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REVIEW ARTICLE – MATERIAL SCIENCE (MISCELLANEOUS)

The Forms and Performance Tuning of Carbon Nanofibers in Composite Phase Change Materials

Rongling Zhang[1*], Xialong Cai[1], Wei Feng[1], Yudi Li[2], Hanjun Wei[3], Hassan Wathiq Ayoob[4], Hanan Osman[5]

[1]School of Mechanical Engineering, Chengdu University, 2025 Chengluo Avenue, Chengdu 610106, China

[2]School of Mechanical Engineering, Southwest Jiaotong University, Chengdu 610031, China

[3]Institute for Advanced Study, Chengdu University, 2025 Chengluo Avenue, Chengdu 610106, China

[4]Chemical Engineering Department, Rovira I Virgili University, Tarragona, Spain

[5]Department of Applied Physics, Faculty of Science and Technology, Al Neelain University, Khartoum, Sudan

[*] Corresponding author E-mail: rinazhang2024@163.com

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Abstract

In the context of the global energy transition, the intermittent challenge of renewable energy urgently requires support from efficient energy storage technologies. Phase change materials (PCMs) have emerged as a key solution due to their high-density latent heat storage/release capabilities. However, solid-liquid phase change materials face bottlenecks such as leakage and low thermal conductivity. Carbon nanofibers (CNFs), with their excellent thermal conductivity, porous structure, and mechanical stability, have become an ideal carrier for optimizing composite phase change materials (CPCMs). CNFs can form a continuous thermal conduction network through uniform dispersion and utilize capillary forces to suppress leakage; when combined with materials like graphene to form hybrid systems, they can create multi-dimensional thermal conduction pathways, synergistically enhancing thermal conductivity and alleviating supercooling degree issues; by self-assembling into two-dimensional films or three-dimensional aerogels and other multi-dimensional structures, they can significantly enhance thermal conductivity and encapsulation stability. By leveraging the multi-form regulation strategy of CNFs, CPCMs achieve high energy storage density, rapid thermal response, and excellent cycle reliability, providing an effective pathway for the efficient storage and utilization of renewable energy.

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Keywords: Phase Change Materials; Carbon Nanofibers; Multi-Form Regulation Strategy; Storage and Utilization of Renewable Energy.

1. Introduction

At this critical stage of the global energy system's transition toward clean and low-carbon energy, the efficient use and sustainable supply of energy have become central issues for the development of all countries[1]. Traditional fossil fuels not only have limited reserves, but their combustion also emits large amounts of carbon dioxide and other greenhouse gases, contributing to global warming, frequent extreme weather events, and posing risks to human health and safety[2–4]. Additionally, overreliance on fossil fuels makes nations vulnerable to shifts in the international geopolitical landscape, while developing domestic renewable energy sources can enhance energy self-sufficiency and ensure energy security.

Solar energy, wind energy, and other renewable energy sources offer significant advantages such as cleanliness, low carbon emissions, and nearly unlimited reserves. However, their power generation is intermittent and volatile[5], they cannot generate electricity at night or in windless conditions, and their output is unstable due to weather and seasonal changes, hindering stable grid integration [6]. This core issue of mismatched temporal and spatial distribution between energy supply and demand requires resolution through efficient energy storage and regulation technologies. Among various energy storage technologies, thermal energy storage technology has garnered significant attention due to its ability to adapt to the thermal energy conversion scenarios of renewable energy sources. Among these, PCMs demonstrate important application value due to their unique energy storage mechanisms[7, 8].

These materials can absorb or release large amounts of latent heat during phase transitions, enabling high-density energy storage and nearly isothermal release. They can provide a buffer for the stable output of intermittent energy sources while meeting temperature control requirements in various scenarios, making them a key material for bridging the gap between renewable energy and continuous energy demand. Phase change materials can be classified based on their phase change mechanisms into gas-liquid, solid-gas, and solid-solid systems [9], with detailed classifications shown in Fig. 1. Solid-gas and liquid-gas phase change materials exhibit extremely high latent heat during phase transitions,

Nomenclature & Symbols

PCM	Phase Change Material	CPCM	Composite Phase Change Material
CNF	Carbon Nanofiber	MP	Methyl Palmitate
TES	Thermal Energy Storage	ETESC	Electrothermal Energy Storage Concrete
CVD	Chemical Vapor Deposition	FG	Flake Graphite
PEG	Polyethylene Glycol	LSPR	Localized Surface Plasmon Resonance
PAA	Polyacrylic Acid	OD	Octadecanol
DDA	Dodecanol	EWG	Expanded Waste Glass
DSC	Differential Scanning Calorimetry	SA	Stearic Acid
SN	Different Mass Ratios of GNP and CNF	GNP	Graphene Nanoplate
PCNF	Porous Carbon Nanofiber	D-AS	Dual-Aligned Scaffold

theoretically endowing them with outstanding energy storage capabilities[10]. However, the significant volume changes accompanying their phase change processes severely limit their practical application in thermal energy storage (TES) systems. Solid-solid phase change materials undergo crystal structure transformations in the solid-state during phase change, resulting in relatively small volume changes, typically within 10%, but their latent heat is lower than that of solid-liquid phase change materials.

Solid-liquid phase change materials are one of the most widely studied and applied types of phase change energy storage materials. They store energy through solid-liquid conversion and have certain advantages in terms of energy storage efficiency. However, these materials face key technical challenges: above the phase change temperature, leakage is likely to occur, which not only causes material loss, reduces system energy storage performance and lifespan, but may also lead to component corrosion, pipeline blockages, and other issues, thereby increasing maintenance costs and technical complexity. Additionally, the inherent limitations of phase change materials restrict their application scenarios. For example, poor thermal conductivity hinders heat transfer rates, while low photothermal conversion efficiency and insufficient stability reduce solar energy utilization efficiency[11-14].

Therefore, to address these inherent issues, researchers have developed various optimization strategies, primarily including special containers[15], filler reinforcement methods[16], and microencapsulation methods[17]. Illustration of the mechanism of the coupling process. Cao et al.[18] treated organically modified sodium phosphate dodecahydrate-based phase change composites using polyethylene glycol (PEG) to suppress undercooling, UV-curable polyurethane acrylate encapsulation, and polydopamine to enhance photothermal performance. Encapsulation prevents material leakage, with the enthalpy value decreasing by only 2.6% after 200 cycles, and significantly improving photothermal conversion efficiency. Li et al[19] studied a Co_3O_4/GO-modified TiO_2 shell microcapsule, where the capsule treatment increased thermal conductivity by 57.7%, improved photothermal conversion efficiency by 88.9%, reduced supercooling degree, and demonstrated excellent cycle stability. CNFs, as a representative class of carbon materials, integrate the advantageous properties of both conventional carbon fibers and carbon nanotubes. They exhibit superior physicochemical characteristics, including excellent corrosion resistance and thermal conductivity comparable to that of metallic materials. These attributes render CNFs highly suitable for the development of high-performance phase change composites[20]. The highly porous structure of CNFs enables the effective adsorption and immobilization of solid-liquid phase change materials through capillary forces, thereby significantly mitigating leakage during phase transitions. Moreover, their high thermal conductivity facilitates the formation of efficient heat transfer pathways, compensating for the inherently low thermal conductivity of phase change materials and thereby enhancing the efficiency of thermal energy storage and release. Additionally, the favorable mechanical strength and flexibility of CNFs contribute to the structural integrity and processability of the composite, making it well-suited for advanced applications such as flexible electronics and wearable devices. The experimental results by Cui et al.[21] indicate that due to the larger diameter of CNF compared to carbon nanotubes (CNT), CNF is more easily dispersed in the PCMs matrix. Consequently, the thermal conductivity of CNF/PCMs composites is higher than that of CNT/PCMs composites at room temperature. CNF can be used as an additive to effectively enhance the temperature response characteristics and thermal conductivity of PCM composites without reducing the latent heat storage capacity of the phase change materials.

Various fabrication techniques, including electrospinning[22], Chemical Vapor Deposition(CVD)[23, 24], and template-assisted synthesis[25], allow for the production of CNFs with diverse structural morphologies[26]. This is manifested not only in the diversity of macroscopic morphologies but also in the existence of distinct structural forms at the microscopic scale[27]. Consequently, CNFs can adopt multiple configurations when incorporated into CPCMs. However, comprehensive investigations into this specific area remain limited, with only fragmented references appearing in general carbon material reviews or other peripheral literature[28, 29]. Therefore, this study aims to systematically summarize and analyze the structural forms of CNFs in CPCMs, as well as their influence on phase change performance.

2. CNF Reinforced Composite Phase Change Material

As a carbon material exhibiting a graphite-like structure, CNF atomic layers serve as ideal additives for enhancing thermal conductivity, capable of directly forming efficient heat transfer pathways within composite materials[30]. In studies focused on improving the overall performance of composites, carbon nanofibers are typically uniformly dispersed within the phase-change matrix. This dispersion state effectively integrates nanoscale effects with interfacial interactions, minimizes undesirable fiber aggregation, and improves interfacial bonding with the matrix, thereby enabling effective functional reinforcement. Elgafy's experimental findings[31] indicate that the incorporation of CNF into paraffin, which serves as the phase change material, leads to a significant enhancement in the thermal conductivity of the composite at room temperature. This improvement is observed across various mass proportions of CNF, with higher mass ratios resulting in greater increases in thermal conductivity.

2.1. Dispersed CNFs

Singh et al[32]. develop CPCMs using methyl palmitate (MP) as the phase change component, expanded waste glass (EWG) as the supporting matrix, and CNFs as thermal conductivity enhancers (Fig. 2a). Fig. 2b demonstrates with CNF contents of 2, 4, and 8 wt%, the thermal conductivity of the composite increased by 29.2%, 62.5%, and 112.5%, respectively. This improvement is attributed to the ultrasonic homogenization process, which ensures uniform dispersion of CNFs in the EWG/MP system, facilitates the formation of a continuous thermal conduction network, and reduces internal and interfacial thermal resistance between EWG and MP by filling interstitial gaps. The composite demonstrates excellent thermal stability and cycling performance, exhibiting a phase change latent heat loss of only 0.5%–0.72% after 1,000 thermal cycles. It remains thermally and chemically stable at temperatures below 227°C. In addition to its enhanced thermal properties, the uniform dispersion of CNFs enables the formation of a conductive network, allowing the composite to perform electrothermal conversion. Ren et al[33] developed a novel electrothermal energy storage concrete (ETESC) by integrating encapsulated phase change aggregate with CNFs. The CNFs are uniformly dispersed throughout the matrix, forming continuous conductive and thermally conductive networks. This structural design effectively reduces the response time of the phase change materials during charging and discharging processes, thereby enhancing the efficiency of thermal energy storage and release. When the CNF content reaches 2.0 vol%, the resistivity of the concrete decreases to 1860 $\Omega\cdot$cm, indicating optimal electrical conductivity, which facilitates efficient electrothermal conversion and supports phase change energy storage. Qin et al[34] employed mechanical stirring and ultrasonic dispersion techniques to achieve uniform dispersion of CNFs in a filamentous configuration within the 1-octadecanol (OD)/nano-TiO_2 system, thereby forming continuous thermal conduction pathways. Upon reaching a CNF mass fraction of 4%, the thermal conductivity of the composite material increases by 34.4% compared to the OD/nano-TiO_2 composite material without CNFs (NPCM3). Furthermore, CNFs and nano-TiO_2 demonstrate synergistic effects that contribute to the suppression of OD leakage. The fibrous structure of CNFs and the particulate structure of nano-TiO_2 collectively enhance the specific surface area of the composite. Through capillary forces and surface tension, this structural configuration improves the retention of liquid OD, resulting in a minimal mass loss of approximately 1.3% after 100 thermal cycles for CPCM3, while maintaining excellent shape stability. Additionally, the melting enthalpy of CPCM3 remains at 209.3 J/g. Gökhan Hekimoglu et al[35] doped CNFs into polyacrylic acid/dodecanol (PAA/DDA) composites, which increased the specific surface area of the PAA network, provided an encapsulation scaffold, and enhanced surface tension and capillary forces. This increased the maximum loading capacity of DDA from 33 wt% to 70 wt% without leakage. As Fig. 2g shows, when 4 wt% and 10 wt% were doped, the thermal conductivity increased by 46.26% and 179.77%, respectively. Similarly, Xu et al. [36]also enhanced thermal conductivity by incorporating CNF into CPCM to form an efficient thermal conduction network, while assisting the supporting material in improving shape stability.

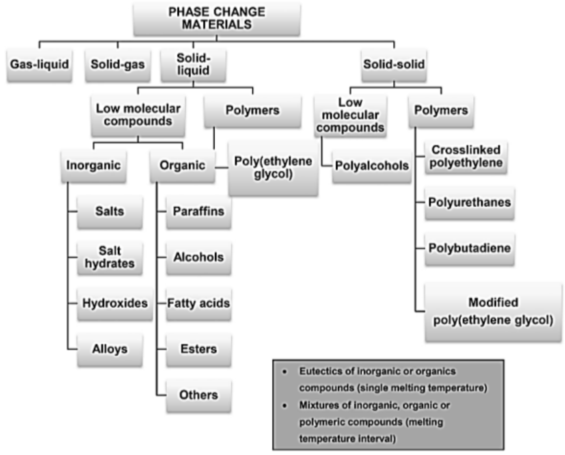

Fig. 1. Classification of energy storage materials, Copyright 2014,Elsevier Ltd. [9]

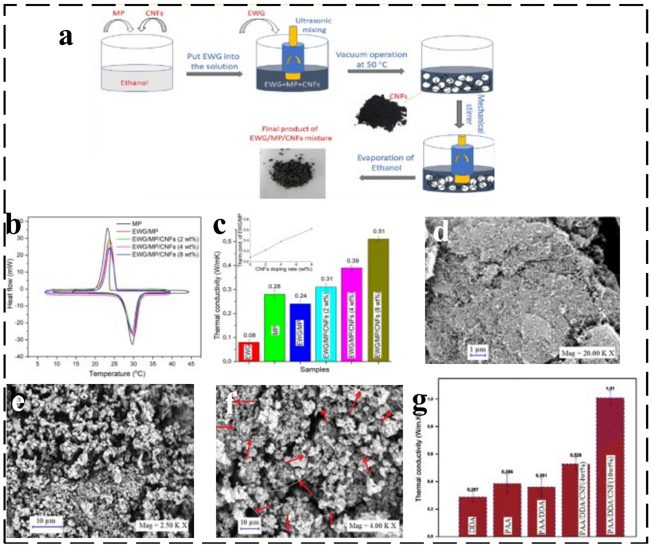

Fig. 2. (a) The experimental process used for the development of the EWG/MP/CNFs composite PCMs, (b) DSC heating/cooling thermograms for pure MP and the prepared LPCPCM with/without CNFs, (c) Thermal conductivity results of EWG, MP, LPCPCM and TE-SSPCMs, Copyright 2023, Elsevier Ltd. All rights reserved [32], (d) SEM photographs of CNF, (e) PAA/DDA(33 wt%), (f) PAA/DDA (70 wt%)/CNF (10 wt%), (g) Thermal conductivity results of DDA, PAA, PAA/DDA(33 wt%), PAA/DDA(48 wt%)/CNF (4 wt%) and PAA/DDA(70 wt%)/CNF (10 wt%), Copyright 2022, John Wiley & Sons Ltd. [35]

2.2. CNFs hybrid fillers

In addition to dispersing CNF individually within the matrix, the use of hybrid filler systems formed by combining CNF with other materials can achieve a more significant enhancement in thermal conductivity. This approach overcomes the limitations associated with a single CNF, which can only marginally improve thermal conductivity, by leveraging synergistic effects between the constituent materials. In the study conducted by Tan et al[37],the incorporation of hybrid-dimensional carbon nanomaterials (HDCN), consisting of CNF and graphene, into a modified lotus root starch matrix significantly enhances the thermal performance of CPCM. Specifically, CNF exhibits inherently high thermal conductivity. When synergistically integrated with graphene, they construct a continuous and stable "thermal conduction pathway," resulting in a maximum thermal conductivity of 1.403 W/(m·K) for the CPCM—approximately 2.5 times higher than that of the pure SAT-STP eutectic material. Moreover, the one-dimensional fibrous structure of CNF is interwoven to form a porous network, which employs capillary forces and surface tension to immobilize the eutectic PCM in its molten state, thereby minimizing leakage and improving the morphological stability of the composite. Additionally, the HDCN facilitates heterogeneous nucleation of the sodium acetate trihydrate-sodium thiosulfate pentahydrate (SAT-STP) eutectic system, thereby lowering the nucleation energy barrier. This results in a reduction in semi-crystallization time from 0.96 minutes to 0.78 minutes, effectively mitigating the undercooling phenomenon. Zhang et al[38] created a "one-dimensional-two-dimensional" hybrid carbon filler system by combining CNF with graphene nanoplates (GNP). In this system, CNF exhibits a slender tubular structure with a diameter of 200–600 nm and a length of 5–50 μm. This one-dimensional morphology enables CNF to span the gaps between the matrix, acting as a “bridge” connecting the two-dimensional GNP layers. In the composite material, GNP and CNF either adhere to the material surface or are embedded within the matrix. Some CNFs also directly bind with GNP to form thermal bridges, thereby filling gaps created during GNP dispersion, reducing thermal resistance, and enhancing phonon heat transfer. Fig. 3b shows that when GNP and CNF are mixed at a mass ratio of 1.7:0.3 (sample SN2), the thermal conductivity of CPCM reaches 0.759 W/(m·K), demonstrating excellent thermal conductivity.

Additionally, its latent heat of fusion is 116.41 J/g, with performance degradation of only approximately 5% after thermal cycling. Li et al[39] grew CNF on the surface of flake graphite (FG) using CVD technology, forming an FG-CNF structure. The fibrous structure of CNF acts as a “bridge,” tightly bonding with the FG surface on one hand, and participating in condensation reactions via surface active groups on the other, achieving chemical bonding with modified bentonite (B_m). This connects FG, CNF, and B_m through chemical bonds to form an integrated carrier. This structure not only enhances the overall stability and cycling reliability of the support material, after 50 thermal cycles, the latent heat change of the composite phase change material fs-PCM3 is only 0.05%, demonstrating excellent thermal stability, but also endows the composite material with a thermal conductivity of 2.803 W/(m·K) shown in Fig. 3f, which is 10.5 times that of pure stearic acid (SA), while effectively suppressing leakage issues.

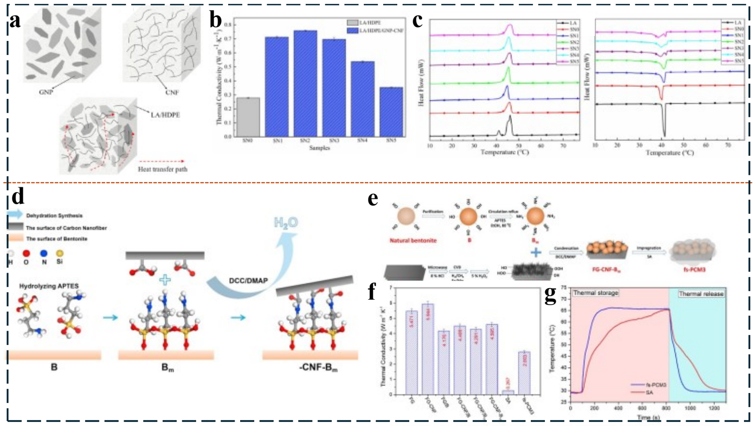

Fig. 3. (a) Schematic representations of thermal conductivity enhancement mechanism, (b) Thermal conductivity histogram of SN0–SN5, (c) DSC curves of LA and SN0–SN5 of melting process and solidification process, Copyright 2023,Society of Plastics Engineers [38], (d) Illustration of the mechanism of coupling process, (e) Scheme of preparation route of composite, (f) Thermal conductivities of samples, (g) Temperature-time curves of SA and fs-PCM3 in thermal storage-release process, Copyright 2019, Elsevier Ltd. [39]

2.3. 2D and 3D carbon nanofiber networks

In phase change energy storage materials, the dispersion state of CNFs plays a critical role in performance regulation. As previously discussed, uniform dispersion of CNFs enables the formation of an effective thermal conduction network, which enhances thermal conductivity and mitigates leakage. However, the influence of CNFs extends beyond mere dispersion; the regulation of both macro- and micro-structures is equally essential for optimizing material performance. Through various preparation strategies, such as self-assembly or template-based methods, CNFs can be precisely engineered into two-dimensional membrane-like or three-dimensional porous network structures. These tailored structural configurations not only affect the interaction between CNFs and PCMs but also directly determine the thermal, mechanical, and functional characteristics of the resulting composite materials. Furthermore, such structures provide PCMs with a stable loading environment and efficient thermal conduction pathways, which constitute the unique advantages offered by CNFs.

Jing et al[40] fabricated porous carbon nanofibers (PCNFs) by encapsulating PEG within electrospun polyacrylonitrile/polystyrene composite nanofibers, followed by pyrolysis, which is shown in Fig. 4a. The resulting CPCM demonstrated excellent performance, including melting and solidification latent heats of 71.9 J/g and 70.9 J/g, respectively. In Fig. 4b, it maintained good thermal stability after 100 thermal cycles, with thermal conductivity improved by 45% compared to pure PEG. Additionally, Fig. 4c and Fig. 4d demonstrate that the material exhibited superior solar-to-thermal and electric-to-thermal energy conversion capabilities, along with notable cycling stability, attributed to the interconnected carbon nanofiber matrix. The PCNFs possess a two-dimensional fibrous membrane structure, in which the fibers are interwoven to form a continuous network. With an average diameter of approximately 740 nm, high porosity, and excellent flexibility, the PCNFs provide a stable structural framework for PEG loading. The capillary forces within the porous structure enable effective immobilization of PEG, allowing the composite material to simultaneously exhibit favorable flexibility, shape stability, and efficient solar-to-thermal as well as electric-to-thermal energy conversion performance. The experimental samples developed by Zeighampour et al. [41]exhibit favorable flexibility, rendering them suitable for wearable applications. The composite framework constructed from CNFs and reduced graphene oxide nanoparticles effectively suppresses the leakage of PEG, with no leakage observed even after exposure to 75°C for 72 hours. Following 3000 cycles of abrasion testing and 5 laundering cycles, the samples demonstrate a weight loss of less than 4%, while the reductions in thermal conductivity and heat transfer efficiency are both confined to less than 10%.

Niu and Yuan's study[42] utilized wood-derived cellulose nanofibers as raw materials to fabricate carbon nanofiber aerogels (CNFAs) with well-defined three-dimensional network structures. These CNFAs were subsequently integrated with SA and 1-tetradecanol (1-TD) to develop shape-stable PCMs. The ultra-high porosity and nano-porous architecture of the aerogels enable the immobilization of molten PCMs through capillary forces and surface tension, effectively preventing leakage while maintaining structural integrity. The interconnected fibrous network provides a continuous pathway for thermal conduction, which, in conjunction with the inherent high thermal conductivity of carbon materials, significantly enhances the heat transfer performance of the PCMs. Additionally, the substantial pore volume allows for a PCM loading capacity exceeding 98%, preserving a latent heat of phase change comparable to that of pure PCMs. Furthermore, as shown in Fig. 4f, the structure leverages the light-absorbing properties of carbon nanofibers to efficiently capture photons and convert them into thermal energy, achieving a solar-thermal energy conversion efficiency of 96.2%.

Similarly, researchers[43] employed a template method to deposit ZIF-67 onto pre-treated bacterial cellulose, followed by the preparation of a CNF@Co/C aerogel with a neural network-like architecture via high-temperature carbonization. Within this aerogel structure, CNF functions as "axons," while Co/C polyhedra act as "neurons." This distinctive design imparts the material with exceptional light absorption properties. By harnessing the synergistic effects of the localized surface plasmon resonance (LSPR) of Co/C polyhedra and the photon transport pathways facilitated by CNF, the solar-to-thermal conversion efficiency achieved a maximum of 95.27%. In terms of carbon nanofiber structural design, in addition to the enhancement provided by the biomimetic network, the ordered alignment of CNF further significantly enhances performance. Zhan et al[44] addressed the challenges of low thermal conductivity and phase leakage in phase change materials by utilizing a magnetic field-assisted freeze-casting technique to construct a dual-aligned scaffold (D-AS), featuring micro-scale CNF alignment and macro-scale oriented channels shown in Fig. 4g. The CNFs are aligned along the direction of heat transfer, thereby fully exploiting their high axial thermal conductivity. In Fig. 4h, when the carbon nanofiber content reaches 16.21 wt%, the thermal conductivity of D-AS/polyethylene glycol attains 5.48 W/(m·K), marking an increase of 1,857.1% compared to pure polyethylene glycol. Moreover, the porous structure of D-AS effectively immobilizes polyethylene glycol through capillary forces and surface tension, with no leakage observed even at 80°C.

In other studies, CNFs, which exhibit high axial thermal conductivity, serve as the fundamental components of the thermal conduction network and collaborate with other carbon-based materials to enhance the thermal conductivity of phase change composites, thereby improving the efficiency of heat storage and release. Song et al[45] synthesized graphene-connected carbon nanofibers, wherein graphene serves as a "bridge" connecting adjacent CNFs, thereby forming a continuous three-dimensional thermal conductivity network referred to as the "fiber-graphene-fiber" structure. This structural design effectively reduces the thermal resistance between CNFs, resulting in a thermal conductivity coefficient of 1.21 W/(m·K) for the composite material, representing a 337% enhancement compared to pure lauric-myristic-stearic acid (LA-MA-SA). Additionally, the heating and cooling efficiencies are improved by 45.7% and 64.9%, respectively. The material's porous structure enables the confinement of molten LA-MA-SA through capillary forces and surface tension, preventing leakage even at 60°C, while achieving a high LA-MA-SA loading capacity of 72.1% with a melting enthalpy of 120.4 kJ/kg. This ensures efficient thermal energy storage and maintains structural stability over 30 thermal cycles. Variations in carbonization temperature were found to influence the pore distribution and specific surface area of the resulting material. In addition to synergistic effects with carbon-based materials, CNFs can also cooperate with metal particles to construct efficient thermal conduction paths. Song et al.[46] shows that form-stable phase change materials with copper nanoparticle-attached carbon nanofibers (Cu/CNFs) as the supporting structure can significantly improve the heat transfer efficiency of composite phase change materials, while shortening the melting and solidification times and enhancing the thermal storage/release rates. Moreover, the high specific surface area and porous structure of CNFs enhance the interfacial bonding with lauric acid-myristic acid-stearic acid ternary eutectic mixture, which can promote the uniform dispersion of phase change materials and reduce the latent heat loss caused by interfacial effects.

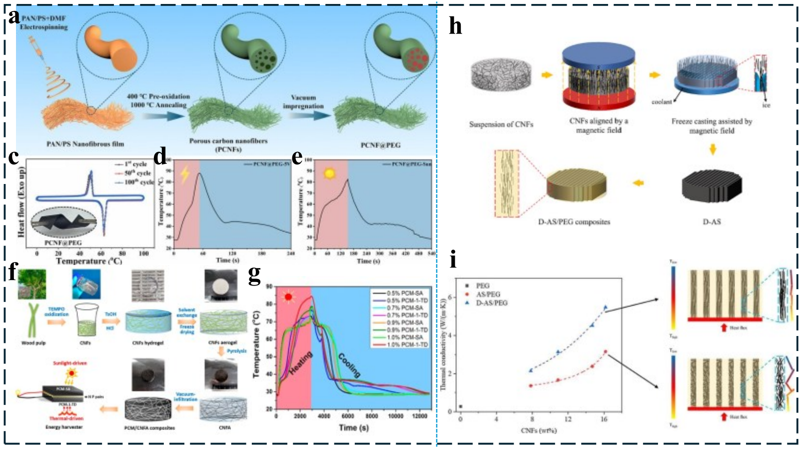

Fig. 4. (a) Schematic of fabrication process of PCNF@PEG, (b) Phase change stability of PCNF@PEG, (c) Solar-/electro-thermal energy conversion and storage performance, (d) Time-temperature evolution curve of the PCNF@PEG under the voltage of 5.0 V, (e) Time-temperature evolution curve of the PCNF@PEG under simulated solar irradiation, Copyright 2024, Elsevier Ltd. [40], (f) Schematic description of the processes of the PCM/CNFA composites and the energy harvesting device, (g) Solar-to-thermal energy conversion curves of the PCM-SA and PCM-1-TD samples under sunlight irradiation, Copyright 2019, American Chemical Society [42], (h) Scheme illustration for the fabrication of D-AS/PEG composites, (i) Schematic diagram of heat transfer of the AS/PEG and the D-AS/PEG composites[43]

3. Conclusion

CNF, with its unique structural characteristics and excellent physical and chemical properties, has become a core functional material for optimizing the performance of PCMs, providing effective solutions to key challenges such as leakage issues, low thermal conductivity, and insufficient cycle stability in solid-liquid phase change materials. CNFs exhibit diverse morphological regulation modes in CPCMs, realizing core functionalities including thermal conductivity enhancement and leakage suppression via tailored structural designs. However, distinct discrepancies exist among different morphologies in terms of performance, cost, and application scenarios.

When CNFs are uniformly dispersed as a dispersed phase, they enhance thermal conductivity by forming a continuous thermal conduction network and restraining PCM leakage through capillary forces and surface tension within the fibrous network. This configuration features relatively low cost, but the improvement in thermal conductivity is limited; moreover, high loading content tends to reduce the latent heat of PCMs, rendering it suitable for scenarios with moderate to low thermal conductivity requirements rather than high-efficiency heat dissipation demand. In hybrid systems composed of CNFs and materials like graphene or flake graphite, CNFs act as "bridges" connecting two-dimensional carbon materials, filling interfacial gaps to reduce thermal resistance and achieve synergistic enhancement of thermal conductivity. Nevertheless, such systems suffer from poor interfacial compatibility between components, complex preparation processes, and high costs, restricting their application to high-end thermal conductivity scenarios with limitations in flexibility and lightweight design. When self-assembled into two-dimensional films, three-dimensional porous structures, or ordered aligned architectures, CNFs optimize heat transfer pathways and provide a mechanically stable framework for PCM encapsulation, endowing the composites with additional functionalities such as flexibility and photothermal/electrothermal conversion capabilities. Despite their superior performance in thermal conductivity and stability, these configurations are characterized by extremely high preparation costs, complex processes, and limited PCM loading capacity, confining their use to high-end applications and hindering large-scale implementation.

The structure and form of CNF have a decisive impact on the performance of composite phase-change materials. Its "structural designability" and "multifunctionality" make it a key link between renewable energy and sustained energy demand, laying an important foundation for improving energy storage system efficiency and promoting the large-scale application of clean energy. Future research could further focus on precise control of CNF structure, in-depth understanding of the mechanisms underlying their interaction with phase change materials at interfaces, and the development of low-cost, scalable production technologies to achieve broader practical applications in fields such as flexible electronics, building energy efficiency, and photovoltaic thermal storage.

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