively charged microorganisms or colloidal particles through electrostatic repulsion, thereby improving the stability of materials in long-term operation. For example, alginate-modified copper sulfide nanoparticles carry an anionic charge and form a strong electrostatic interaction with the cationic chitosan matrix, which not only improves dispersion while improving its mechanical performance and photothermal characteristics of the material[32]. Similarly, hyaluronic acid (HA)-modified Fe-MOF-derived nanoparticles (Bi-NC@HA) enhance the accumulation capacity of tumor tissues due to surface negative charges, while reducing non-specific protein adsorption and improving biocompatibility and targeting[36]. In water treatment applications, the introduction of directional covalent modification at the edge of graphene membrane nanopores by pre-anchoring method significantly improves the surface charge density, greatly improves the selectivity of K^+/Cl^- and the conversion performance of osmotic energy, and achieves a volumetric power density of 81.6 W m^{-2} at a 100-fold ionic concentration gradient [37]. In addition, the Ag/GO-GelMA platform not only achieves efficient photothermal conversion through synergy with functional groups on the surface, but also significantly downregulates the inflammatory pathway of MAPK and PI3K-Akt, showing excellent anti-inflammatory and anti-pollution potential[31]. The above evidence suggests that precise regulation of surface charge is an important way to achieve highly stable and anti-pollution photothermal materials.

5.4 Principles of Collaborative Optimization Design of Multi-Scale Structures

Advanced functional performance of graphene-based photothermal composites hinges on multi-scale co-design from macroscopic configuration, microscopic pores to surface chemistry. Single-scale optimization is often difficult to take into account multiple requirements such as optical absorption, thermal localization, water transport and steam escape, while cross-scale integration strategies can achieve functional complementarity and performance multiplication. For example, the composite of a three-dimensional graphene network with FeO nanoparticles possessing localized surface plasmon resonance effect for fabricating ODA@FeO/C-GP phase change materials not only achieves a high photothermal conversion efficiency of 88.18%, but also has excellent heat storage and cycling stability[38]. For example, the GO/Au NPs sandwich structure is constructed by means of layer-by-layer self-assembly, and the multi-level interface between two-dimensional sheets and zero-dimensional nanoparticles is used to achieve wide spectral absorption and photothermal efficiency of 96.7%, and the thermoelectric module is integrated to achieve energy recovery[39]. At the interfacial behavior level, PEG-modified AgS quantum dot probes achieve targeted delivery and high PCE (45.17%)

through surface chemical regulation while maintaining good biocompatibility [40]. The molecular-level hybridization of polydopamine (PDA) and AgS resulted in a combinatorial index (CIs) as low as 0.3–0.7 through the synergy of dual photothermal components, confirming the synergistic enhancement mechanism of organic-inorganic interfaces[41]. On the whole, the core principles of multi-scale collaborative optimization are: macroscopic structure ensures thermal locality and mechanical stability, microscopic pores regulate mass transfer and salt resistance, and surface chemical modification accurately regulates interfacial energy barrier, charge state and biocompatibility. Only by organically integrating the three can graphene-based photothermal materials be applied efficiently, stably, and multi-functionally in the fields of energy, environment, and biomedicine.

The multi-scale structural synergistic design framework is shown in Figure 2, which systematically integrates macroscopic geometry, microscopic mass transfer, and interfacial chemistry to achieve efficient light-heat-mass conversion and stable solar interfacial evaporation.

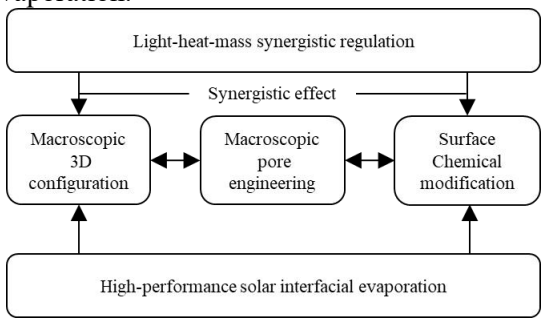

Figure 2. Schematic Diagram of Multi-scale Collaborative Design Framework for Graphene-based Photothermal Materials

6. Conclusion and Development Trends

This review provides a systematic overview of the multi-scale structural engineering of graphene-based photothermal materials utilized in solar interfacial evaporation, and extracts three universal laws underlying cross-scale optimization:Macroscopic 3D configuration determines light-harvesting and thermal localization capacity, which sets the upper limit of photothermal conversion efficiency.Microscopic pore structure dominates water/vapor transport and salt-resistant stability, which ensures long-term operational reliability.Surface chemical modification tailors interfacial energy barrier and anti-fouling ability, which further boosts adaptability to complex water environments.These three levels work synergistically rather than independently, forming a complete structure–performance regulation mechanism for light–heat–mass transfer.Despite the remarkable progress achieved, several critical challenges remain for practical deployment:Precise control of hierarchical structures in large-scale fabrication;Long-term durability under high-salinity, organic-fouling, and complex wastewater conditions;Integration of desalination, pollutant degradation, and resource recovery in one system.

To address these issues, future research can focus on the following specific technical routes:Template-guided precision manufacturing:Using ice-templating, 3D printing, or electrospinning to realize controllable construction of vertically aligned channels and graded pores, ensuring structural consistency in scale-up production.Janus wettability interface engineering:Constructing asymmetric hydrophilic–hydrophobic surfaces to directionalize water supply, accelerate vapor escape, and achieve spontaneous salt crystallization/dissolution without external stimuli.Heteroatom doping & defect engineering:Regulating energy band structure and non-radiative relaxation paths via N, P, S doping or vacancy introduction to further enhance photothermal conversion without increasing material cost.Multi-functional heterostructure assembly:Integrating catalytic sites (e.g., TiO_2, MOFs) into photothermal frameworks to realize synchronous solar evaporation, organic degradation, and heavy metal adsorption.

With rational cross-scale structural design and targeted interface modification, graphene-based photothermal materials are expected to achieve high efficiency, long stability, and low cost in real-world solar-driven water purification and seawater desalination.

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