

Review

Cellulose aerogel-based phase change materials for thermal energy storage: Advances and future perspectives

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ABSTRACT

Cellulose aerogel-based phase change materials (CA-PCMs) combine the latent heat storage capability of phase change materials (PCMs) with the hierarchical porous architecture, low density, and structural stability of cellulose aerogels (CA), providing a promising platform for sustainable thermal energy storage. This review critically examines recent advances in CA-PCMs from a materials-by-design perspective by correlating fabrication strategies, thermophysical properties, and structure-property-performance relationships. A comparative assessment of impregnation, in situ polymerization, encapsulation, and direct mixing establishes the influence of fabrication route on PCM loading, latent heat retention, thermal conductivity, mechanical stability, and cycling durability. Furthermore, graphene-, MXene-, CNT-, polymer-, and other cellulose-based hybrid systems are critically compared, demonstrating that conductive nanofillers can enhance thermal conductivity by 50–200% while introducing important trade-offs between heat transfer, latent heat storage, multifunctionality, and manufacturing scalability. Unlike previous reviews, this work integrates comparative performance analysis, commercialization perspectives, and emerging computational approaches into a unified materials design strategy for rational CA-PCM development. Finally, the review identifies key challenges, including long-term durability, scalable manufacturing, sustainable processing, and multifunctional integration, and proposes a research roadmap to accelerate the industrial translation of next-generation CA-PCMs.

1. Introduction

In recent years, pursuing environmentally friendly and efficient energy storage solutions has garnered the focus of researchers and industry sectors, driven by the escalating energy demands and to mitigate climate change [1,2]. Scientists have introduced many materials with energy storage capability such as phase change materials (PCMs), graphene, metal oxides, etc which can fulfil these needs [3–6]. Out of these, PCMs have emerged as appealing candidate across diverse industries and research domains, representing a paradigm shift in thermal energy management. PCMs demonstrate the appealing property to store thermal energy, they keep the energy stored in one phase and during the phase change they can absorb and release a substantial amount of latent

heat [7–13], enabling efficient thermal regulation in a wide range of applications. Organic PCMs such as paraffin waxes and fatty acids, as well as inorganic PCMs including salt hydrates and metallics, have been extensively studied and deployed in building thermal management [14, 15], solar energy storage [16,17], electronics cooling [18,19], thermal protection for spacecraft and aircraft [20,21], temperature-regulated textiles and wearables [22,23], food preservation and medical storage [24,25], and industrial heat recovery systems [26,27]. Recent advances have expanded PCM applications to include latent heat thermal energy storage in concentrated solar power plants, phase change slurries for district heating networks, and composite PCMs for smart responsive materials [28–33]. These Phase change materials are broadly classified as organic, inorganic, and eutectic PCMs, each differing in thermal

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performance, cost, and compatibility with host matrices. As, this review focuses on Cellulose aerogel-based phase change materials (CA-PCMs) employing organic and organic-eutectic PCMs, which constitute over 95% of reported CA-PCM systems and offer practical long-term thermal energy storage. Inorganic hydrate-based PCMs are mentioned only for context, as they are generally incompatible with Cellulose aerogels (CAs) and not considered viable candidates for CA-PCM integration. Despite excellent heat storage and release capabilities of the PCMs, the conventional PCMs face limitations in practical applications due to their structural deformation and leakage issues during phase transitions, which restrict their broad applications in real time and necessitate the adaptation of advanced encapsulation and stabilization approaches. The structural characteristics of the supporting matrix or encapsulation materials greatly influence the energy storage efficacy and functional reliability of shape-stabilized PCMs. Aerogel-based PCMs like CA-PCMs are considered a state of the art approach compared to other composite PCMs, as they allow for increased functionality, enhanced structural stability, and significantly reduced leakage of PCMs [34–38].

Mineral based composite PCMs are form-stable systems encased in natural or inorganic mineral supports including clays, diatomite, kaolin, porous silicates or other mineral matrices to prevent leakage during melting and solidification [39]. Researches have demonstrated that mineral scaffolds can offer a porous and chemically active matrix for vacuum impregnation or adsorption of fatty acids, paraffins, and other PCMs, thus enhancing form stability, thermal reliability, and practical applicability [40,41]. For instance, the preparation of form-stable composite PCMs with enhanced encapsulation by utilizing clay-mineral systems and the effective solar-thermal storage and leakage suppression by employing diatomite-based composites with strong confinement of the PCM within the mineral network have been described. In CA-PCM systems, mineral fillers or mineral-like hybrid networks may have a complementary role to play by improving the thermal conductivity, mechanical robustness, flame resistance and dimensional stability especially when the cellulose aerogels (CA) are combined with clays, diatomite, silica or other inorganic phases [19].

Aerogels are one of the most effective porous scaffolds for shape-stabilized PCMs. Their interconnected three-dimensional topologies may concurrently provide high PCM loading, remarkable capillary confinement and low density. Aerogels also have the merit of customizable pore architectures and surface chemistry to optimize the thermal energy storage, structural stability and multifunctional performance simultaneously, compared with conventional porous supports. PCM systems with polymer, silica, carbon, metal, and cellulose-based aerogels are making progress in generating multifunctional CA-PCMs [24,38, 42–48]. CA-PCMs have gained substantial attention because of their remarkable thermal characteristics, adjustable porosity, enhanced biocompatibility, sustainability credentials, and environmental friendliness compared to synthetic alternatives.

Cellulose is a naturally abundant and renewable biopolymer which is a biodegradable matrix with high-porosity, low-density aerogels with low environmental effect. Various nanocelluloses, such as nanofibers, nanocrystals, and bacterial nanocellulose, have been used in thermal insulation, optical sensing, anti-counterfeiting, and energy management systems [49–54]. The cellulose matrix can be loaded with PCMs using multiple approaches like impregnation (vacuum or direct), in-situ polymerization, Pickering emulsion stabilization, and blending strategies. CA-PCMs provide almost all critical benefits for practical thermal energy storage, temperature control, building thermal regulation, and thermal protection applications via enhanced mechanical strength, reduced phase transition supercooling, improved thermal cycling stability, and robust form stability [55–61]. Substantial progress has been reported in developing advanced CA-PCM composites inspired by principles of materials science and nanotechnology [62–64].

Based on these foundational advances, researchers have studied CA-PCMs synthesis, characterization, thermal characteristics, and performance, as well as its use in thermal energy storage and management

systems [19,65–67]. Several investigations have demonstrated that thermal properties and mechanical stability can be improved by incorporating CA with an organic PCM (PA16) [19], carbon nanotubes (CNTs) [68], metal-organic frameworks (MOFs) [69], and graphene oxide (GO) [70]. CA-PCMs with CNTs for improved thermal management as well as energy storage. The authors used octacosane as a PCM for their experiments and derived nanocellulose to achieve greater molecular consistency, which Du and coworkers presented [71]. Also, the combination of functional materials such as metal-organic frameworks (MOFs) and graphene oxide (GO) has provided new paths towards modification of the thermal and mechanical properties of CA-PCMs. The report showed an improvement in the thermal performance of CA-PCMs by augmenting them with graphene nanoparticles and PEG, thus broadening the scope of high-performance thermal storage materials, as reported by Wei et al. [72].

In the recent 5 years, aerogel-supported PCMs have progressed fast from simple shape-stabilized thermal storage materials to multifunctional platforms integrating temperature control, energy conversion, sensing, electromagnetic shielding and intelligent thermal management. Therefore, various comprehensive studies have described aerogel-based PCMs from different aspects, including aerogel chemistry, multifunctionality, porous supports, and application-oriented advancements. However, the rapid development of CA-PCMs has given rise to a distinct research area where hierarchical pore engineering, renewable feedstocks, multifunctional nanofillers and computationally assisted materials design jointly govern thermal performance. What is still missing is a key synthesis that explicitly links these features through quantitative structure–property–performance correlations and engineering design concepts. To put the present study into perspective with the current literature, we give a comparison of recent review papers in Table 1.

Recent review articles, detailed in Table 1, have significantly enhanced understanding of aerogel-supported phase change materials, either by focusing on specific categories (e.g., graphene aerogels, 2D nanomaterial aerogels, porous supports, or multifunctional aerogel composites) or by providing comprehensive overviews of aerogel-based PCMs. Nonetheless, these assessments frequently classify the literature by aerogel type or application, providing a limited understanding of the quantitative design concepts relevant to CA-PCMs. There is a notable deficiency in a thorough comprehension of the relationships among manufacturing techniques, hierarchical pore structures, interfacial interactions, thermophysical properties, multifunctional performance, computational modeling, and practical scalability. Nonetheless, the advancement of CA-PCMs from sustainable porous scaffolds to multifunctional thermal energy storage systems has not been thoroughly evaluated through a materials-by-design perspective.

This review addresses this gap by shifting the emphasis from material classification to design-centric analysis. This review transcends mere summarization of published studies by offering systematic comparisons of fabrication methods, establishing quantitative structure-property-performance correlations in cellulose-based hybrid systems, critically assessing thermal storage metrics and multifunctional behavior, and incorporating advanced computational techniques such as molecular dynamics, density functional theory, finite element modeling, and machine learning. It demonstrates how experimental and data-driven methodologies can be synergistically integrated to expedite the rational development of next-generation CA-PCMs. This paper addresses scalability, green manufacturing, and commercialization, offering a comprehensive research framework for the practical implementation of sustainable thermal energy storage systems.

This review provides an in-depth, systematic overview CA-PCMs and their advanced composites, addressing the need for a structured and comprehensive literature on CA-PCMs for advanced thermal energy storage applications. The review is organized in 8 sections as: Section 2 discusses CA-PCMs' practical use and real-world applications in energy storage, thermal control, and climate resilience. Section 3 provides chronological context for recent CA-PCM technology advances by

Table 1
Comparative assessment of recent review articles on aerogel-based phase change materials and the positioning of the present review.

Review	Primary scope	Material system	Structure–property–performance relationship	Fabrication strategy comparison	Computational modelling	ML/AI-assisted design	Distinct limitation addressed by the present review
Yang et al., [73] 2019, <i>Mater. Horizons</i>	3D structural materials for composite PCMs	Carbon foams, graphene aerogels, porous scaffolds, metallic foams, CA	Strong emphasis on thermal conductivity and shape stability	Partial	No	No	Focuses on scaffold engineering rather than cellulose-specific design principles and lacks a comprehensive comparison of fabrication routes and multifunctionality.
Kashyap et al., [74] 2020, <i>J. Mater. Sci.</i>	Graphene aerogel-based PCMs	Graphene aerogels	Focused on graphene-enhanced heat transfer	No	No	No	Restricted to graphene aerogels and does not discuss other cellulose-derived architectures or processing strategies.
Liu et al., [75] 2022, <i>ACS Nano</i>	General aerogel-based multifunctional PCMs	Carbon, silica, polymer, metal, biomass, MXene, BN aerogels	Excellent discussion of multifunctionality	Limited	No	No	Comprehensive overview but broad in scope, with limited cellulose-specific critical analysis, quantitative benchmarking, or computational materials design.
Rostami et al., [44] 2023, <i>Mater. Adv.</i>	2D nanomaterial aerogel PCMs	Graphene, MXene, h-BN and related 2D aerogels	Focused on aerogel architecture and functionality	Moderate	No	No	Restricted to 2D nanomaterials and does not establish cellulose-specific structure–property relationships.
Suchorowiec et al., [24] 2024, <i>Materials</i>	Functional and multifunctional aerogel/PCM composites	General aerogels	Focus on functionality (energy conversion, flame retardancy, conductivity)	Limited	No	No	Functional-material perspective with little quantitative comparison or processing-performance correlation.
Kong et al., [76] 2025, <i>Energy Built Environ.</i>	Shape-stabilized aerogel PCMs	Carbon, silica, polymer, cellulose, MXene and other aerogels	Preparation-oriented discussion	Moderate	No	No	Broad classification and applications, but limited critical benchmarking, computational integration, and cellulose-centered design framework.
Huang et al., [77] 2019, <i>Chem. Eng. J.</i>	Porous supports for shape-stabilized PCMs	Metal foams, expanded graphite, graphene aerogels, CNTs, silica, minerals	Strong discussion of porous support effects	No	No	No	Addresses porous supports in general rather than aerogels or cellulose-derived porous networks and does not consider multifunctionality or computational design.
Present review	CA-PCMs from materials design to application	CA and cellulose-based hybrid systems	Integrates fabrication–microstructure–property–application relationships into unified design principles	Systematic comparison of impregnation, in situ polymerization, encapsulation, and direct mixing, including processing–performance trade-offs	Integrates MD, DFT, FEM, and multiscale modeling with experimental findings	Introduces emerging ML-assisted materials design and optimization	Provides a design-oriented framework that links experiments, modeling, and sustainable engineering for next-generation CA-PCMs.

tracing their historical development, scientific milestones, and its evolution with time. The fundamentals and characterization of CA-PCMs are covered in Section 4, including freeze-drying, vacuum impregnation, in-situ polymerization, 3D printing, and their material properties like density, porosity, morphology, thermal conductivity, latent heat capacity, and mechanical properties. Section 5 reviews advanced CA-PCM composites, including Cellulose/graphene aerogels, Cellulose/MXene aerogels, Cellulose/CNT aerogels, Cellulose/polymer aerogels, and other emerging composite systems, with an emphasis on thermal conductivity enhancement, phase change characteristics, and multifunctional properties, in thermal energy storage applications. Section 6 examines how molecular dynamics simulations, density functional theory, and finite element analysis forecast CA-PCM performance and optimize material design strategies for thermal characteristics. Section 7 addresses technical, economic, and environmental issues in CA-PCM development and deployment, including scalability, cost-effectiveness, long-term stability and cycling performance, environmental impact, and possible solutions and research paths. Section 8 covers future

research directions, emerging opportunities (e.g., shape-memory effects, EMI shielding, acoustic-thermal conversion), potential industrial applications, and strategic recommendations for commercializing and socially impacting CA-PCM research.

2. Utilization of CA-PCMs

The unique combination of hierarchical porous architecture, low density, high latent heat storage, and structural stability enables CA-PCMs to address a broad spectrum of thermal management challenges that cannot be readily achieved using conventional PCMs alone. Depending on the PCM composition and aerogel architecture, CA-PCMs typically exhibit thermal conductivities ranging from 0.018 to $0.075 \text{ W m}^{-1} \text{ K}^{-1}$, porosities of 80–99%, latent heat values approaching 200 J g^{-1} , and encapsulation efficiencies close to 90%, making them attractive for both passive and active thermal management [52,64,78–85]. Consequently, their application has expanded from conventional thermal insulation to multifunctional thermal regulation in buildings,

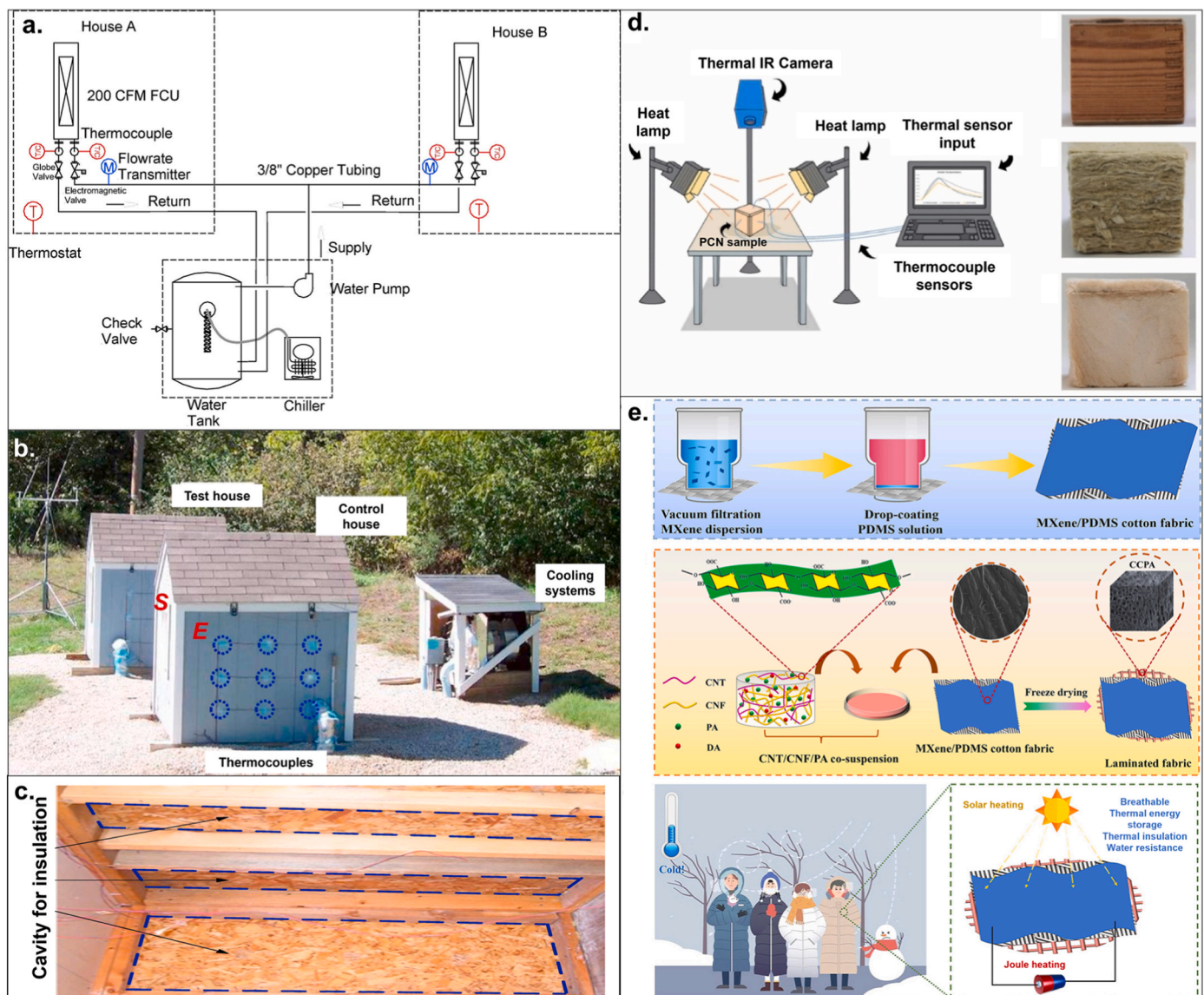


Fig. 1. Utilization of CA-PCMs (a.) Schematic of the field setup of two houses. (b.) real-time images of houses. (c.) images of walls from inside the house. Adapted from Ref. [86] with permission from Elsevier, copyright 2018 (d.) Experimental setup diagram of insulated box behavior, uninsulated wooden box “model house,” mineral wool insulated wooden box, and PCM insulated wooden box. Adapted from Ref. [80] with permission from Elsevier, copyright 2023 (e.) Schematic representation of the manufacturing process for MXene/PDMS cotton fabric, laminated fabric, and laminated fabric utilized as a smart wearable heater for individual temperature regulation. Adapted from Ref. [68] with permission from Elsevier, copyright 2023.

cold-chain logistics, wearable electronics, solar energy conversion, battery thermal management, and aerospace technologies [76,78–80, 86,87]. The following sections discuss these representative application areas and highlight the relationship between material design, thermo-physical performance, and application-specific requirements.

CA-PCMs play an essential role in building construction materials to store and release heat as per the requirement, improving energy efficiency by lowering energy requirements for heating and cooling buildings [2,88]. The cellulosic aerogel and PCMs function synergistically, with the aerogel structure providing good thermal insulation and the PCMs absorbing or releasing heat, stabilizing indoor temperatures, and decreasing reliance on external energy sources. Integrating PCMs into a cellulose matrix does not alter the composite's melting and solidification temperatures [86]. Experimental evaluation on test houses (Fig. 1a–c) showed a peak heat flux reduction of ~25.4% for individual walls; when all four walls were included in the calculation, the reduction was 20.1% [86]. The critical role of cellulose-based cryogens with the help of an experimental setup was established by Le et al., as shown in Fig. 1d [80]. Moreover, PCMs mixed into cellulose matrix in the form of cryogels exhibited robust mechanical resilience and a high energy storage capacity of 204 J/g. At a thickness of 1.5 cm, the material also provided strong insulating performance against a temperature gradient of 33.5 °C [80]. Wall panels, roofing materials, and window inserts can contain CA-PCMs. They passively manage building temperature, saving energy.

CA-PCMs are also being utilized in cold-chain logistics by helping maintain the required temperature range of 2–8 °C, ensuring the freshness of perishable items like food and pharmaceuticals during transportation. The CA-PCMs demonstrated enhanced thermal energy storage, exhibiting a phase change at 6 °C with an enthalpy of 112 J/g and maintaining its heat storage capacity over more than 300 cycles, making it highly suitable for logistic applications using n-tetradecane as PCM (Yan et al.) [89]. These PCMs can be employed in temperature-controlled shipping containers and packing materials protect sensitive goods in transit, making perishable product shipment safer and more reliable.

Personal temperature management wearable electronics can also be benefited via implication of CA-PCMs. Their lightweight, breathable construction and phase shift capabilities help regulate body temperature via cooling or heating. A eutectic combination of lauric acid and myristic acid can be loaded onto graphene oxide and CA as PCM [70]. Mixing lauric acid and myristic acid lowers their melting point compared to individual acid, resulting in a eutectic mixture, tuning the phase change temperature for wearable thermal management. These composites have shown an superior thermal conversion efficiency of 96.5% and electro-thermal conversion of 82.3% with a latent heat of 124.6 J/g. Furthermore, these PCMs exhibited sufficient mechanical strength and flexibility to function as a free-standing, wearable substrate, enabling effective personal thermal management [70]. $\text{Ti}_3\text{C}_2\text{T}_x$ MXene and cellulose-based aerogels for loading paraffin as PCM material. The resulting aerogels have been fabricated into laminated, breathable fabrics for wearable thermal management (Zhao et al.) (Fig. 1e) [68]. These materials can be incorporated into intelligent textiles, coats, or footwear to improve comfort during extreme weather conditions. Potential end-users include military personnel, athletes, and outdoor workers who require reliable thermal regulation in harsh environments.

CA-PCMs are also ideal for solar energy applications, due to their ability to store and release heat as needed. These materials can be integrated into solar panels or thermal batteries, storing heat throughout the day and releasing it at night or during periods of low solar activity, considerably increasing the efficiency of solar thermal systems, allowing them to provide energy continuously. The MXene/CNC-based aerogels loaded with tetradecylamine exhibited a latent heat of 221.5 J/g, which is 106.5% higher than that of pure tetradecylamine, defying theoretical predictions. This enhancement is attributed to the 3D MXene/CNC aerogel structure, which promotes more efficient heat exchange with the environment, while MXene organizes PCM molecules into a denser

arrangement through van der Waals interactions. Notably, these composites demonstrated a phase change temperature of 60 °C and a high solar thermal energy conversion efficiency of ~94.1% (Jiang et al.) [90]. In another report, the use of bacterial cellulose with CNT aerogels for loading PEG as PCM, with a high latent heat of 145.1 J/g, a solar-to-electrical energy conversion of 423 mV, and a high power density of 30.26 W/m² (Zhang et al.) [91]. The thermal insulation properties and temperature resilience of these cellulose PCMs allow them to effectively manage heat from engines and battery systems, enhancing energy efficiency and performance. CA are also suitable for airplanes, spaceships, and electric vehicles due to their low density, high porosity, structural stability and great thermal management characteristics. CA-PCMs combine high energy-storage density, effective solar-thermal-electric conversion, and robust thermal control into a single, flexible film, making them ideal for energy and electronics applications. The silylated bacterial cellulose/CNT/PEG network delivers high latent heat with >94% enthalpy retention over 60 cycles, low leakage, good flexibility after 800 deformation cycles, and regulated thermal conductivity of 0.45 W m⁻¹ K⁻¹, alongside rapid, tunable thermal storage (394 s g⁻¹) and photothermal heating above 80 °C. Under triple solar irradiation, it achieves 423 mV and 30.26 W/m² with remarkable cycling stability, while also enabling thermal buffering and waste-heat utilization for solar cells and LED chips. This illustrates how cellulose-aerogel PCMs can convert and regulate intermittent or localized heat in device-relevant scenarios.

As shown in the above applications, CA-PCMs have been well evolved from traditional latent heat storage materials, and are increasingly explored for multifunctional thermal management in buildings, wearable systems, cold-chain logistics, solar energy conversion, electronics and aerospace technologies. However, the large scale application is still limited by the problems of scalability, durability and economical fabrication. To understand the origin of such issues, it is necessary to review the historical development of CA-PCMs starting from standard PCM systems to modern multifunctional designs based on cellulose. Therefore, the next section is dedicated to the historical evolution of CA-PCMs and the main scientific milestones that have shaped the area.

2.1. Positioning cellulose aerogels within the broader PCM stabilization landscape

In order to solve the leakage issue of solid-liquid PCMs, various stabilization strategies have been developed with different physical confinement mechanisms, while at the same time affecting the thermal conductivity, latent heat retention, mechanical integrity, processability and long-term durability. Conventional stabilization techniques include microencapsulation, porous minerals, expanded graphite, metal foams, silica aerogels, and polymeric porous matrices. These methods have been successful to different extents for different applications. Therefore, the selection of suitable support material should be undertaken by a holistic consideration of the relative trade-off among the heat storage performance, structural stability, multifunctionality, sustainability and manufacturing feasibility. Thus, placing CA in the context of the larger set of PCM stabilizing techniques helps to comprehend their distinct benefits and current limits (Table 2).

In summary, as shown in Table 2, CA do not outperform every PCM stabilization strategy in individual performance metrics. Expanded graphite and metal foams provide superior intrinsic thermal conductivity, polymeric matrices generally offer simpler large-scale manufacturing, and microencapsulation exhibits outstanding leakage control. However, CA occupy a distinctive position within the PCM stabilization landscape by integrating renewable feedstocks, lightweight hierarchical porous architectures, high PCM loading, improved mechanical flexibility, and outstanding compatibility with multifunctional nanofillers. Unlike conventional porous supports, which are typically optimized for a single function, CA provide a versatile platform capable of simultaneously integrating thermal energy storage, structural

Table 2

Comparative positioning of CA-PCMs among the major PCM stabilization strategies for latent thermal energy storage.

PCM stabilization strategy	Stabilization mechanism	Thermal conductivity	Mechanical flexibility	Sustainability	Scalability	Primary engineering advantage	Primary engineering limitation
Microencapsulation	Polymer or inorganic shell confines molten PCM	Low	Moderate	Moderate	High	Excellent leakage control, easy incorporation into coatings, textiles and slurries	Low heat-transfer rate, inactive shell reduces effective energy-storage density
Expanded graphite (EG)	Capillary adsorption within interconnected graphite pores	Excellent	Poor	Moderate	High	Outstanding heat transfer and rapid thermal charging/discharging	High density, fossil-derived precursor and limited mechanical flexibility
Metal foams	Three-dimensional metallic porous framework	Excellent	Poor	Poor	Moderate	Superior thermal conductivity for high-power thermal management	High cost, heavy weight, corrosion and difficult machining
Silica aerogels	Nanoporous confinement through capillary forces	Low	Poor	Moderate	Moderate	enhanced thermal insulation and high PCM loading	Brittle structure and energy-intensive processing
Porous minerals (diatomite, vermiculite, perlite, etc.)	Physical adsorption within mineral pores	Moderate	Poor	Good	Excellent	Low cost, abundant resources and mature manufacturing	Limited multifunctionality, relatively low PCM loading and poor mechanical adaptability
Polymeric porous matrices	Polymer network confinement	Low	Excellent	Moderate	Excellent	High flexibility and simple large-scale processing	Low intrinsic thermal conductivity and limited thermal response
CA	Hierarchical porous capillary confinement reinforced by hydrogen bonding	Moderate (can be considerably enhanced through conductive nanofillers)	Excellent	Excellent	Moderate–High	Unique balance of high PCM loading, lightweight structure, renewable feedstocks, multifunctionality and mechanical flexibility	Low intrinsic thermal conductivity, freeze-drying cost and challenges associated with industrial-scale manufacturing

integrity, multifunctionality, and environmental sustainability. Consequently, the principal advantage of CA-PCMs lies not in maximizing a single thermophysical property, but in achieving a balanced combination of performance characteristics required for next-generation multifunctional thermal energy storage systems.

3. History of CA-PCMs: from PCM technology to cellulose-based systems

The development of CA-PCMs is a good example of the evolution of phase change materials from simple latent heat storage media to multifunctional thermal management platforms. CA-PCMs are not an isolated materials system but rather the result of sequential developments in PCM chemistry, porous scaffold engineering, nanomaterials and sustainable materials processing. Understanding such historical evolution is essential as many of the contemporary design strategies such as hierarchical pore engineering, multifunctional filler integration and computationally assisted optimization were derived from earlier technological developments rather than recent discoveries only. The evolution of CA-PCMs is roughly classified into four major phases, each marking a different scientific milestone that has together defined the present status of the subject.

Phase I: Early development of phase change materials (1960s–2000) discusses the first discovery of PCMs in the 1960s when Ovshinsky demonstrated reversible switching effects in chalcogenide alloys, marking the beginning of PCM research. However, early PCM devices faced critical limitations: sluggish transition rates, high programming currents, and inability to sustain stable phase changes, restricting commercial viability. PCM technology stagnated for approximately two decades with limited improvements. The field revived at the beginning of the 21st century when Ovshinsky's team introduced the Ge-Sb-Te (GST) alloys, which exhibited rapid switching between high-resistance and low-resistance states within nanoseconds [92]. This breakthrough in solid-state PCM performance reignited research interest and drove measurable technological advancements in optoelectronic applications.

Phase II: PCM Expansion Beyond Semiconductors (2000–2010) led to the success of GST alloys expanded PCMs research beyond semiconductor to broader thermal energy storage applications. While PCMs excellently absorbed and released latent heat during phase transitions, researchers identified a fundamental problem during this phase of PCMs transition that it suffered from severe structural deformation and liquid leakage during thermal cycling in practical applications limiting their deployment in real time applications and expansion beyond lab scale. To overcome these limitations, researchers pursued two parallel strategies: (1) develop novel PCMs with improved phase transition properties, and (2) create structural supports and matrices to prevent leakage while maintaining thermal performance.

Phase III: Aerogel-Based PCM Composites (2010–2015) were introduced to overcome the issues of leakage. Recognition of PCM limitations as discussed in Phase 2, led researchers to investigate porous matrices as containment matrix. Aerogels emerged as ideal candidates because their unique properties to overcome the limitations of PCMs, like preventing leakage while maintaining high energy storage density and thermal insulation [75]. This phase was the transition time of the PCMs with the porous materials as they are one of the best available candidates to overcome leakage issues. During this time, research evolved to encompass silica, carbon, polymer, metal, and cellulose-based aerogels, each with unique PCM integration benefits. CA were particularly promising due to their renewable nature, biodegradability, and environmental sustainability, which other synthetic alternatives lack.

Phase IV: CA-PCM Emergence and Rapid Growth (2016–Present) this phase can also be termed as a foundational Milestone in the field of the CA-PCMs for thermal management. Yang et al. incorporated graphene nanoplatelets into 3D CA and impregnated them with polyethylene glycol to synthesize CA-PCMs for the first time. This pioneering work showed that CA can stabilize PCMs and improve thermal performance with filler. PEG's latent heat capacity (>150 J/g) was preserved while the composite provided good shape stability and thermal insulation, outperforming bare PCMs. [93]. Following this breakthrough, CA-PCMs research expanded rapidly. Two major research directions emerged:

First was that, with time researchers progressively focused diverse forms of cellulose providing different mechanical properties, porosity characteristics, and making choice for PCM affinity. The choice of cellulose type became critical parameter for controlling thermal performance and mechanical stability. Second direction was towards addition of fillers to the CA-PCM matrix. In this direction, MXenes, graphene nanoplatelets (GNPs), carbon nanotubes (CNTs), boron nitride (BN), carbon fiber, and conductive polymers (polyaniline, polypyrrole) were incorporated in CA matrix to increase thermal conductivity for faster thermal response. These improvements provided efficient thermal routes across the aerogel matrix, increasing thermal conductivity by 15 times.

Overall, Phase 4 witnessed accelerated publication growth (Fig. 2a and b): research publications on CA-PCMs surged. Milestone Timeline of CA-PCMs and their multifunctional evolution is demonstrated in Fig. 3 [17,55,93–100] First graphene/cellulose/PCM hybrid aerogel synthesis was reported in 2016 (Yang et al.) [93] In between 2018 and 2019 research focused on Widespread adoption of MXenes, CNTs, and graphene in CA-PCM systems [55,86,101–103] The era of 2020–2021 brought the development of multifunctional CA-PCMs with simultaneous thermal, optical, and mechanical properties [17,71,81,83,85,104] Integration of advanced composites combining multiple nanofillers (MXene + CNT, GNP + BN) was targeted between 2022 and 2023 [79, 98,105–109] After 2024 to till date the emergence of smart/sensing CA-PCMs with embedded functionality (self-healing, strain sensing, EMI shielding) is being focused [14,89,100,110–115].

As shown in the historical evolution above, the development of CA-PCMs has gradually evolved from solving the basic problem of PCM leakage to designing multifunctional, high-performance thermal management platforms by rationally controlling the material composition, pore architecture and interfacial design. This development naturally triggers a full discussion on the physicochemical properties and fabrication approaches that regulate the structure–property–performance relationships of modern CA-PCM systems.

4. Characteristics and preparation of CA-PCM systems

CA have become one of the most attractive scaffold materials for organic and eutectic organic phase change materials due to their renewable origin, hierarchical porous architecture, low density, and high chemical compatibility, which together contribute to high PCM loading and long-term structural stability. However, the ultimate performance of CA-PCMs is dictated by the tight connection among cellulose chemistry, PCM selection, fabrication technique, and

microstructural development throughout processing. Ultimately, these characteristics govern pore architecture, interfacial interactions, thermal conductivity, latent heat retention, mechanical integrity, and long-term cycling stability. Thus, it is important to understand the structure–property–performance correlations created during material preparation for the rational design of high-performance CA-PCMs. In the next sections, these regulating parameters are discussed from the point of view of material characteristics, PCM compatibility, fabrication methodologies and the ensuing thermophysical properties.

4.1. Fundamental characteristics of cellulose aerogels for PCM integration

4.1.1. Chemical and structural foundations

At the nanoscale, cellulose exists in diverse forms; cellulose nanofibers (CNFs), cellulose nanocrystals (CNCs), and bacterial nanocellulose (BNC); each providing different mechanical properties, porosity characteristics, and PCM affinity [78]. With its variable characteristics, the choice of cellulose type becomes critical for controlling thermal performance and mechanical stability in final CA-PCMs. The surface of cellulose-based materials is augmented with hydroxyl groups (3–5 mmol/g) plays a vital role in governing its properties, enabling chemical modification possibilities such as esterification with degree of substitution (DS) values of 0.45–0.68 through interactions with fatty acids or inorganic acids [116–118]. Additionally, the pore architecture determines whether fabrication strategies like impregnation will fully penetrate the aerogel or leave unfilled voids. The thermal properties establish the baseline performance that enhancers must improve upon. Thus, careful characterization and control of CA properties at this stage directly determines the success of downstream fabrication and final CA-PCM performance [119]. Importantly, the interconnected nature of aerogel pores, rather than isolated cavities enables effective capillary forces which holds the PCMs within the matrix and avoid the leakage during phase transitions while maintaining rapid heat transfer between encapsulated PCM and the exterior environment.

4.1.2. Thermal and mechanical properties

The very low thermal conductivity of CA (0.020–0.040 W/m·K) results from nanoscale porous structures. These CA hinder phonon and gas-phase heat transfer due to the Knudsen effect [120]. However, this low baseline thermal conductivity necessitates strategic incorporation of thermal conductivity enhancers (discussed Section 4.4) when rapid heat exchange is required to release or store the latent heat. High PCM encapsulation rates (~90%) and low thermal expansion coefficients

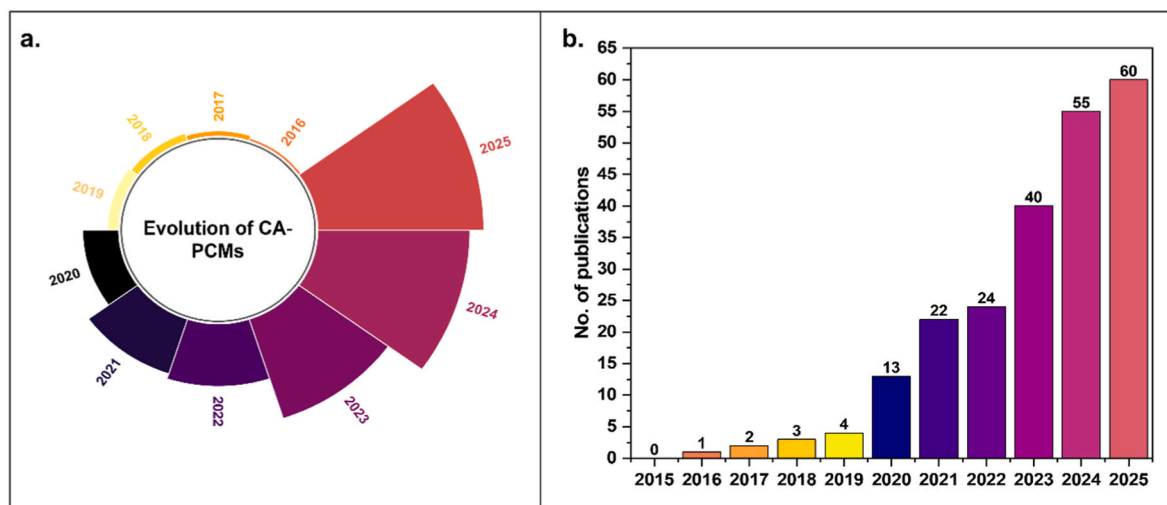


Fig. 2. Statistical evaluation of research publications across 2015 and 2025 using the terms “cellulose aerogels,” “phase change material,” and “thermal energy storage” on Scopus in July 2024. (a.) Yearly variation in the total number of publications between 2014 and 2024, and (b.) Year-by-year contribution of studies on CA-PCMs.

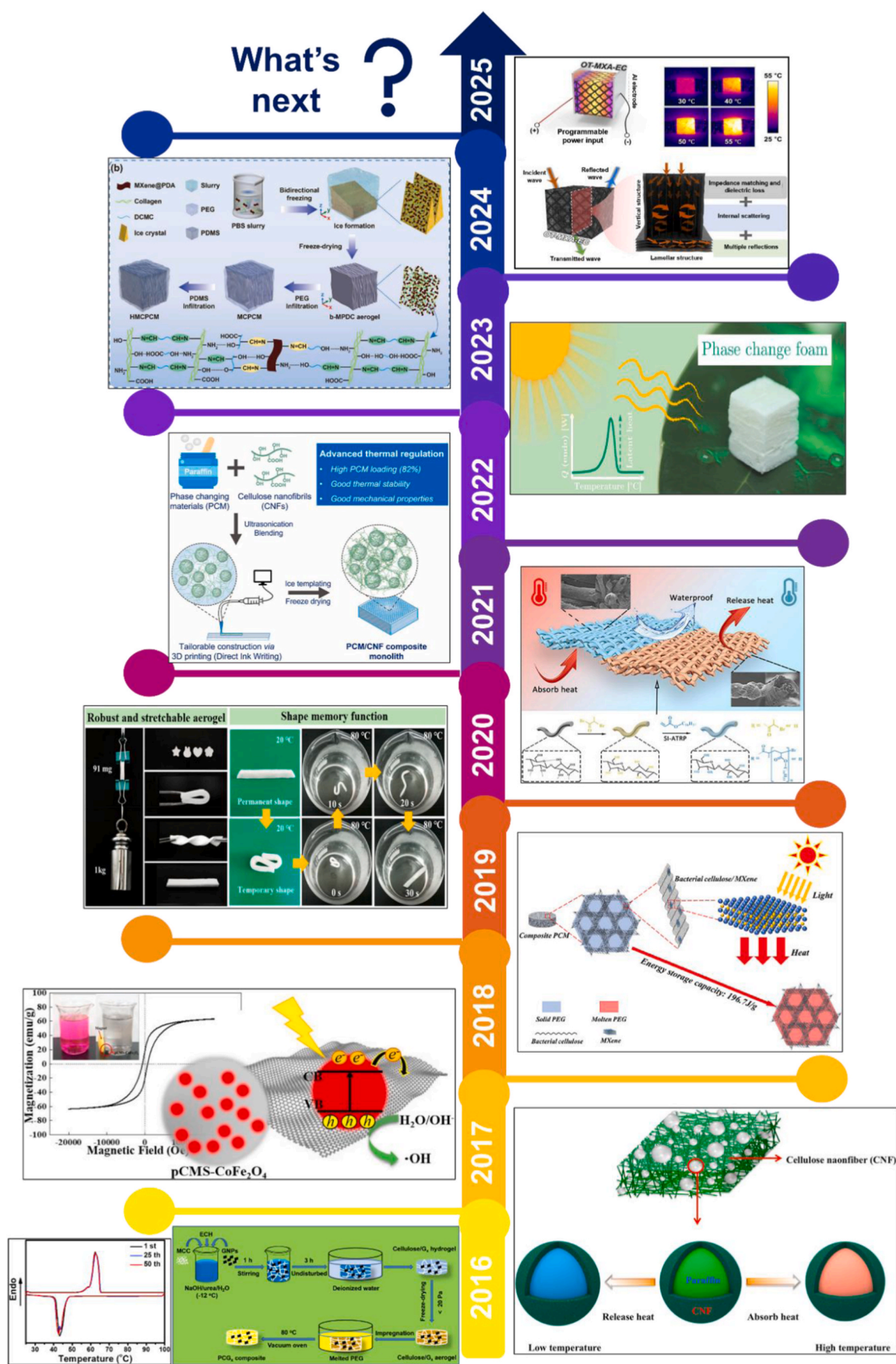


Fig. 3. Timeline of major breakthroughs of CA-PCMs in recent years.

(5–10 ppm/K) of CA-PCMs enable them to store thermal energy with high latent heat storage capacities around 150 J/g. Moreover, CA maintain the structural stability through repeated thermal cycles achieved by low thermal expansion coefficient [121]. These metrics directly translate to practical performance of a 90% encapsulation rate means that nearly all incorporated PCM is contributing to storage the energy enabling minimal loss, while low thermal expansion coefficients prevent stress cracking or warping during temperature fluctuations.

4.2. PCM selection criteria and compatibility with cellulose aerogels

PCMs must be selected for chemical compatibility, thermal characteristics, and long-term stability when integrated with CA. Most successful CA-PCM systems use organic and eutectic organic PCMs, which are more compatible with CA matrices than inorganic hydrate-based alternatives.

4.2.1. Chemical compatibility requirements

Organic PCMs like paraffins, fatty acids, polyethylene glycol, and esters are hydrophobic or weakly polar, allowing them to integrate with CA hydroxyl group rich surface through van der Waals interactions. These PCMs do not release free water during phase transitions, eliminating the possibility of hygroscopic degradation of CA matrix. While working with inorganic salt hydrate PCMs can be challenging due to hydration-dehydration cycles during thermal cycling, releasing H₂O molecules that gradually degrade CA matrix through disruption of intermolecular hydrogen bonding networks, swelling and shrinkage of cellulose due to repeated moisture absorption/release. The chemical attack by concentrated salt solutions on CA's glycosidic bonds and hygroscopic uptake from ambient humidity, leading to uncontrolled water content, and phase separation and leakage of salt-saturated solutions through aerogel pores can also result to the structural deformation of CA. This phenomenon resulting to the structural deformation of CA matrix render the inorganic hydrate-based PCMs incompatible for extended thermal cycling. This necessitates complex encapsulation strategies that notably increase manufacturing complexity and material costs to incorporate inorganic PCMs in the CA matrix. Therefore, CA-PCM research has strategically focused on organic and eutectic organic systems, which offer inherent compatibility without requiring protective barriers.

4.2.2. Thermal property optimization

While the choice of PCMs is somehow limited to Organic and eutectic PCMs with latent heat around 120–200 J/g, they provide adequate energy density for most practical applications n day to day life. Therefore, eutectic organic mixtures enable tunability by blending two organic PCMs, melting point of the forming PCMs can be precisely tuned to match application requirements for instance around 34 °C for wearable thermal management, 60 °C for building thermal regulation, and 6 °C for cold-chain logistics. This flexibility with the organic and the eutectic PCMs are particularly valuable for advanced thermal management applications requiring temperature control within specific heat transfer windows.

4.2.3. Long-term cycling stability

Organic and eutectic-organic PCMs also demonstrate efficient thermal cycling stability, maintaining latent heat capacity and physical integrity over 1000+ thermal cycles. This performance is critical for practical applications in buildings which requires decades of operation, aerospace systems where it can undergo thousands of flight cycles, and renewable energy storage where it requires to show seasonal scale cycling. While the inorganic hydrate PCMs exhibit substantial degradation within 50–100 cycles due to progressive water loss and salt segregation, limiting their practical utility despite higher raw latent heat capacity. These are also in one of the reasons that the most of the CA-PCMs result focuses on the organic or eutectic PCMs and inorganic hydrate

based PCMs in CA matrix are explored less.

4.2.4. Selection framework for CA-PCM design

When developing CA-PCMs, material scientists should prioritize: Organic PCMs for general applications requiring reliable long-term performance. Eutectic organic PCMs when temperature tunability and suppressed supercooling are required. Inorganic PCMs only in specialized applications where: (a) hermetic encapsulation can be achieved at reasonable cost, (b) cycling is limited, or (c) non-hydrate inorganic systems (metallic alloys) are employed with accepting trade-offs in density and mechanical properties. All CA-PCM systems discussed in this review (Sections 5–6) employ organic or eutectic-organic PCMs unless explicitly noted.

Having established the fundamental properties of CA and clarified which PCM types are compatible with cellulose matrices, we now examine how to integrate these components into unified CA-PCM systems. The kinetics of PCM incorporation, loading efficiency, dispersion, thermal conductivity, and mechanical stability are contingent upon the fabrication method employed. Impregnation, in-situ polymerization, encapsulation, and direct mixing are predominant in CA-PCM research. Each approach possesses distinct advantages and employs various PCM-matrix interaction processes, as detailed below.

4.3. Fabrication strategies of CA-PCMs

The fabrication technique is the most important parameter affecting the behavior, such as density, pore structures, and mechanical and thermal properties of CA-PCMs. Synthesis of cellulose-based aerogels involves a set of specific processes that are performed in specific conditions [122–125]. A common process for synthesizing CA consists of several interconnected steps, such as selecting appropriate dissolving solvents, gel preparation and its pore structure, solvent exchange, and the control of pore formation [120]. The aerogel production normally follows a sol-gel method, where molecular precursors are converted into strongly cross-linked gels—inorganic or organic. Subsequently, the liquid components of the wet gels are replaced by gas (typically air) using various drying processes, such as freeze-drying, supercritical drying, or ambient pressure drying, resulting in a 3D solid network [126].

The most crucial component of CA-PCMs is the integration of PCM into the cellulose matrix.

There are several types of PCMs that can be used to prepare CA-PCMs, and there are many ways of PCM integration, such as (a) Impregnation, (b) In-situ polymerization, (c) Encapsulation, and (d) Direct mixing, as shown in Fig. 4a and b. The following subsections explain each fabrication technique in detail.

4.3.1. Impregnation

In the impregnation method, a phase change substance is added and subsequently absorbed into an aerogel's porous structure, commonly using a vacuum oven (in a method known as the 'vacuum impregnation method'). Impregnation remains the predominant manufacturing method for CA-PCMs, since it preserves the pre-established porosity structure of the CA while facilitating the loading of substantial PCM content through capillary-driven infiltration. The efficacy of this procedure primarily relies on pore connectivity, pore size distribution, surface wettability, viscosity of the phase change material, and infiltration conditions. Impregnation is frequently characterized by pronounced latent heat storage capacity and processability; however, partial pore filling and heterogeneous PCM dispersion may occur when using complex pore geometries or high-viscosity PCMs.

For example (Fig. 5a), PEG is liquefied in a vacuum oven at 90 °C. After the PEG melts, the bubbles in the molten PEG are eliminated, and the PEG is impregnated into the bacterial cellulose-based aerogel scaffold using vacuum [17]. Similarly (Fig. 5b), Erythritol is heated to 130 °C to achieve total melting. Then, a P-MXene/CNF composite

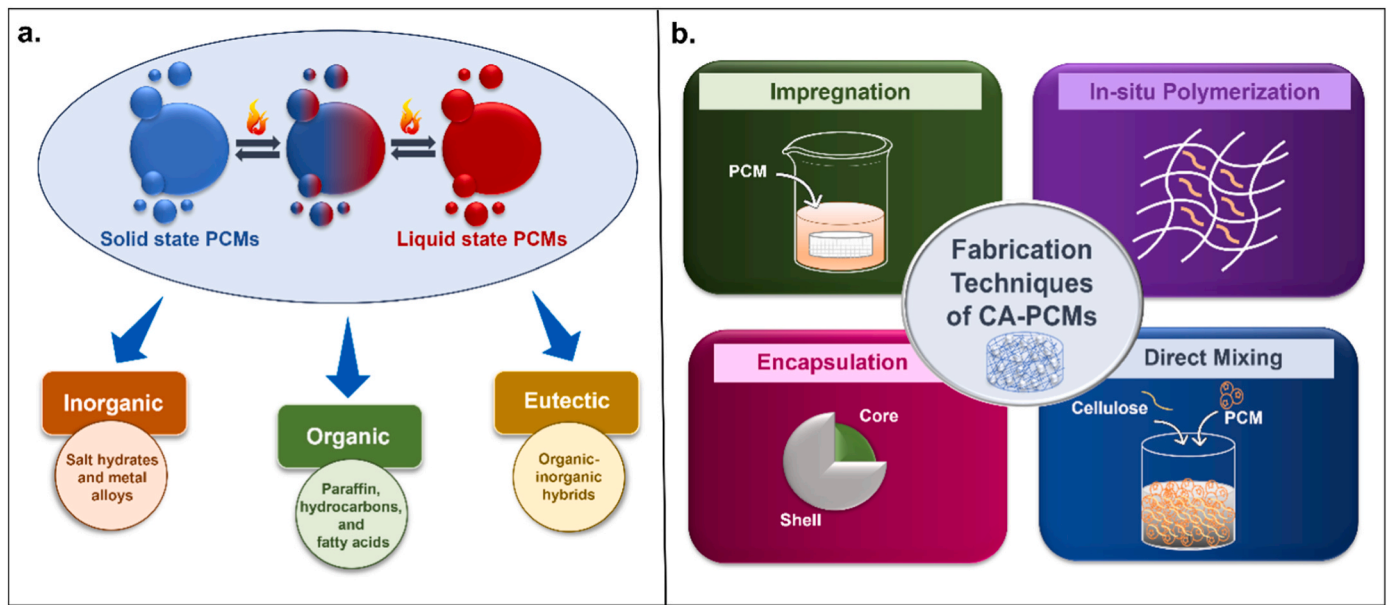


Fig. 4. (a.) Classification of PCMs. (b.) Various fabrication techniques of CA-PCMs.

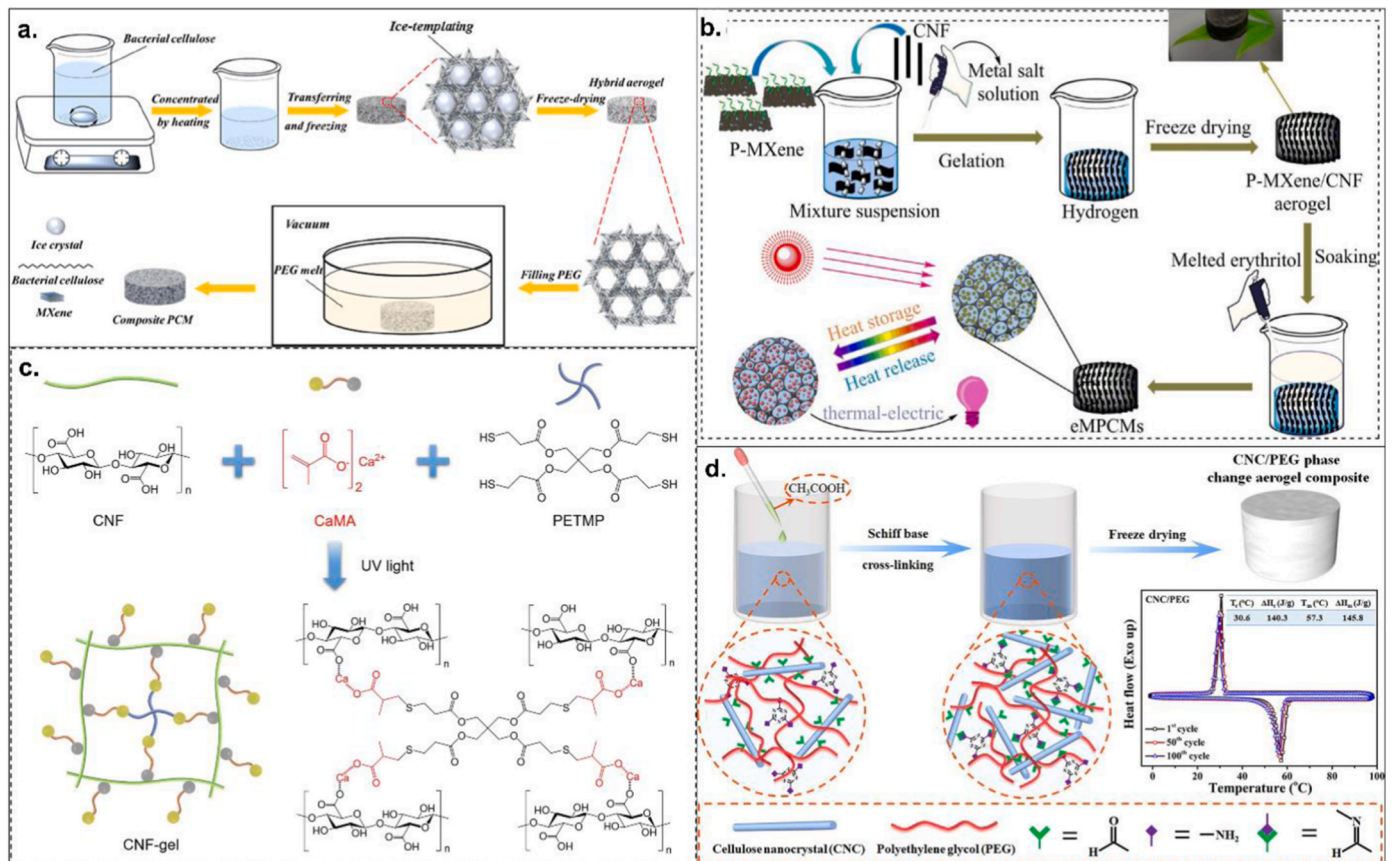


Fig. 5. (a.), (b.) Fabrication of CA-PCMs by impregnation [17,127]. Adapted from Ref. [17] with permission from Elsevier, copyright 2019, and adapted from Ref. [127] with permission from the American Chemical Society, copyright 2022 (c.), (d.) Fabrication of CA-PCMs by in-situ polymerization [108,128]. Adapted from Ref. [128] with permission from Elsevier, copyright 2023, and adapted from Ref. [108] with permission from Elsevier, copyright 2022.

aerogel is prepared by submerging in molten erythritol at 130 °C via vacuum impregnation by Du et al. [127]. The resultant porous and linked 3D aerogels proficiently retain erythritol, inhibiting liquid leakage during thermal energy storage. The erythritol loading rates for the eMPCM composites vary from 1906% to 1934%, respectively. The

composite PCMs show remarkable thermal storage density and PCM loading capacity, with melting enthalpy between 325.4 and 330.6 J/g and crystallization enthalpy from 304.6 to 309.8 J/g, indicating their considerable potential for effective thermal energy storage [127]. This simple process may be less effective as the PCM does not fully cover the

pores, which may result from complex pore structure, viscosity mismatch, or surface chemistry.

While impregnation achieves high PCM loading through physical absorption into pre-formed aerogels, this approach cannot guarantee complete pore filling or prevent concentration gradients that may compromise uniform thermal properties. In contrast, in-situ polymerization approaches build the PCM into the matrix during aerogel formation itself, potentially enabling more homogeneous PCM distribution. The following subsection explores how this alternative strategy trades off complexity for improved uniformity.

4.3.2. In situ polymerization

In the *in-situ* polymerization method, the PCM is polymerized or encapsulated within the CA during the gelation or drying stage. In this method the monomers are polymerized directly within a matrix often in the presence of other components like fibres or nanoparticles to form a composite. In contrast to impregnation, in-situ polymerization offers superior control over PCM distribution, as the supporting network is established during composite fabrication. This approach enhances interfacial interactions between cellulose and PCM, leading to improved structural integrity, leakage resistance, and more uniform thermophysical properties. These advantages, however, entail more intricate processing and stricter requirements for precursor compatibility, polymerization kinetics, and reaction control. Consequently, in-situ polymerization is particularly beneficial for multifunctional CA-PCMs, where structural uniformity and long-term durability are prioritized over processing convenience.

Experimental evidence indicates, CNF can be combined with binding agents such as calcium methacrylate and pentaerythritol tetra(3-mercaptopropionate). This method rapidly forms a three-dimensional network with many cross-links quickly because certain calcium ions connect with the carboxyl groups of CNF, and the vinyl groups click with the thiol molecules of pentaerythritol tetra (3-mercaptopropionate) (Fig. 5c). This network structure makes it possible for PCMs with different phase change temperatures to exhibit good phase change enthalpy, better thermal stability, and improved cycle stability [128]. In another method, PEG is incrementally included in a CNC-CHO suspension, subsequently followed by the use of melamine. Acetic acid is added to commence the Schiff-base reaction between the CNC-CHO groups and melamine, promoting cross-linking and gelation. The resultant gel was freeze-dried to yield 3D aerogels, as shown in Fig. 5d. The CNC/PEG phase change aerogel composites exhibited remarkable shape stability, a high phase change enthalpy of 145.8 J/g, and outstanding cyclic reversibility, preserving good performance after 100 heat cycles [108]. This method allows the PCM to be embedded more uniformly.

4.3.3. Encapsulation

In the encapsulation method, the PCM is encapsulated in micro- or nano-capsules, which are then embedded into the CA structure. In this method, an active substance termed as core (PCM) is surrounded by a protective matrix, which is termed as wall material, resulting in the formation of capsules. This method is used to improve the material's stability and reduce leakage. Encapsulation is particularly advantageous in scenarios where long-term structural stability and leakage prevention are prioritized over maximum latent heat storage. Unlike impregnation, in which PCM directly permeates the aerogel pores, encapsulation involves the physical confinement of PCM within micro- or nanosized shells prior to its integration into the cellulose matrix. This approach considerably improves thermal reliability, mechanical robustness, and cycling stability; nevertheless, it always includes inactive shell elements, hence diminishing the overall energy-storage density. Consequently, encapsulation is more beneficial for applications necessitating extended service life, enhanced safety, and regulated thermal release rather than optimal latent heat capacity.

Representative studies include, paraffin wax (PW) is augmented with CNCs as Pickering emulsifiers that are then embedded into silica

aerogels (SA) stabilized by polyvinyl alcohol (PVA) binders (Zhang et al.; Fig. 6a). The composite comprises 63 wt% PW and exhibits a substantial latent heat capacity of 106 J/g. The material's distinctive blend of low heat conductivity and low density characteristics is due to the homogenous distribution of hydrophobic, porous SA inside the PVA matrix [111]. Similarly, n-hexadecane, as shown in Fig. 6b, using oil-in-water Pickering emulsions kept stable by CNF that had not been chemically tuned. The microcapsules got as big as $12 \pm 3.4 \mu\text{m}$ in diameter, and CNF covered 67% of their surface area, which matches the fact that the freeze-dried capsules had a porous shape. The PCM-CNF microcapsules had a PCM loading of 59% and could store and release 132.5 and 141.1 J/g of heat energy, respectively. The microcapsule slurry also showed a change in storage stiffness that could be undone by an order of magnitude across the n-hexadecane phase transition temperature [109]. This technique helps in controlling the release and absorption of heat. Encapsulation strategies sacrifice direct contact between PCM and heat transfer additives in exchange for enhanced leakage prevention and mechanical robustness. On the other hand, the direct mixing methods allow simultaneous incorporation of PCM and additives in a single step, potentially achieving superior thermal performance at the cost of reduced control over final morphology. The final fabrication approach, described below, exemplifies this simplified one-pot synthesis pathway.

4.3.4. Direct mixing

In the direct mixing method, PCM is directly mixed with the cellulose matrix, where the PCM solidifies inside the aerogel's structure upon cooling. Representative work by Wang et al. synthesize PEG/MXene-CA as freeze-stable hybrid PCMs using a one-step in situ encapsulation approach based on freeze casting (Fig. 6c). They first mix MXene colloid with 1.34 wt% cellulose hydrogel in varying mass ratios and stir the mixture at room temperature for 2 h. Then add 90 wt% PEG and continue stirring at 65°C for 3 h. The synthesized aerogels demonstrated augmented light-to-heat conversion, better thermal conductivity, and higher thermal retention capabilities [121]. In another study, they prepare form-stable PCMs from PEG/CA-dopamine using an in situ freeze casting method (Fig. 6d). They begin by vigorously stirring cellulose hydrogel at room temperature, then add dopamine hydrochloride. After thorough mixing, they introduce PEG and continue stirring at 65°C for 3 h. Finally, the composite was freeze-dried to obtain the aerogels. Adding CA and dopamine together had a positive effect that made it possible for the PCD fs-CA-PCMs to have a very high encapsulation capacity of about 95% by weight and a latent heat of about 194.3 J/g. The new way that PEG, CA, and dopamine interacted with each other changed the hydrogen bonding network, which explained the higher latent heat [82].

In terms of thermal performance, structural integrity, and simplicity of production, each of the four fabrication strategies for CA-PCMs demonstrate unique advantages and constraints. During impregnation, molten PCM is poured into the porous structure of the CA. This creates a high heat storage density and PCM loading capacity (for example, a melting enthalpy of 325.4–330.6 J/g), but the pores may not be fully covered. In-situ polymerization allows for homogeneous integration of PCM during gelation or drying, resulting in composites with high phase change enthalpy (e.g., 145.8 J/g) and outstanding cyclic stability, this method associate some challenges like monomer diffusion in the polymer matrix and reaction compatibility. Encapsulation, with latent heat capacities of up to 141.1 J/g, provides regulated heat release and absorption while maintaining structural integrity by embedding micro- or nanocapsules containing PCM into the aerogel matrix, this method faces challenges due to low encapsulation efficiency and thermal stability of the shell material. When PCM and cellulose gels are mixed directly, the manufacturing process is sped up. This creates aerogels with high packing efficiency (up to 95%) and latent heat values of about 194.3 J/g, for this approach to be successful it gets very important to choose the matrix and PCM very wisely as if they are not compatible it can lead to

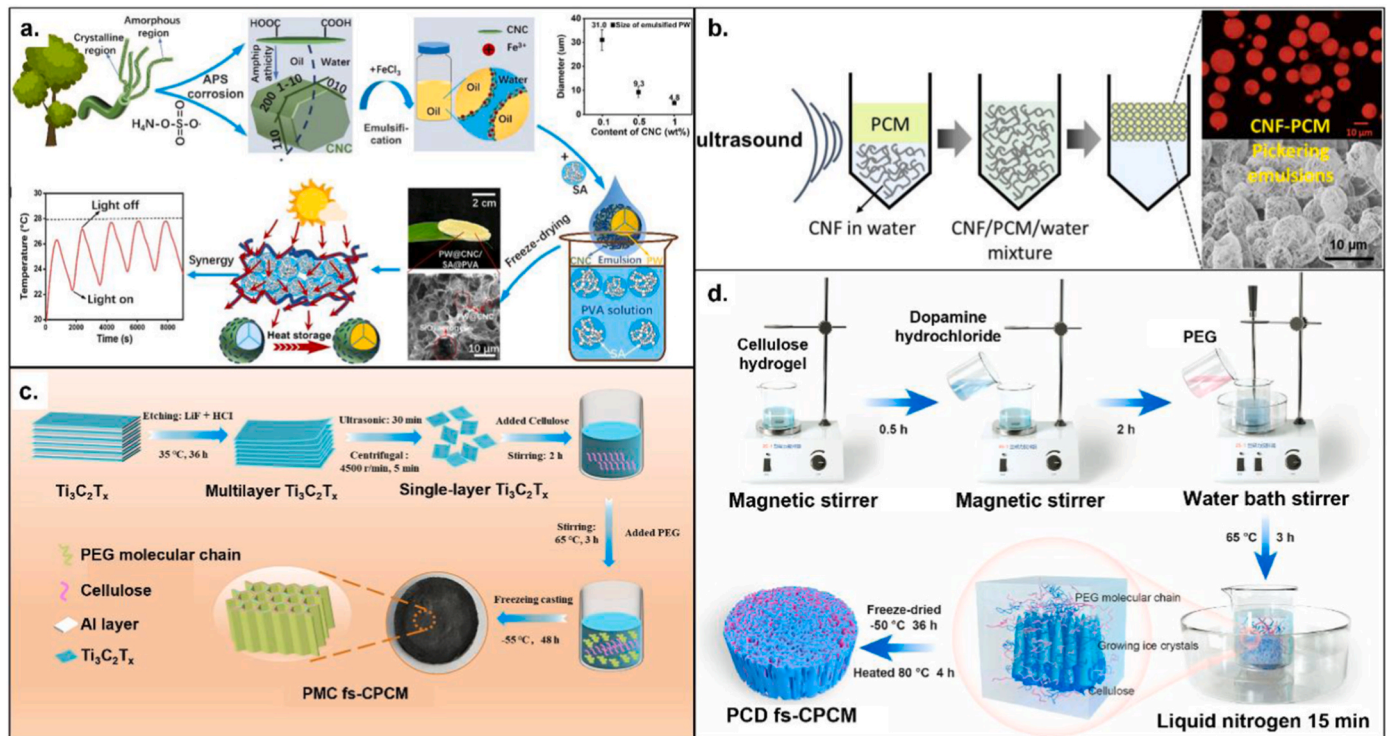


Fig. 6. (a.), (b.) Fabrication of CA-PCMs by encapsulation [109,111]. Adapted from Ref. [111] with permission from the American Chemical Society, copyright 2024, and adapted from Ref. [98] with permission from Elsevier, copyright 2022 (c.), (d.) Fabrication of CA-PCMs by direct mixing [82,121]. Adapted from Ref. [121] with permission from Elsevier, copyright 2022, and adapted from Ref. [82] with permission from Elsevier, copyright 2021.

leakage issues. Collectively, these strategies underscore the adaptability of CA-PCMs in customising thermal properties and structural designs for a wide range of applications in thermal management and energy storage systems.

The four primary ways of manufacture of CA-PCMs differ process complexity, distribution of PCM, resistance to leakage and thermal performance. Impregnation is the simplest way and usually has a high loading of PCM, although it is highly sensitive to pore accessibility and interfacial compatibility. Table 3 presents the comparison between all these methods. In-situ polymerization usually gives greater structural integration and more uniform dispersion of PCMs, while encapsulation is particularly useful when leakage control and long-term stability are of primary importance. Direct mixing has the advantage of simple processing, which is scalable, but formulation control must be carefully optimized to ensure morphological stability and prevent leakage.

4.4. Properties of CA-PCMs

CA-PCM material characteristics, such as density, morphology, and thermal conductivity, notably impact their phase change properties, such as phase change efficiency and thermal energy storage (Fig. 7).

Density is an important feature affecting how well heat moves through an aerogel matrix and how well PCMs can be incorporated in the matrix. The low density and corresponding high porosity of CNF aerogel allow for successfully catching and holding PCMs, which makes them a good candidate for storing energy [129]. Furthermore, the Low density allows for high PCM loading without compromising thermal insulation performance, which is needed for efficient heat storage [98, 130].

Processing parameters during the composite formation highly influence the morphology of CA-PCMs, including pore structure and distribution. This in turn directly influences thermal absorption and emission efficiency of CA-PCMs during phase transitions and thermal energy storage effectiveness. Interconnected voids of CA enhance PCM

encapsulation, minimizing phase transition leakage and ensuring stability over prolonged thermal cycles. The porous open-cell structure of CA matrix provides the PCMs with an extensive surface area for interaction, hence enhancing heat storage capability in the matrix. Morphology influences the mechanical stress resistance of the material. This indicates that CA-PCMs shape is crucial for enhancing performance and yielding high-quality PCMs with superior thermal and mechanical properties [131,132].

The thermal conductivity of CA matrix plays a crucial role in heat absorption and release during the phase change because organic and eutectic PCMs exhibit low thermal conductivity. The low thermal conductivity reduces the heat transfer rate and thereby increases the energy storage time, leading to a prolonged phase change which results more effective thermal energy storage [133,134]. However, heat transport also plays a vital role for rapid latent heat storage and release. To enhance the heat transport within the matrix, the strategies via certain nanofillers, such as graphene [135], CNTs [113], MXenes [127], or conductive polymers [136] are incorporated to expedite heat transport and may be desirable in situations requiring rapid heat storage or release. Thus, achieving and optimizing the thermal conductivity of CA-PCMs greatly governs the phase change characteristics of the final composite material. Nanofiller integration time impacts manufacturing. In-situ polymerization creates coherent nanofiller networks during matrix synthesis, whereas direct mixing integrates phase change materials and fillers, improving thermal conductivity. The fabrication method determines heat conductivity and complexity to meet targets.

4.4.1. Connecting to application requirements

Application domains possess varying requirements for thermal properties. Effective thermal management in buildings necessitates substantial latent heat capacity and moderate thermal conductivity to reduce response time to daily temperature fluctuations (8-12 h). Electronics require robust thermal conductivity to dissipate heat rapidly. Aerospace thermal protection must maintain stability after thousands of

Table 3
Comparison of major fabrication strategies for cellulose aerogel-based phase change materials.

Method	Definition	Advantages	Limitations	Typical performance tendency
Impregnation	PCM is absorbed into a pre-formed CA, usually by vacuum impregnation.	Simple process, relatively low cost, high PCM loading, easy implementation.	May not ensure complete or uniform pore filling; leakage may still occur if pore structure or surface chemistry is not favorable.	Often provides high latent heat and high PCM loading.
In-situ polymerization	PCM or PCM-related monomers are integrated during gelation or network formation within the CA.	More uniform PCM distribution, strong network formation, improved shape stability, good cycling durability.	More complex chemistry; requires compatibility among monomers, cellulose matrix, and additives.	Often provides balanced thermal performance with good structural stability.
Encapsulation	PCM is first enclosed in micro- or nanocapsules, which are then incorporated into the aerogel matrix.	Excellent leakage prevention, improved mechanical robustness, and controlled heat release behavior.	Shell materials may reduce effective heat transfer; encapsulation efficiency may be limited.	Suitable for stable and safe thermal storage, although latent heat may be lower than direct loading methods.
Direct mixing	PCM and cellulose components are mixed together before gelation, freezing, or drying.	Simple one-pot process, fewer fabrication steps, good potential for scale-up, high packing efficiency when optimized.	Less control over final morphology; poor compatibility may lead to leakage or phase separation.	Can provide high encapsulation efficiency and enhanced thermal performance when formulation is optimized.

cycles. Cold-chain logistics must operate in humid conditions. Wearable thermal regulation requires mechanical pliability and low density. By understanding how fabrication methods control properties, researchers can select methods that naturally produce materials meeting their application requirements, minimizing the need for costly post-processing modifications.

5. Different type of CA-PCMs in thermal energy storage

A diverse array of enhancers, such as MXenes, graphene nanoplatelets, CNT, boron nitride, carbon fibre, and conductive polymers like polyaniline and polypyrrole, have been widely incorporated into CA-PCM composites to enhance thermal efficiency [137–141]. This section provides an in-depth exploration of the several types of CA-PCMs and their hybridization with other materials like graphene, MXene, CNT etc. Table 4 discusses the phase change properties of these CA-PCMs.

5.1. CA-PCMs

Pure cellulose aerogel-based PCMs represent the fundamental design platform upon which subsequent hybrid systems have been developed. In these materials, the porous cellulose network alone provides capillary confinement, structural support, and leakage resistance without relying on conductive nanofillers or multifunctional additives [153–158]. Consequently, studies on pristine CA-PCMs establish the baseline relationships among pore architecture, PCM loading, latent heat retention, thermal insulation, and cycling stability, thereby providing an essential reference for evaluating the benefits and limitations introduced by graphene, MXenes, CNTs, polymers, and other reinforcing phases discussed in the following sections.

Song et al. employed CNFs to stabilize paraffin-based PCMs in a Pickering emulsion system. The emulsion was subsequently incorporated into a 3D interconnected network of CNFs to produce a composite aerogel (Fig. 8a). The resulting aerogels have exceptional strength and can withstand loads up to 5000 times its weight. Furthermore, it retains its structural integrity and effectively resists any leakage after undergoing 20 heating and cooling cycles at 80 °C, as shown in Fig. 8b. The CNF/PCM composite aerogel has a latent heat capacity of 173.59 J/g, which is ~84.4% of the latent heat capacity of paraffin. Moreover, its thermal conductivity is low and does not exceed 37.7 mW/m K. Thus, this composite aerogel enhances remarkable thermal insulation and durability (Fig. 8c) [19].

Guo et al. employed the Pickering method to efficiently encapsulate paraffin into a 3D porous crosslinked structure formed by CNTs and methyltrimethoxysilane (MTMS), stabilized through chemical and hydrogen bonding (Fig. 8d). The resulting structure exhibits superior multifunctionality, including hydrophobicity, compressive strength, photothermal conversion, and effective thermal energy storage and release. The materials also maintain hydrophobicity and compressive strength under light-induced heating conditions and remain structurally stable up to 100 thermal cycles (Fig. 8e). Additionally, under irradiation at 100 mW/cm², the aerogels reach temperatures exceeding 80 °C and demonstrate the ability to adsorb and recover viscous crude oil (Fig. 8f) [147].

Wang et al. adopted the freeze casting technique, which is a simple one-step in-situ method to develop form-stable CA-PCMs (PCD fs-CA-PCMs) using PEG and dopamine-modified CA. The resulting PCD fs-CA-PCMs were found to encase up to about 95 wt% of the CA and produce a latent heat of roughly 194.3 J/g owing to the synergistic activities of CA and dopamine. Adding dopamine enhanced the latent heat by around 18.9% compared to the materials without alteration (Fig. 9a). This increase in the latent heat was mostly due to the weakening of hydrogen bonding caused by the combination of PEG, CA, and dopamine (before and after modification: O-H O > O-H N or N-H O), which made surfaces less confined and improved surface-induced phase change

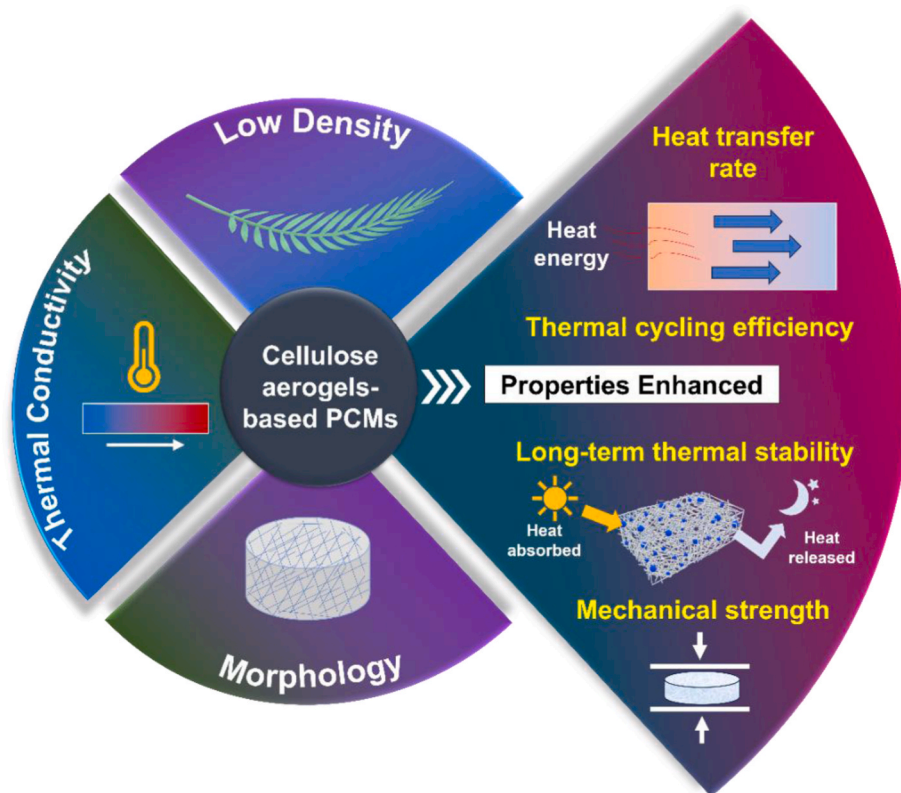


Fig. 7. Illustration of enhanced properties of CA-PCMs.

behaviour. Furthermore, it effectively minimized the difference between the theoretical and experimental results. This suggests the alteration improved PEG's phase transition behavior and heat storage capabilities [82].

Zhou et al. used UV-induced thiol-ene click chemistry to create CNC hydrogels with vinyl-functionalized CNCs, then maintained the integrity of the CNC network by impregnating it with molten PEG through a solvent-exchange process (Fig. 9b). The resulting PEG/CNC-PCM retains PEG's chemical and crystalline structure, with PEG loading reaching a maximum of 97 wt%, exhibits a high latent heat capacity of over 150 J/g, and resulted in a drop in PEG's crystalline phase transition temperature from 40.1 °C to 33.5 °C. This may be due to the CNC nanofibers having a high surface area and the strong hydrogen bonding between CNC and PEG. Notably, the thermal stability of the PEG/CNC PCM is robust, and it has a fast exothermic response and a high thermal conductivity of 0.44 W/m K, (Fig. 9c). In summary, the hybrid PCM composite material demonstrated a 3D CNC network with effective PEG loading, followed by phase transitions and undergoing temperature changes while retaining high mechanical integrity and clarity [159].

Li et al. used directional freeze drying and vacuum impregnation techniques to create porous hybrid PCM aerogels composed of CNFs, silver nanowires (AgNWs), and encapsulated octadecanol (OCO) and octadecane (OCC). The obtained material had a good order structure and anisotropic thermal transmission properties. Due to the tight contact between silver nanowires (AgNWs) and CNFs, energy from NWs induced by solar light can be efficiently transferred to the CNFs. This interaction increases the transmission of phonons within the cellulose lattice and facilitates heat transfer. There is a considerable improvement in the thermal conductivity of CA-PCMs by 72.7%, and the thermal time constant is within a close range of the expected. Fig. 9d-f shows the potential of CNF/AgNWs hybrid aerogels in thermal energy storage systems [160].

Zheng et al. used a direct fabrication approach to produce PCM/CNF composite monoliths by formulating a PCM/CNF ink, in which a small

amount of CNF gel acted as both a stabilizer and a viscosity modifier, enabling high printability for Direct Ink Writing. The process involved incorporating microencapsulated paraffin-based PCM into the CNF matrix, followed by ice templating and freeze-drying to form a robust 3D-interconnected CNF scaffold. This structure allowed for high PCM loading (up to 82% by weight of the composite) and substantial heat storage capacity (up to 153 J/g). The resulting composite monolith aerogels exhibited a high Young's modulus (up to 1.14 MPa), with enhanced thermal stability. These monoliths also retained their microscopic morphology and thermal storage properties even after prolonged exposure to heat at 80 °C, owing to the CNF coating on PCM particles and the mechanically resilient CNF network [97].

Collectively, these studies demonstrate that pristine CA-PCMs provide an effective baseline for understanding the relationships among pore architecture, PCM loading, latent heat retention, and cycling stability. Future advances should therefore focus on scalable fabrication routes and microstructural engineering that preserve these advantages while enabling industrial implementation.

5.2. Cellulose/graphene aerogel-based PCMs

Graphene is widely recognized for its outstanding thermal conductivity of 3000 to 5000 W/m K at room temperature. The additional benefit of using it is broad solar light absorption spectrum, substantially improving solar light absorption, solar-thermal energy conversion, and thermal conductivity in PCMs [130,161,162]. Various forms of graphene, including 2D graphene sheets, 3D graphene networks, graphene oxide, and graphene nanoplatelets, offer advanced characteristics for thermal management [163–166]. Integrating graphene into CA matrix creates efficient thermal conduction pathways, reducing phonon scattering between graphene layers and lowering thermal resistance at the interfaces between graphene and PCMs [167]. Which results in the rapid energy storage and release during the phase change of the CA-PCMs due to the thermal conduction provided by the graphene moiety.

Table 4

Representative thermophysical performance of reported cellulose aerogel-based phase change materials used for comparative analysis in this review.

S. no	Representative CA-PCM	T _m (°C)	H _m (J/g)	T _c (°C)	H _c (J/g)	Ref.
CA-PCMs						
1.	CNF/PEG-based PCM aerogels	48.3	164.3	36.0	152.0	[128]
2.	CNC/PEG-based PCM aerogels	57.3	145.8	30.6	140.3	[108]
Cellulose/graphene aerogel-based PCMs						
3.	PEG-grafted CNF/graphene hybrid aerogels	69.8	187.4	35.2	173.9	[81]
4.	PEG/CNCs/graphene hybrid aerogels	66.4	183.3	34.3	167.0	[107]
5.	Melamine foam/GNP/CNF/Paraffin wax composite aerogels	52.4	154.3	46.7	159.7	[142]
6.	Cellulose/expanded graphene/palmityl alcohol	45.6	147.1	45.4	146.3	[143]
7.	PEG/CNFs/graphene aerogel	53.96	167.5	32.94	158.1	[144]
Cellulose/Mxene aerogel-based PCMs						
8.	Bacterial cellulose/MXene/PEG hybrid aerogels	67.0	196.7	41.5	191.7	[17]
9.	CNF/CNT/paraffin wax-based PCM aerogels decorated Ti ₃ C ₂ T _x laminated fabric	50.5	133.0	44.0	140.2	[68]
10.	Bacterial cellulose/polyvinyl alcohol/Ti ₃ AlC ₂ /PEG aerogel	64.91	172.94	41.02	146.63	[16]
11.	CNF/PEG/TiC	38.59	107.95	27.49	-	[145]
Cellulose/CNT aerogel-based PCMs						
12.	CNF/CNT/PEG composites aerogels	63.55	158.3	35.0	157.1	[146]
13.	CNT-doped photothermal cellulose-based aerogels	51.74	130.23	47.00	127.80	[147]
14.	CNT/CNF/paraffin aerogels	50.5	133.01	44.0	140.0	[68]
15.	BC/MWCNT/paraffin aerogels	62.5	172.0	49.3	157.9	[148]
Cellulose/polymer aerogel-based PCMs						
16.	CNF/polypyrrole/n-octacosane hybrid aerogels	64.8	260.2	50.1	257.8	[83]
17.	CNF/polydopamine/n-octacosane hybrid aerogels	64.1	263.1	56.9	262.1	[136]
18.	CNF/polyurethane/octadecane hybrid aerogels	37.6	190.0	16.6	189.7	[95]
19.	PEG/CNF/waterborne polyurethane	61.14	132.63	28.42	126.68	[149]
Miscellaneous						
20.	carboxymethyl cellulose/BN/lauryl acid-based aerogels	41.02	185.1	40.15	184.3	[150]
21.	CNF/Paraffin-3D printed aerogels	63.0	150.0	43.6	141.0	[97]
22.	CNF/melanin/PEG-based hybrid aerogels	67.9	168.3	38.1	161.8	[151]
23.	BNNS-g/CNF/PEG	39.9	146.1	21.0	143.8	[152]

Su et al. created reduced graphene oxide/cellulose sodium frozen aerogels (rGCA) combined with a binary eutectic PCM consisting of lauric acid and myristic acid. With a phase change temperature around 34 °C, the rGCA melting latent heat is 124.6 J/g with 99.7% adsorption efficiency, and the composites exhibit high photothermal conversion efficiency (~96.5%), enabling effective temperature regulation for personal thermal management (Fig. 10a-c) [70].

Shu et al. utilized vacuum-assisted paraffin impregnation to fabricate vertically aligned graphene-CNF (GCA) aerogel composites, starting

with the preparation of GCA and subsequently infiltrating them with paraffin to create thermal-structured PCMs. These composites, containing 3.35 wt% GCA, exhibited a high thermal conductivity of 15.9 W/m K, maintained 98% of their latent heat, and demonstrated excellent form stability, with phase deformation beginning at 55 °C. In addition to effective greenhouse temperature control, the GCA/paraffin composites showcased the ability to convert solar-thermal energy into electricity, achieving an output of 823.2 V under 5 kW/m² solar irradiation (Fig. 10d), highlighting their potential for advanced temperature regulation and solar energy harvesting applications [167].

Wei et al. synthesized aerogels with a porous structure from microcrystalline cellulose (MCC) reinforced with graphene nanoplatelets (GNPs). The aerogels were synthesized with appropriate layering/stacking order arrangements. For the preparation of CA-PCMs, the anisotropic aerogels were blended with PEG to produce CA-PCMs. Composite PEG at a GNP content of only 1.51 wt% had a notable thermal conductivity of 1.03 W/m K and a latent heat capacity of 182.6 J/g, close to unmodified PEG (99.84%). Furthermore, the composite possessed remarkable encapsulation and mechanical stability. On top of this, the CA-PCMs showed a strong capacity for solar energy capturing, conversion, storage, and release. Moreover, a rudimentary heating apparatus was devised to evaluate the efficacy of the CA-PCMs as a thermal safeguard component, as shown in Fig. 11a, along with the mechanisms for energy harvesting, transition, storage, and release procedures [103].

Qian et al. used atom transfer radical polymerization (ATRP) to graft comb-like poly(hexadecyl acrylate) (PA16) onto a cellulose substrate, and then enhanced the thermal performance and physical characteristics of the resulting PCM (CPmG-x) by incorporating modified graphene (GN16) through direct addition during the composite synthesis process. The CPmG-x composites exhibit exceptional flexibility and adaptability after the phase transition of PA16. Adding GN16 enhances the thermal conductivity to 1.32 W/m K (with 9 wt% GN16) and the enthalpy is raised to 103 J/g (with 5 wt% GN16) compared to the composite without GC16, indicating that the CPMg-x composites can effectively harvest and store solar energy (Fig. 11b) [168].

Wei et al. employed a solution-compounding approach to fabricate hybrid CA-PCMs by incorporating CNCs and graphene nanoplatelets (GNPs) into polyethylene glycol (PEG). GNPs function as heat conductors in this configuration, while CNCs act as barriers to prevent PEG leaking during transitions in phase. The addition of CNCs had a considerable favorable effect on the structure of the CA-PCMs while still maintaining their very high thermal conductivity. Composites with 8 wt % CNCs and 4 wt% GNPs exhibited a latent heat of 145.5 J/g, with 88% of the enthalpy contribution originating from pure PEG. Similarly, the thermal conductivity of CA-PCM was 2.018 ± 0.067 W/m K, larger than the thermal conductivity of PEG (0.3 W/m K). Further, these CAPCMs also had high light-to-thermal energy conversion and electrical energy-to-heat conversion capabilities. Therefore, these components can be used effectively to stabilize temperature, thus providing advanced electrothermal management of electronic components in extreme environments [72].

Wu et al. fabricated a form-stable PCM (FSPCM) by embedding PEG into a melamine foam scaffold coated with a hybrid layer of cellulose fibrils and GNPs, resulting in the composite CG@MF/PEG FSPCM. This material, with a melting point around 60 °C, exhibits efficient encapsulation efficiency, high thermal conductivity, and a melting enthalpy of 178.9 J/g. The high elasticity of the melamine foam and the superior photothermal and electrical conductivity of the GNP network enable the composite to display outstanding light- and electro-driven shape memory effects (Fig. 11c). These effects are activated by the phase transition of PEG, allowing the material to change geometry in response to light or electricity. Moreover, the composite efficiently converts light or electrical energy into heat and retains this energy during phase change, shown in Fig. 11d, making it highly effective for thermal energy storage and multifunctional energy conversion applications [169].

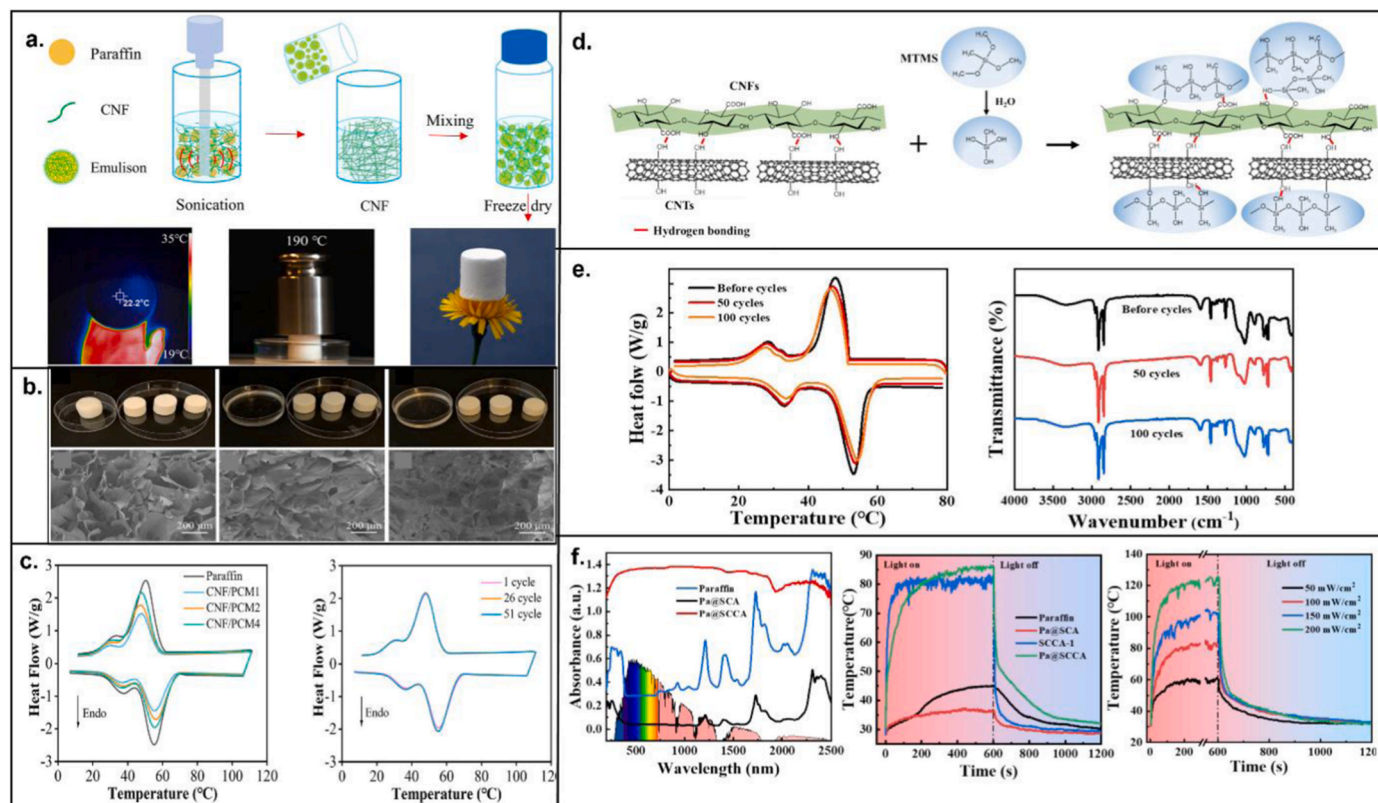


Fig. 8. (a.) Diagram for the synthesis of CNF/paraffin emulsified aerogels. Digital photos illustrating the integrity of the structure of pure paraffin and CNF/PCM composite aerogels, along with SEM images of the CNF/PCM aerogels post-heating at 80 °C for 48 h. (c.) DSC graphs for pristine paraffin and CNF/PCM composite aerogels following 51 thermal cycles of heating and cooling [19]. Adapted from Ref. [19] with permission from Elsevier, copyright 2021 (d.) Diagrams illustrating the interactions among MTMS, CNFs, and CNTs within the hybrid cell wall. (e.) DSC curves and FTIR spectra of composite aerogels post-cycling. (f.) UV-vis-NIR absorption spectrum, the surface temperature of various materials in the air under 100 mW/cm² illumination, and surface temperature in the air under varying light intensities [147]. Adapted from Ref. [147] with permission from Elsevier, copyright 2024.

Li et al. introduced an advanced composite PCMs (CA-PCM) engineered for comprehensive thermal management of Li-ion batteries across diverse environmental conditions. The system is built around a dual-network aerogel matrix, consisting of PVA and carboxylated CNC (PVA/CNC-C), cross-linked using borax, and loaded with tetradecanol (TD) as the phase change medium. To enhance photothermal response, GNPs were integrated into the structure. This CA-PCM exhibits high latent heat storage (199.1 J/g), robust shape stability, and strong mechanical performance, alongside an exceptional photothermal conversion efficiency of 94.5%. These features collectively enable the CA-PCM to buffer extreme temperatures experienced by Li-ion batteries resulting in the prevention of overheating by keeping maximum battery temperatures under 55 °C and providing low-temperature protection by sustaining temperatures above 10 °C for 30 to 40 min. Furthermore, the material can warm batteries for cold-start performance. This work presents a versatile, multifunctional thermal regulation material with energy storage, mechanical integrity, and solar responsiveness that could improve battery safety and performance in high-heat and sub-zero situations [170].

Graphene-based CA-PCMs exhibit the highest improvement in thermal conductivity among existing cellulose-based hybrid systems, while virtually maintaining latent heat storage capacity. Their interlinked conductive networks facilitate photothermal conversion, electrothermal response, and multifunctional energy management, rendering them especially appealing for electronic cooling, solar thermal harvesting, and sophisticated thermal regulation. Nonetheless, achieving uniform graphene dispersion, reducing filler-induced reductions in latent heat, and establishing economically scalable production processes remain the primary obstacles to their widespread industrial use.

5.3. Cellulose/Mxene aerogel-based PCMs

MXenes are an advanced type of 2D nanomaterials with the general formulation $M_{n+1}X_nT_x$. In this notation, M represents a transition metal, X represents nitrogen or carbon, and T_x represents one of several surface terminations such as —OH, —F, and =O. Lately, there has been interest in these materials for their solar-thermal conversion applications. This is due to its great surface area, good conductivity, and efficient solar-thermal conversion capabilities [171–174]. MXenes possess special structural and functional properties that enhance PCMs as they facilitate effective heat transmission and conversion processes [114,121,127].

Quan et al. synthesized a set of composite aerogels by incorporating CNFs, Ti₃AlC₂ MXene, PEG, and Fe₃O₄, and the resulting composites exhibited phase change behavior as well as the ability to convert solar energy into magnetothermal energy (Fig. 12a) They obtained aerogels with a solar-to-thermal conversion efficiency of more than 97.16% and the lowest enthalpy of 151 J/g using freeze-drying and vacuum impregnation processes. Fig. 12b shows the heat release rate and sub-cooling of PCMs. The composite aerogels with Fe₃O₄ at 15% concentration but strong magnetothermal conversion and appreciable phase change underwent a temperature increase from 17 °C to 85 °C in 130 s. Moreover, these composites show exceptional thermal stability below 350 °C and preserve their enthalpy values even after 1000 cycles, indicating remarkable cycling stability [175].

Du et al. integrated erythritol into P-MXene/CNF hybrid aerogels using 2D-layered nanosheets of Ti₃C₂T_x MXene (P-MXene) that they developed by selectively etching Ti₃AlC₂ particles and polymerizing dopamine on their surface. These aerogels possess a porous and interconnected 3D structure that efficiently supports erythritol and avoids

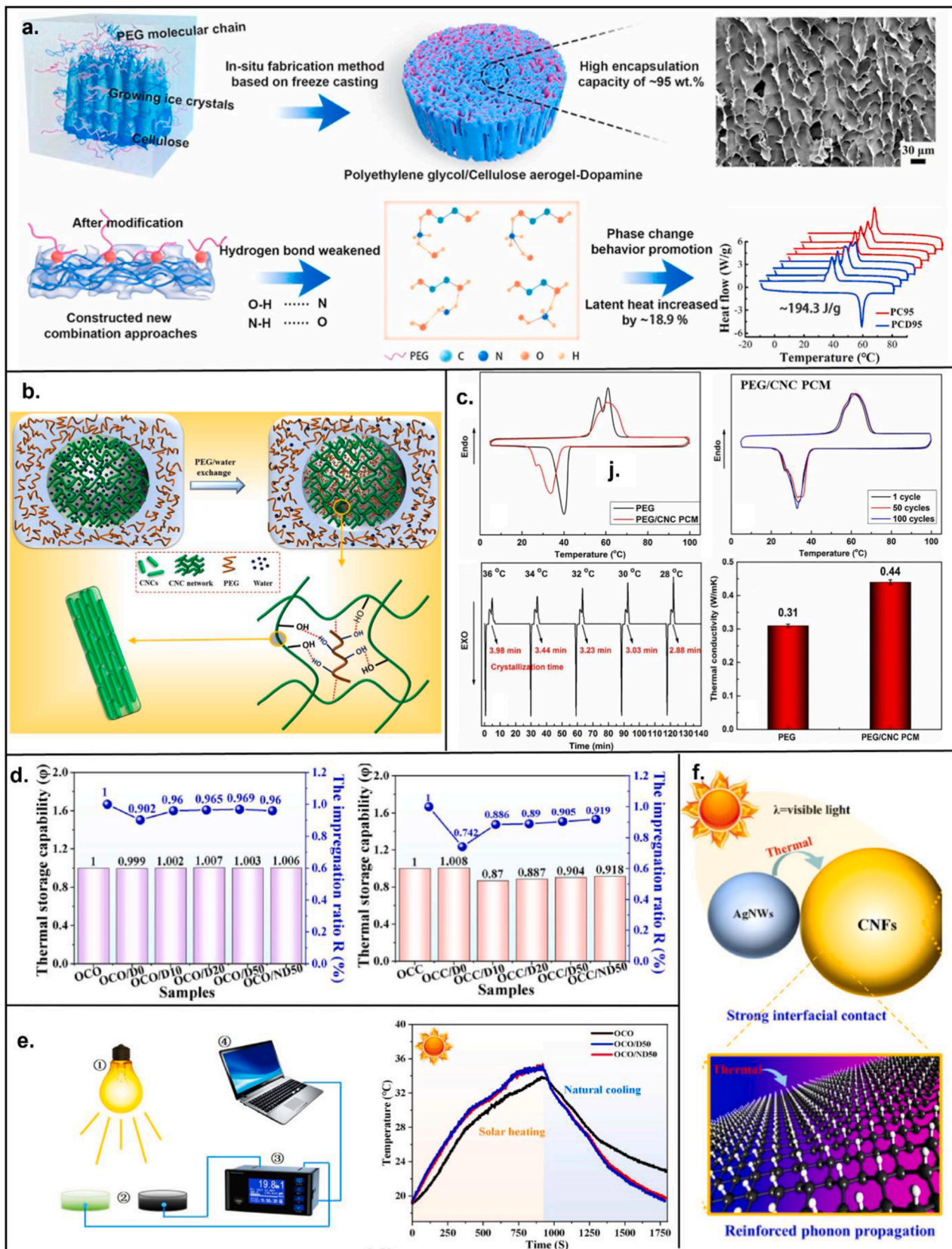


Fig. 9. (a.) Schematic representation of the production procedure for PCD fs-CA-PCMs and the augmentation of latent heat following dopamine modification [82]. Adapted from Ref. [82] with permission from Elsevier, copyright 2021 (b.) Scheme of the solvent exchange and the interaction between PEG and CNC gel. (c.) DSC curves and thermal conductivity of pure PEG and PEG/CNC PCM [159]. Adapted from Ref. [159] with permission from Elsevier, copyright 2020 (d.) Thermal storage capability and impregnation ratio of composite sponges. (e.) Schematic representation of the thermal transfer process of the composite sponges, along with the diagram illustrating the typical thermal conduction strategy of the composite aerogels. (f.) thermal conduction mechanism diagram of the composite sponge [160]. Adapted from Ref. [160] With permission from Elsevier, copyright 2021.

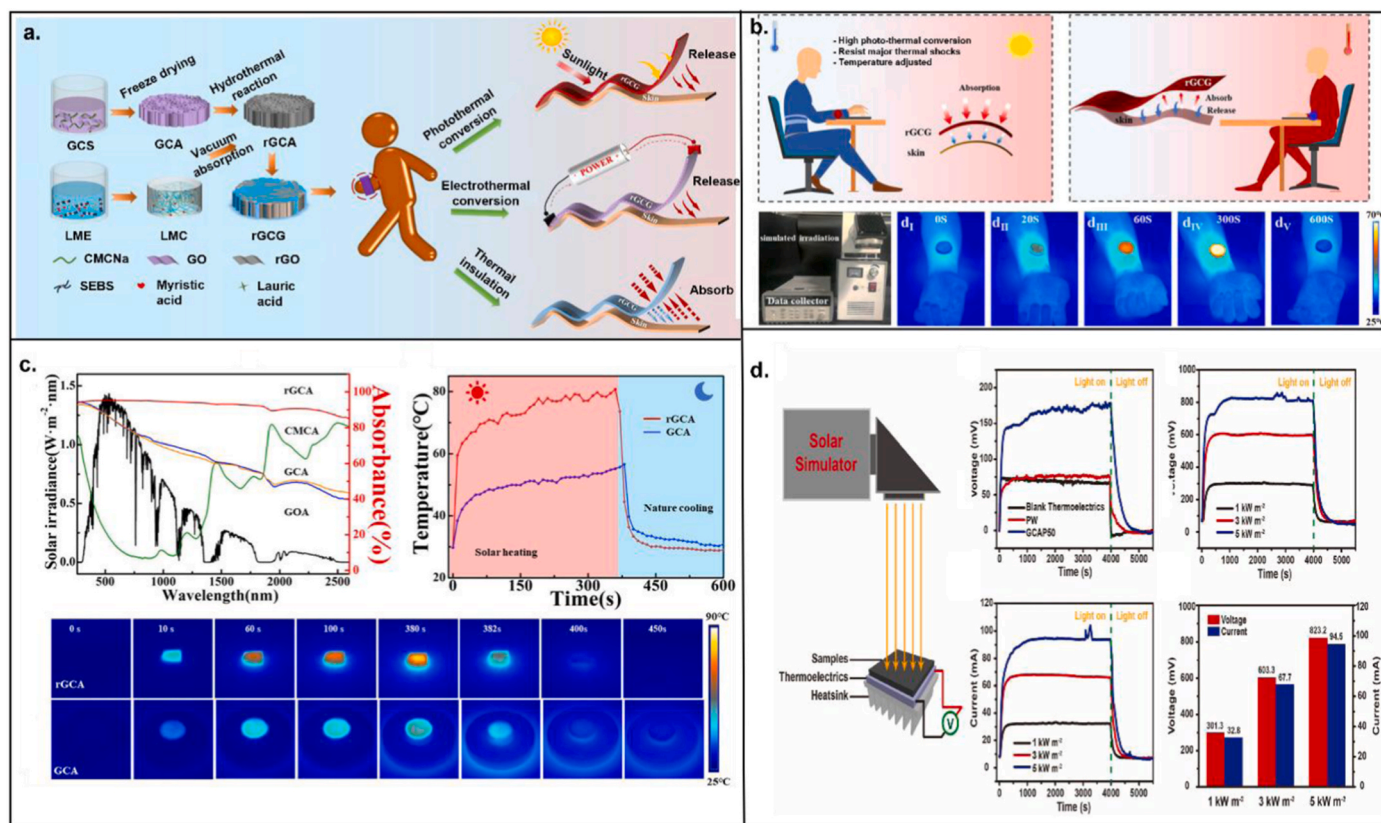


Fig. 10. (a.) Development of innovative thermal management materials and their utilization in personal thermal regulation. (b.) Personal thermal management efficacy of rGCA. (c.) UV-vis-NIR absorption spectra, temperature profiles under simulated sunlight, and infrared thermal pictures during heating and cooling of rGCA for solar thermal conversion and thermal energy storage performance. [70]. Adapted from Ref. [70] with permission from the American Chemical Society, copyright 2024 (d.) Schematic representation of the experimental apparatus for solar-thermal-electric conversion of electricity and output voltage characteristics of GCA-coated thermoelectric devices when the heat sink is subjected to air exposure, immersed in an ice-water mixture, and under varying sun irradiation intensities [167]. Adapted from Ref. [167] with permission from Elsevier, copyright 2023.

any loss of liquid during thermal storage. Based on the findings, these hybrid PCMs retained substantial thermal storage density varying from ~ 325.4 to 330.6 J/g. They also possessed extremely high PCM loading ratios, reaching as high as 1929%. When P-MXene nanosheets were added, the composites had better solar-to-thermal energy conversion, thermal energy storage capacity, solar-to-thermal and electrical energy conversion, and thermal conductivity. Furthermore, these composites exhibited exceptional durability and consistency in terms of heat resistance over extended periods [127].

Wang et al. created PEG/Ti₃AlC₂-CA (PMC fs-CA-PCMs) utilizing a one-step in situ encapsulation process using freeze casting. These aerogels demonstrate enhanced light conversion into heat and increased transport and storage of heat. The light-to-heat conversion efficiency of the photonic metamaterial composite (PMC) systems, which comprised carbon-based PCMs(fs-CA-PCMs), was $\sim 91.6\%$. Such a high value may be attributed to the MXene, a material that absorbs photons and acts as a solar heater. However, developing a conductive network caused by the incorporation of MXene led to improved heat transmission. The increased latent heat capacity of down to 183 J/g in the PMC fs-CP-CMs can be explained by the 90 wt% PEG confined in the aerogel matrix. In addition, the composites exhibited superb chemical compatibility and enhanced thermal stability [121].

Researchers designed CA-PCM scaffolds by cross-linking microcrystalline cellulose (MCC), MXene, and silver-loaded multi-walled carbon nanotubes (Ag-MWCNT) to form a three-dimensional (3D) network hydrogel. This network was processed via cryodesiccation to yield MCC/Ag-MWCNT/MXene (Ag-MMCT) scaffolds with a microcrystalline porous structure capable of forming a multi-level pore system wide enough to accommodate palmitic acid (PA). Using vacuum

impregnation, PA was introduced into the scaffolds to synthesize MCC/Ag-MWCNT/MXene/PA (Ag-PMMCT) CA-PCMs. The resulting Ag-PMMCT materials demonstrated efficient phase form stabilization and high latent heat storage (153.8 J/g), as well as enhanced light-to-heat conversion with a response time of 223 s under exposure. Notably, a 1:1 ratio of MXene to Ag-MWCNT in the composite achieved a thermal conductivity of 0.653 W/m K, representing a 202.3% increase compared to pure PA. These findings highlight the potential of Ag-PMMCT CA-PCMs for applications in energy conversion, storage devices, and thermal management in electronics [177].

Liu et al. fabricated anisotropic CA-PCMs (MXene/CNF-C/PEG aerogels) utilizing a straightforward directed freeze-casting technique, yielding a distinct anisotropic porosity structure. These aerogels exhibited effective absorption of sound waves, conversion of acoustic energy into thermal energy, and capacities for managing thermal conditions, as shown in Fig. 12c. The MXene/CNF-C/PEG aerogels exhibited form stability due to H-bonding between carboxylated CNFs (CNF-C) and PEG, eliminating the need for chemical cross-linking and adding Ti₃AlC₂ as MXene resulted in a 150% increase in thermal conductivity and a substantial improvement in acoustic-to-thermal conversion efficiency. The CA-PCMs had an impressive energy storage density of 143.0 J/g and displayed enhanced acoustic-to-thermal conversion capabilities due to the directionality of the pores (Fig. 12d), indicating that the DMCPs might have potential in thermal storage devices and acoustic energy transducers [176].

Jiang et al. used a vacuum impregnation process, given in Fig. 12e, to fabricate tetradecylamine/MXene/CNC modified form-stable CA-PCMs (TDA/MXene/CNC CA-PCMs), which is the first use of TDA as supporting material. The shape-stable Ti₃C₂T_x MXene and CNC composite

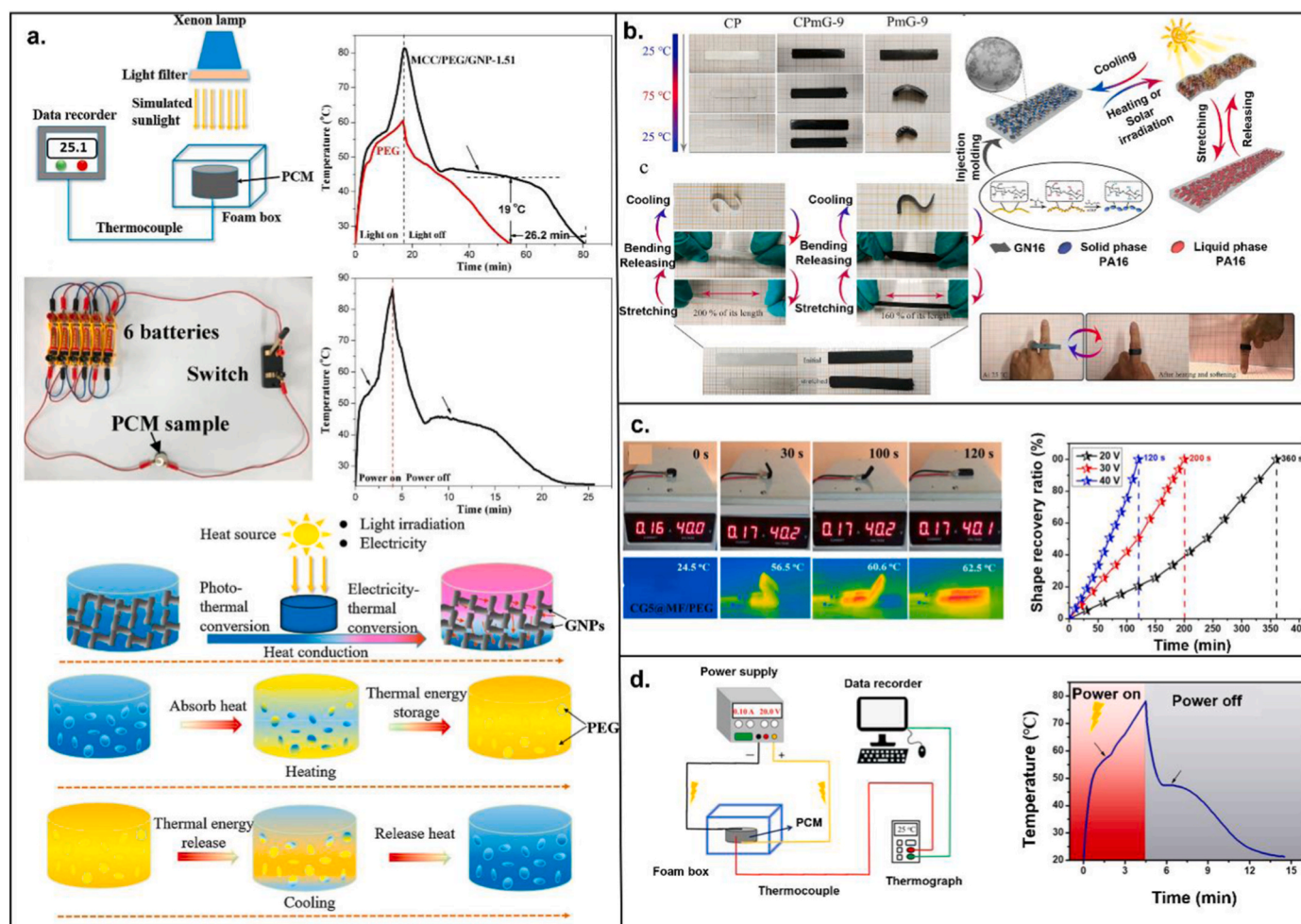


Fig. 11. (a.) The solar energy capturing and electrical energy conversion assessments utilize the representative CA-PCM samples, along with the mechanisms for energy harvesting, transition, storage, and release procedures [103]. Adapted from Ref. [103] with permission from Elsevier, copyright 2019 (b.) Images depicting the heating, bending, and stretching deformations of CP and CPmG-9 at temperatures exceeding and falling below the melting point of PA16 [168]. Adapted from Ref. [168] with permission from the American Chemical Society, copyright 2019 (c.) Digital and infrared thermographic photographs of the electrically induced shape recovery behavior of CG5@MF/PEG under an applied voltage of 40 V, and the associated shape recovery ratios under different applied voltages. (d.) Schematic of the electro-to-thermal energy conversion setup and temperature evolutions of example CG5@MF/PEG sample with or without electrical input. [169]. Adapted from Ref. [169] with permission from the American Chemical Society, copyright 2019.

aerogel have been tested for the first time as the supporting material in the energy storage application of TDA as the PCM. The TDA/MXene/CNC composites possess a latent heat of fusion of 221.5 J/g. The MXene/CNC-2 aerogel had 0.0614 g/cm³ density and 90.6 wt% loading of TDA. The TDA/MXene/CNC composite PCM/cryogel, TDA/MXene/CNC CA-PCM-5, possessed a thermal conductivity of 0.491 W/m K or 1.87 times more than pure TDA. The composites were found to have good potential for use in the cover layer of solar thermal energy collectors (Fig. 12f), with high dimensional stability at 60 °C and improved thermal stability at up to 120 °C. The conversion efficiency from solar to thermal energy was also high, with an efficiency of 94.1% due to increased photothermal conversion because of MXene [90].

Kim et al. introduced a structurally engineered multiscale meta-structure, OT-MXA-EC, that integrates an octet-truss (OT) framework, an asymmetric MXene/TEMPO-oxidized CNF aerogel (MXA), and nicosane (EC) as the PCM. The MXA, fabricated via directional freeze-drying, featured asymmetric lamellar structures and vertically aligned channels that enabled directional heat conduction while suppressing out-of-plane PCM leakage, achieving 98.9% PCM retention under compression. The presence of EC provided considerable latent heat storage, extending sustained heat release by 2.58 times during Joule heating and delaying temperature rise to 60 °C by 304%, functioning as

both an active thermal regulator and thermal buffer. Mechanically, the reported composites shown remarkable strength enhancement by a 6-fold increase at 25 °C where PCM was in solid state and 130-fold at 50 °C where PCM was liquid state, and maintained its structural integrity over 200 thermal cycles. Moreover, the composite shown enhanced EMI shielding, showcasing the potential for efficient electromagnetic attenuation through careful material design. Altogether, the OT-MXA-EC meta structure represents a durable, and multifunctional material with high potential for wearable electronics, energy-efficient enclosures, autonomous systems, and robotics, offering an effective strategy for integrating mechanical robustness, thermal energy storage, and EMI shielding in a single architecture [100].

MXene-based CA-PCMs exhibit superior thermal conductivity, effective photothermal conversion, and remarkable cycling stability, rendering them very appealing for solar thermal energy conversion, wearable heating, and sophisticated electronic thermal control. Their multifunctional properties set them apart from traditional conductive fillers; yet, susceptibility to oxidation, comparatively high material costs, and challenges in production scalability continue to hinder wider industrial adoption. Future advancements should concentrate on enhancing long-term environmental stability while preserving the multifunctional benefits inherent to MXene-based structures.

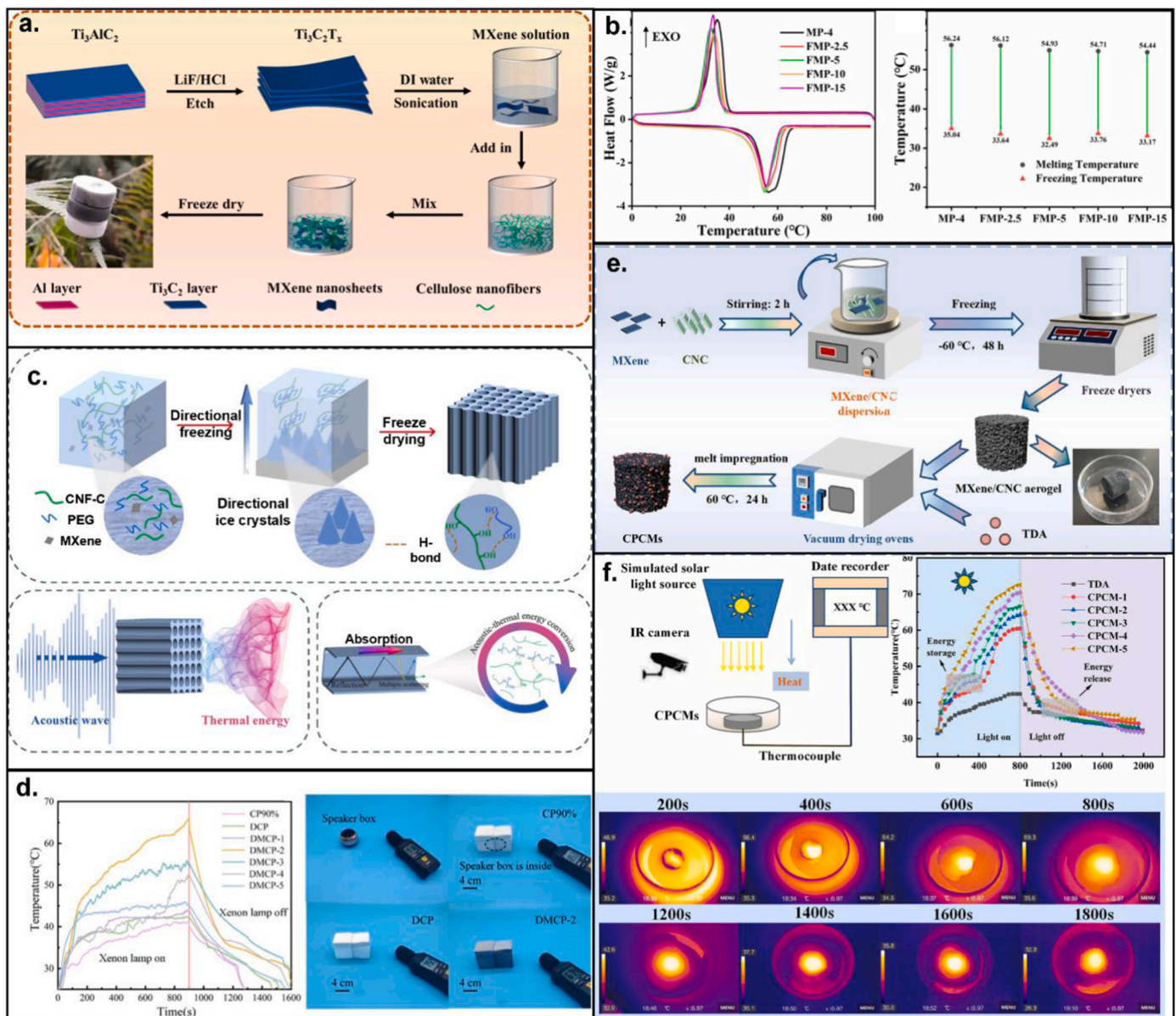


Fig. 12. (a.) Schematic representation of the synthesis of MXene nanosheets and composite aerogels. (b.) DSC cycling graphs of composite PCMs and their degree of subcooling [175]. Adapted from Ref. [175] with permission from Elsevier, copyright 2023 (c.) The production of CA-PCMs through directed freezing and the methods by which aerogels facilitate acoustic-thermal energy conversion via friction damping within the pores. (d.) Curves depicting the conversion of acoustic to thermal energy in aerogels, together with boxes constructed from aerogels, illustrate sound absorption capabilities [176]. Adapted from Ref. [176] with permission from Elsevier, copyright 2024 (e.) The preparation procedure of TDA/MXene/CNC CA-PCMs. (f.) Photothermal transformation graphs of TDA and MXene/CNC CA-PCMs under solar illumination [90]. Adapted from Ref. [90] with permission from Elsevier, copyright 2023.

5.4. Cellulose/CNT aerogel-based PCMs

CNTs are also a promising candidate which can be incorporated to the CA matrix to enhance the conduction within the matrix. These promising nanomaterial recognized for their low-density and outstanding thermal conductivity, establishing them a suitable candidate to overcome the heat transfer challenge in CA-PCMs [178–182]. CNTs have been extensively investigated as additives in CNF aerogels to enhance the thermal conductivity of PCMs.

Du et al. effectively created form-stable CA-PCMs utilizing n-octacosane, and CNFs, where they used CNTs for enhanced heat transfer, as shown in Fig. 13a n-octacosane was infused into hybrid aerogels composed of alkylated CNF and CNT. The alkylation method markedly improved the compatibility of the aerogels with n-alkanes, leading to superior form stability, higher thermal reliability, and increased loading

capacity (Fig. 13b). Differential scanning calorimetry indicated that the CA-PCMs exhibited a substantial phase change enthalpy between 250.9 and 252.9 J/g. Using CNTs enhanced photothermal conversion and storage efficiency from 46.9% to 83.4% and elevated thermal conductivity from 0.247 to 0.583 W/m K. These CA-PCMs thus combine high phase-change enthalpy (~ 251 J/g) with improved heat transfer and photothermal response [71].

Shen et al. prepared foams based on CNF as a porous framework for PEG and paraffin wax to act as barrier inhibitors of encapsulant leakage and added multi-walled CNTs (MWCNTs) for enhanced thermal efficiency. The resultant foams had lower density (less than 67.3 kg/m^3) and higher porosity (more than 94.5%), while the foams possessed targeted structural alteration and attachment with PCM due to changes in chemistry. The highly porous structure allowed a high amount of PCM, with 93.63% for PEG and 91.77% for paraffin wax with low

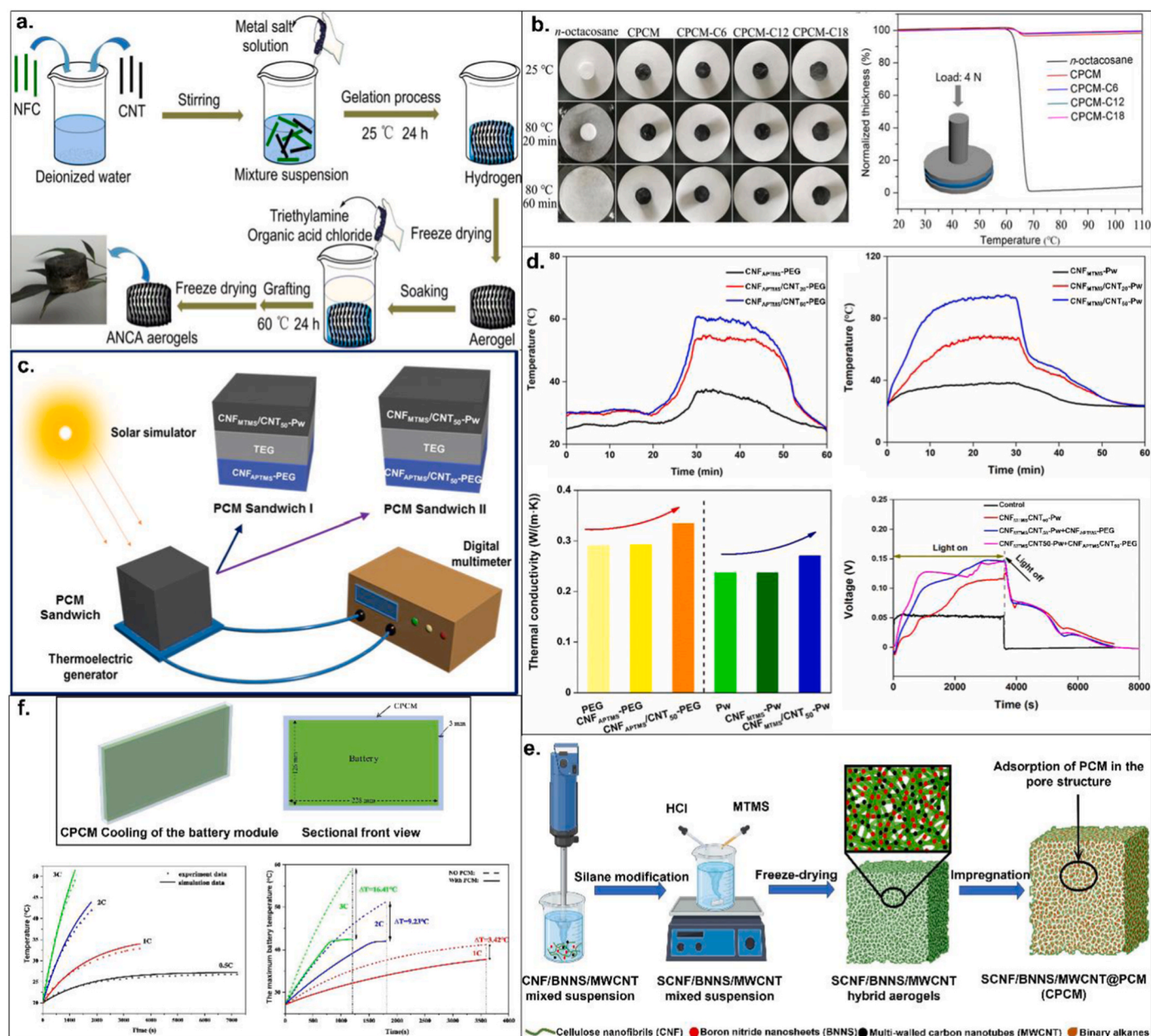


Fig. 13. (a.) Process schematic depiction for CA-PCMs. (b.) Shape stability of *n*-octacosane and CA-PCMs, together with thickness versus temperature curves of the samples under a constant load of 4 N [71]. Adapted from Ref. [71] with permission from the American Chemical Society, copyright 2020 (c.) Diagram depicting the solar-heat-electricity system utilizing innovative PCM sandwich structures. (d.) Temperature-heating time profiles of CA-PCMs under a solar simulator, thermal conductivity measurements of PCM samples, and the temporal progression of the open-circuit voltage of the thermoelectric generator with and without sandwich PCMs [183]. Adapted from Ref. [183] with permission from Elsevier, copyright 2022 (e.) The preparation techniques of SC/B/M@PCM composites. (f.) CA-PCM cooling of the battery module and validation of the 3D battery model at various discharge rates [113]. Adapted from Ref. [113] with permission from Elsevier, copyright 2024.

leakage (<2%). It also showed that heat transfer improved considerably with the addition of CNTs through adequate thermal conductivity and remarkable temperature rise due to solar heating (Fig. 13c and d). CA-PCMs were found to be thermally more stable than the control PEG PCM and had an enthalpy of phase change of 143 kJ/kg at a 25.2 °C melting temperature. The paraffin wax-based PCM also demonstrated a higher melting temperature of 53.4 °C with an enthalpy of phase change at 184 kJ/kg [183].

Zhang et al. introduced a novel approach to fabricating high-performance aerogel fibers by employing a dual gelation technique combined with gravity-assisted alignment. BC serves as the structural framework, while hydroxylated carbon nanotubes (HCNTs) provide photothermal functionality. The resulting silanized BC/HCNT (SBT)

aerogel fibers exhibit an interconnected porous network with high mechanical strength (up to 26 MPa), flexibility, and the ability to be knitted. When infused with the PCM, these fibers (SBTE) achieve impressive thermal energy storage capacity (105 J/g), outstanding mechanical durability (23.9 MPa strength, 8.7 MJ/m³ toughness), and resistance to leakage. Moreover, textiles made from SBTE fibers show sustained heat release under low temperatures, highlighting their strong potential for application in solar-responsive, wearable thermal garments for cold-weather environments [184].

Fu et al. developed a hybrid aerogel (Fig. 13e) by embedding boron nitride nanosheets (BNNS) and MWCNTs as dual thermally conductive fillers into a silanized hydrophobically modified CNF (SCNF) 3D porous flexible structure. This hybrid aerogel was a fibrous material to support a

shape-stable thermal CA-PCM of binary alkane blends. Results of the experiments demonstrated that the porous structure of the hybrid aerogel effectively prevented the leakage of PCM while allowing sufficient flexibility even during the melting process. The incorporation of just 1 vol% MWCNT with BNNS at an optimal 7:3 ratio notably enhanced thermal conductivity, with the SC/35B/15M@PCM composite showing a 91.8% improvement over pure PCM, along with a high PCM content of 94.51 wt% and a latent heat of 213.27 J/g. Moreover, from Fig. 13f, calculations suggested that SC/35B/15M@PCM can reduce the commercial Li-ion batteries' temperature from 3.42 °C to 16.41 °C at discharge rates of 1–3C. The CA-PCM developed in this study features a compromise between the form's stability and density, plus energy transfer [113].

Zheng et al. fabricated a rotunda-shaped aerogel consisting of carboxymethylcellulose and CNTs (CA) by freeze casting with a specialized mold, subsequently impregnating it with PEG. The resultant PEG/CA demonstrated an enthalpy of 183.21 J/g and an effective thermal conductivity of 0.324 W/m K, representing a 57% increase compared to pure PEG. The thermal conductivity of the composite aerogel exhibited a positive connection with the incorporation of CNTs, improving its capacity to transform solar energy into thermal energy. After 10 min of simulated sunlight, surface temperatures reached 65 °C and inside temperatures reached 44.67 °C, achieving 94.41% solar-thermal conversion efficiency, sowing a great potential in solar to thermal conversion. Due to its high solar-thermal conversion and thermal storage capabilities, this PEG/CA may capture renewable solar energy [185].

Wang et al. created CA-PCM scaffolds composed of MCC, MXene, and silver-loaded-MWCNT. Palmitic acid (PA) was incorporated into the scaffold using vacuum impregnation, resulting in MCC/Ag-MWCNT/MXene/PA (Ag-PMMCT) CA-PCMs with hierarchical porosity and efficient light to heat conversion. The Ag-PMMCT scaffolds exhibited a characteristic hierarchical pore structure, offering sufficient capacity for PA encapsulation and establishing an efficient thermal conduction network resulted by MWCNT incorporation. As a result, the Ag-PMMCT materials exhibited superior encapsulation and form stability, a substantial latent heat of 153.8 J/g, and effective light-to-heat conversion, with phase change happening after 223 s of irradiation. The MXene to Ag-MWCNT ratio of 1:1 yielded a thermal conductivity of 0.653 W/m K, representing a 202.3% enhancement relative to pure PA. The Ag-PMMCT CA-PCMs exhibit considerable potential for use in energy conversion, storage systems, and thermal control of devices and electronics [177,186].

CNT-based CA-PCMs provide an optimal equilibrium of enhanced thermal conductivity, mechanical flexibility, and structural integrity, rendering them especially appealing for wearable thermal management, battery thermal regulation, and flexible energy-storage applications. In comparison to graphene- and MXene-based systems, carbon nanotubes (CNTs) offer superior mechanical compliance while preserving effective heat-transfer channels. Nonetheless, obstacles related to uniform CNT dispersion, production expenses, environmental factors, and large-scale processing persistently hinder wider commercial implementation.

5.5. Cellulose/polymer aerogel-based PCMs

Polymers are crucial in CA-PCMs because they provide shape stability, suppress leakage and improve mechanical robustness over repeated phase-change cycles [187,188]. Conductive or photothermal polymers such as PPy, polyaniline and polydopamine further enhance thermal conductivity and solar-to-heat conversion [189,190], while polymer networks improve PCM encapsulation and loading [99].

Meng et al. developed a variety of reversible cellulose-based phase change gels (CPCGs) by physically enclosing PEG inside a dual 3D network cross-linked with hydroxyethyl methacrylate and carboxymethyl cellulose (CMC). The material has exceptional thermal storage dependability, with PEG encapsulation efficiency and energy storage

density of 80.6% and 99.5%, respectively. The maximal latent heat is 90.6 J/g, which retains 97% of its original enthalpy after 100 cycles (Fig. 14a). CPCGs provide exceptional form stability, cycle endurance, and optical reversibility and are manufactured using ecologically green processes. The phase change gels exhibit a pronounced transmittance range of 25 to 78% at 550 nm, efficiently filtering UV radiation upon melting. Adding a small quantity of CMC (about 1 wt %) resulted in a tensile strength enhancement of around 131% in the phase change gel, markedly improving its shape stability. CPCGs maintain a degree of haze when clear, which mitigates glare in optical applications (Fig. 14b). Their adaptability facilitates progress in PCM research, intelligent optics, building thermal regulation, and other fields [191].

Wu et al. fabricated anisotropic hybrid aerogels using a directed freeze-drying technique that incorporated cellulose, silver nanowires (AgNWs), graphene nanosheets (GNPs), and polyacrylamide, which created aerogels (CPGxAy). Using a vacuum-assisted impregnation technique, these aerogels were found to be x-promising as they included PEG structures. As a result, the PEG@CPGxAy ss-CA-PCMs achieved enhanced thermal conductivity values due to the synergistic effect of the graphene/polyacrylamide/cellulose/PEG composites with specific volume ratios of GNPs and AgNWs. Most importantly, these values represented a 200% improvement in thermal conductivity from pure PEG. They also showed anisotropic mean electromagnetic interference (EMI) shielding effectiveness along the transverse and longitudinal axes directions of 71.08 dB and 35.21 dB while maintaining decent melting and crystallization efficiency ratios of 93.47% and 93.08%, respectively. Accordingly, the findings of this study open new perspectives for the design and fabrication of ss-CA-PCMs that will serve both thermal and EMI shielding functionalities in advanced electronic systems [194].

Zhou et al. fabricated robust CNC/poly(N-isopropylacrylamide) (PNIPAM) hydrogels using UV light cross-linking to create carbon-carbon bonds and then incorporated hexadecanol as a PCM by a solvent-exchange process involving ethanol evaporation and hexadecanol diffusion. The resulting CNC/PNIPAM (CP) hydrogel architecture was enhanced by the addition of CNCs, which considerably increased porosity and mechanical properties, raising compressive strength and modulus of the composite by 417% and 331%, respectively, compared to pure PNIPAM. The CP/hexadecanol composite achieved a high PCM loading of ~92 wt %, exhibited a substantial latent heat capacity of ~178 J/g, and maintained excellent thermal stability and shape retention both in water and air. Importantly, the composite's surface was coated with a thick hydrophobic PNIPAM layer (Fig. 14c), resulting in strong water repellence demonstrating the suitability for outdoor applications [192].

Tao et al. introduced a shape-stabilized thermal energy collection and storage device that reacts to solar and electrical stimuli. The resultant CA-PCMs had a substantial thermal storage density of 160.9 J/g and showed remarkable phase transition stability, even following 100 heating and cooling cycles. The integration of PPy as a binder among CNTs markedly improved thermal conductivity to 0.64 W/m K, which is 2.56 times superior to paraffin wax. Molecular dynamics simulations demonstrated that in-situ polymerization of PPy by vapor deposition establishes supplementary thermal transfer channels and enhances heat transmission by synchronizing the vibrations of carbon atoms. This novel method may yield substantial insights for developing CA-PCMs with improved thermal conductivity and multifunctional energy applications. Fig. 14d shows electro-to-thermal energy conversion performance under varying operating voltage and electro-to-thermal energy storage efficiency of composite PCMs with different supporting matrix [193].

Zou et al. developed a shape-stable CA-PCM by embedding a eutectic mixture of myristic acid and tetradecanol (MA-TD) as PCM in CA matrix. The porous CA framework served as an effective structural support, suppressing leakage of the PCM during phase transitions and notably enhancing mechanical stability. The resulting M-T@CNF/PVA composite demonstrated superior thermal performance, with a high latent

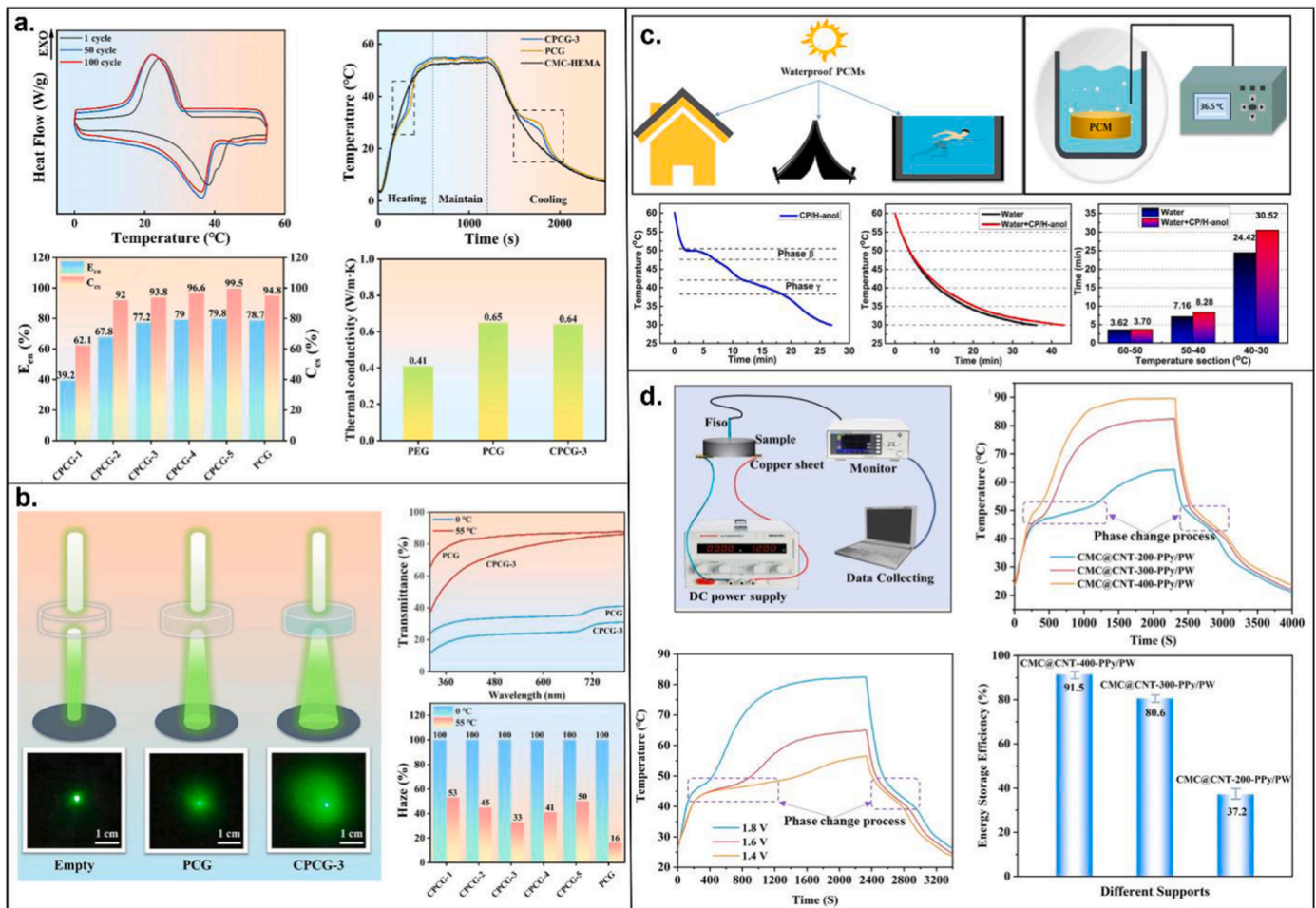


Fig. 14. (a.) Thermal energy storage performance of CPCG-3 including DSC heating/cooling curves, temperature–time profiles, encapsulation efficiency (E_{en}), thermal energy-storage capacity (C_{es}) and thermal conductivity. (b.) Light scattering analysis of CPCG-3 and PCG at elevated temperatures [191]. Adapted from Ref. [191] with permission from the American Chemical Society, copyright 2024 (c.) Potential utilization of waterproof PCMs as outdoor thermal storage substances [192]. Adapted from Ref. [192] with permission from the American Chemical Society, copyright 2020 (d.) Electro-to-thermal energy conversion performance, discussion on measurement setup, temperature evolution under varying operating voltage and electro-to-thermal energy storage efficiency of composite PCMs with different supporting matrix. [193]. Adapted from Ref. [193] with permission from Elsevier, copyright 2023.

heat of 191.5 J/g, low thermal conductivity, and outstanding thermal stability. Even after 100 heating cooling cycles, the material maintained consistent enthalpy, underscoring its reliability and durability. Moreover, these CA-PCMs also proved themselves effective in dynamic thermal regulation, contributing to reduced energy consumption in indoor environments. This study showcases a promising approach for integrating eutectic PCMs in CA framework for sustainable and efficient thermal management solutions [195].

Polymer-modified CA-PCMs are characterized by their superior mechanical robustness, structural flexibility, and long-term thermal reliability compared to existing cellulose-based hybrid systems. Despite their thermal conductivity typically being inferior to that of graphene or MXene alternatives, polymer networks provide superior PCM confinement, substantial latent heat retention, and robust cyclic performance. Thus, these technologies are especially appropriate for adaptable thermal management, intelligent textiles, and architectural applications where mechanical resilience is as crucial as thermal efficacy.

5.6. Other CA-PCM composites

In addition to the more widely developed composites discussed earlier, the incorporation of other promising functional additives, such as boron nitride (BN) [196–198], modified cellulose fibers [199,200], lignin-infused cellulose fibers [201,202], gelatine [203,204], silicon

dioxide [205,206], black phosphorus [207], and chitosan [115,208, 209] has also been investigated to enhance CA-PCM thermal conductivity, mechanical strength, and phase stability. This section highlights the role of diverse additive materials in optimizing the performance of CA-PCMs.

Lei et al. created an anisotropic and resilient 3D-interconnected framework of cellulose and boron nitride nanosheets (BNNS) embedded in PEG composites. This was accomplished by self-assembling concentrated cellulose/BNNS aqueous solutions by vacuum impregnation and cold compression. The distinctive architecture enabled the PEG composites to use the elevated in-plane thermal conductivity of BNNSs, enhanced by their effective alignment during cold compression, which diminished phonon scattering among the BNNS routes due to their interlaced connections. The cellulose/BNNS scaffold demonstrated a remarkable compressive strength of 3.03 MPa, similar to that of reinforced concrete. The strength was ascribed to the synergistic reinforcement from the CNF and the robust BNNSs, allowing the material to maintain its form post-PEG vacuum impregnation. The in-plane thermal conductivity of PEG composites reached 4.764 W/m K with only 10 vol % BNNS loading, representing an ~1400% increase relative to pure PEG. Moreover, these composites preserved their original form at temperatures of 101 °C, even when subjected to a load 500 times their weight. The PEG composites exhibited a pronounced latent heat capacity of 136.8 J/g with a comparatively modest loading of BNNS. These

exceptional features indicate considerable promise for energy conversion and storage systems applications [101].

Le et al. combined PEG and modified CNFs, especially lignin-containing CNF (LCNF) and acetylated CNF (ACNF), through various fabrication techniques (Fig. 15a). The PEG/CNF hybrids, known as phase-change nanohybrids (PCN), that are bound by primarily non-covalent interactions and extremely porous, possess ultra-lightweight characteristics, with densities between 0.022 and 0.043 g/cm³, and demonstrate remarkable mechanical robustness alongside a substantial latent heat storage capacity of up to 204 J/g. The PCN systems possess a specific heat capacity of 2.24 J/g K and efficiently adjust the temperature by 2.23 °C with a thickness of merely 1 mm, all while ensuring thermal stability. Notably, the PCNs deliver substantial temperature regulation during extreme sun exposure, providing insulative protection of 33.5 °C with a thickness of 1.5 cm (Fig. 15b). Besides their thermal characteristics, these materials exhibit remarkable acoustic absorption, attaining 100% absorption at 1600 Hz and 50% at lower frequencies

such as 500 Hz. The integration of titanium dioxide (TiO₂) nanoparticles significantly improves the thermoregulating properties of the PCNs, suggesting their use in several multifunctional domains [80].

Zhang et al. created a hydrophobic silicon dioxide-phase change microcapsule/gelatin-hydroxyethyl cellulose composite aerogel employing a simple directed freezing method (Fig. 15c). The prepared material showed a melting temperature of 18.0 °C and an enthalpy of 129.9 J/g. This novel material exhibits remarkable passive daytime radiative cooling properties, with a solar reflectance of 92.61% and an atmospheric emissivity of 95.47%. Field studies demonstrate that, under typical sunshine circumstances of 726.9 Wm⁻², the material attains an average sub-ambient cooling of 4.3 °C. During the night. The prepared composites sustains thermal comfort via the exothermic phase transition of the PCC, yielding a temperature differential of 0.78 °C, thus mitigating the problem of overcooling. Furthermore, they efficiently inhibit heat backflow and preserves thermal stability due to its superior thermal insulation characteristics. These composites has self-cleaning properties

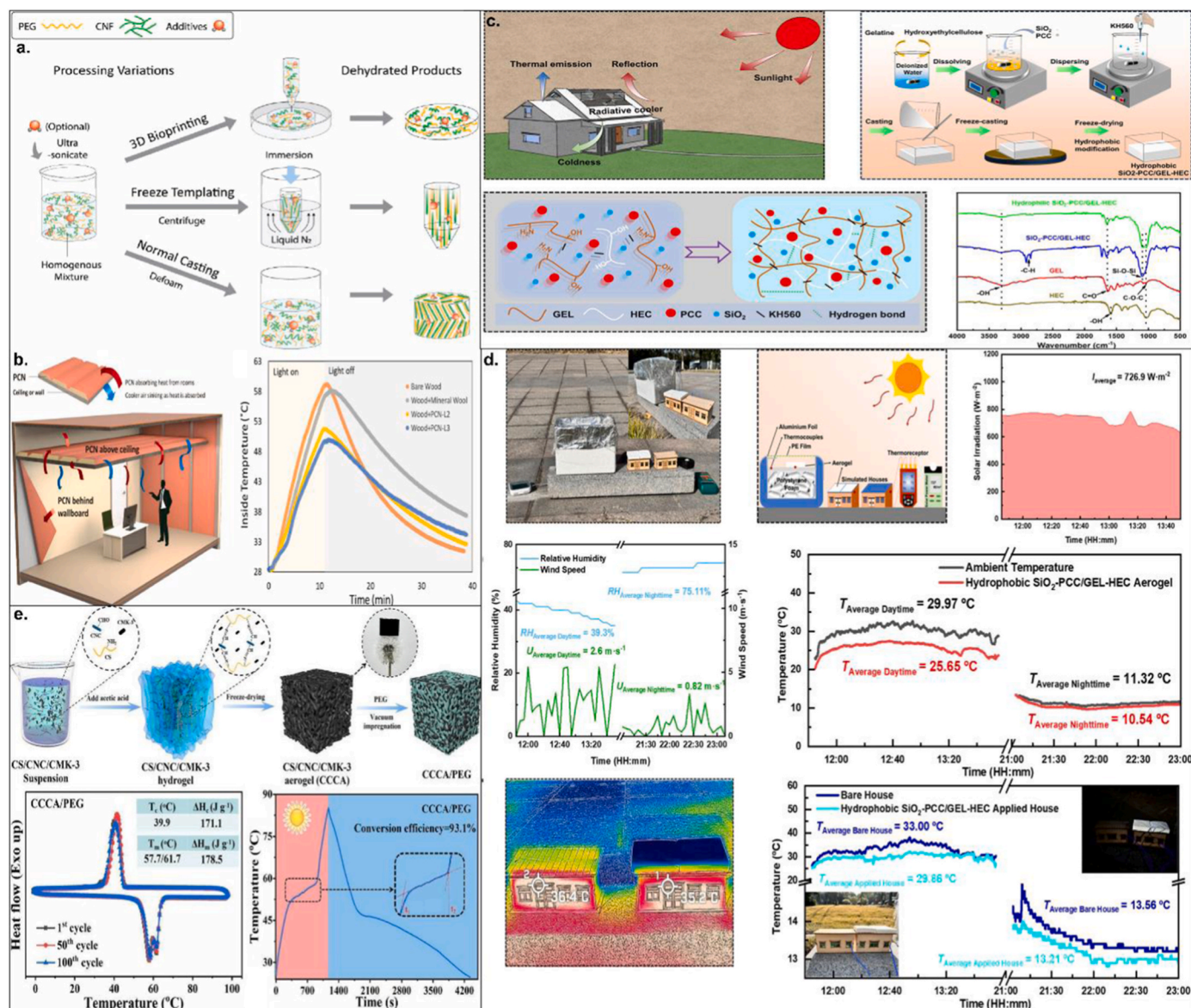


Fig. 15. (a.) Schematic illustrating changes in PCN processing: 3D Printing, Freeze drying, and oven drying. (b.) Evaluation of the thermal regulation capability of PCN under light irradiation [80]. Adapted from Ref. [80] with permission from Elsevier, copyright 2023 (c.) Schematic representation of a radiative cooler model utilized on a building's roof, along with a flowchart detailing the preparation method of the hydrophobic SiO₂-PCC/GEL-HEC composite aerogel. (d.) SiO₂-PCC/GEL-HEC composite aerogels for thermal regulation in buildings [210]. Adapted from Ref. [210] with permission from the American Chemical Society, copyright 2024 (e.) The preparation procedure of CA/PEG and their conversion of solar energy to thermal energy [211]. Adapted from Ref. [211] With permission from Elsevier, copyright 2024.

with a water contact angle of 152.3° , improving its efficacy in outdoor applications. Energy-saving calculations indicate that employing the hydrophobic SiO_2 -PCC/GEL-HEC composite aerogel as a roofing material can decrease energy consumption by 5.27% during summer relative to baseline structures. This composite aerogel provides a straightforward and efficient method for regulating thermal comfort in radiative cooling applications, as shown in Fig. 15d [210].

Cheng et al. created an innovative shape-stable PCM utilizing a chitosan/CNC/CMK-3 aerogel. This composite consists of chemically cross-linked chitosan and CNC, which provides a 3D-support structure, while CMK-3 enables solar-to-thermal energy conversion, and PEG functions as the latent heat storage element (Fig. 15e). The aerogel infused with PEG exhibits remarkable melting and crystallization enthalpy values of 178.5 and 171.1 J/g, respectively, and outstanding form stability, with a melting temperature around 67.8°C . The hierarchical porous structure efficiently incorporates PEG, enabling the composite to retain its form without leaking, even when subjected to compression above PEG's melting point. Moreover, the phase change aerogels have exceptional thermal stability, exhibiting an exceedingly low enthalpy degradation rate of $0.030 \pm 0.012\%$ per cycle after 100 thermal cycles. The integration of CMK-3 substantially improves the solar-to-thermal energy conversion efficiency, reaching an impressive rate of 93.1%. This study developed a high-performance, eco-friendly CA-PCM, emphasizing its use in solar energy conversion and storage [211].

Wang et al. designed a multifunctional CA-PCM with improved thermal management and photothermal performance by constructing a directionally frozen CNF foam scaffold embedded with lignosulfonate-functionalized BNNS (BNNS@LS). The scaffold was subsequently vacuum-impregnated with PEG to yield a shape-stable CA-PCM. The 3D porous structure formed through directional freezing successfully prevented PEG leakage during phase transitions while retaining a high latent heat of 177.2 J/g, equivalent to 93.56% of pristine PEG. The introduction of a BNNS@LS-CNF network notably enhanced thermal conductivity, achieving 1.00 W/m·K with only 1.8 wt% filler, a 2.1-fold increase compared to pure PEG. In addition to enhanced phase stability and thermal performance, the CA-PCM exhibited photothermal conversion capability, allowing it to store solar energy efficiently [112].

The various CA-PCM systems presented in this section demonstrate that developing functional additives are largely being developed to fulfill particular technical needs that cannot be met by conventional conductive fillers alone. Rather than increasing thermal conductivity, these solutions prioritize flame retardancy, radiative cooling, acoustic insulation, photothermal conversion, mechanical reinforcing, and environmental compatibility. They reveal the amazing adaptability of

cellulose aerogel platforms and point to future prospects for application-specific multifunctional thermal energy storage materials.

The aforementioned CA-PCM systems demonstrate distinct advantages arising from their composition and intended applications. Nonetheless, direct comparison of their performance is sometimes hindered by differences in PCM chemistry, filler loading, fabrication techniques, and testing methodologies. Nonetheless, various prominent engineering trends can be discerned in the literature, enabling a comparative assessment of CA-PCM systems with respect to thermal energy storage, heat transfer, structural integrity, multifunctionality, scalability, and application appropriateness. A comparative evaluation of these systems is shown in Table 5 and examined in Section 5.7.

5.7. Comparative analysis and design principles of CA-PCMs

Sections 5.1-5.6 focus on the different CA-PCM systems based on filler type and composition; however, a broader literature comparison reveals certain common engineering characteristics that may not be apparent when studies are examined independently. The investigations together indicate that the performance of CA-PCM represents a balance among thermal energy storage, heat transfer, structural stability, multifunctionality, and manufacturability, rather than identifying a single optimal material, since material selection is inherently application dependent. Consequently, the selection of materials should be governed by the performance requirements relevant to the application rather than by the optimization of a particular thermophysical property.

5.7.1. Thermophysical performance landscape

Across the literature summarized in Table 4, latent heat values of CA-PCMs generally range from ~ 108 to 263 J g^{-1} , whereas the incorporation of conductive fillers increases thermal conductivity from modest improvements to several-fold enhancement depending on filler chemistry, loading, and network architecture. These broad performance windows indicate that no single material simultaneously maximizes every performance parameter, reinforcing the need for application-oriented material optimization rather than universal material selection.

5.7.2. Thermal energy storage versus heat-transfer trade-off

A common observation in the literature is the inherent trade-off between thermal energy storage capacity and the enhancement of thermal conductivity. Generally, pure CA exhibit optimal PCM loading and latent heat retention due to the porous cellulose matrix occupying a minimal volume of the composite, hence offering adequate capillary confinement to avert leakage. Nonetheless, their intrinsically low thermal conductivity constrains the rate of heat absorption and release. The

Table 5

Comparative assessment of CA-PCM systems: performance trade-offs, engineering advantages, and application suitability.

CA-PCM family	Typical latent heat range (J/g)	Thermal energy storage performance	Heat transfer performance	Structural reliability	Scalability potential	Recommended application	Primary engineering limitation
Cellulose aerogels	145–165	Excellent	Limited	Excellent	Excellent	Building thermal regulation	Low intrinsic thermal conductivity
Cellulose/Graphene	147–187	Very Good	Excellent	Very Good	Moderate	Electronics, solar harvesting	Reduced latent heat at high filler loading
Cellulose/MXene	108–197	Very Good	Very Good	Excellent	Moderate	Solar thermal conversion, wearable heaters	Oxidation stability and cost
Cellulose/CNT	130–172	Very Good	Excellent	Good	Moderate	Wearables, battery thermal management	CNT dispersion
Cellulose/Polymer	133–263	Excellent	Moderate	Excellent	Very Good	Flexible thermal storage, smart textiles	Lower thermal conductivity
Miscellaneous hybrids	146–185	Good–Excellent	Moderate–Excellent	Good–Excellent	Moderate	Specialized multifunctional devices	Complex fabrication

Thermal energy storage performance is evaluated considering reported latent heat retention, PCM loading, and cycling stability across representative literature. Performance classifications are based on comparative analysis of representative literature summarized in Table 4 and should be interpreted qualitatively rather than as absolute rankings.

incorporation of thermally conductive fillers, including graphene, MXenes, CNTs, boron nitride, metallic nanowires, and conductive polymers, establishes continuous heat-transfer pathways and considerably improves thermal sensitivity. Nevertheless, the increased quantity of these inactive fillers inevitably reduces the effective PCM percentage, leading to diminished latent heat storage despite improved thermal conductivity. Consequently, the highest thermal conductivity does not necessarily equate to optimal overall thermal energy storage efficacy.

5.7.3. Influence of hierarchical pore architecture

A notable trend is the function of CA architecture. The hierarchical porosity structure of CA influences PCM loading, leakage resistance, crystallization behavior, and long-term cycling stability, independent of the filler chemistry. The extensively interconnected pore networks can generate sufficient capillary force to effectively immobilize the molten phase change material (PCM) and maintain a significant PCM loading, while excessive confinement in exceedingly small pores may impede molecular crystallization and reduce latent heat. Hierarchical pore designs comprising macro-, meso-, and micropores appear to provide the optimal balance between energy storage density, dimensional stability, and thermal reliability.

5.7.4. Processing-performance relationships

The manufacturing method also influences the final performance of CA-PCMs. Vacuum impregnation often yields the highest PCM loading and latent heat, as the porous scaffold is established prior to PCM infiltration. Conversely, in-situ polymerization facilitates a more uniform dispersion of phase change materials and enhances interfacial contact, resulting in improved structural durability after repeated thermal cycles. Encapsulation offers remarkable leakage suppression and durability; however, it generally results in a loss of latent heat from inactive shell materials. Direct mixing is the most straightforward and scalable manufacturing procedure; yet, achieving homogeneous dispersion and long-term phase stability is challenging. Consequently, fabrication methods must be selected according to the intended use, rather than only on latent heat or PCM loading.

5.7.5. Engineering roles of conductive fillers

The comparison with several hybrid reinforcing methods indicates that no singular conductive filler optimizes all performance measures concurrently. Graphene-based devices exhibit superior thermal conductivity and effective photothermal conversion owing to their densely interconnected conductive networks, rendering them particularly suitable for electronic cooling and solar thermal harvesting. Although MXene-containing aerogels exhibit superior thermal conductivity and exceptional photothermal and electrothermal conversion efficiencies, their long-term oxidation stability and comparatively high material cost pose challenges for widespread application. CNT-based devices exhibit pronounced flexibility and rapid heat transfer, making them appealing for wearable thermal management. Polymer-modified systems often exhibit enhanced mechanical robustness, flexibility, and cyclic stability, albeit with inferior thermal conductivity. Hybrid systems including diverse conductive fillers exhibit synergistic enhancements in multifunctionality, but accompanied by heightened processing complexity and manufacturing costs.

5.7.6. Variability across reported studies and the need for standardization

Nonetheless, despite these general trends, certain oddities persist in the documented literature, underscoring the necessity for more stringent comparison analysis. While the incorporation of conductive fillers often enhances thermal conductivity, the degree of improvement significantly fluctuates even with equivalent filler quantities, owing to differences in filler dispersion, orientation, pore architecture, interfacial thermal resistance, and fabrication techniques. Likewise, the retention of latent heat does not consistently diminish with higher filler loading, indicating that the optimized hierarchical networks of cellulose can partially offset

the reduction in PCM content by enhancing confinement efficiency and crystallization behavior. The documented thermal dependability has a broad spectrum, with cycling stability ranging from about 100 to over 1000 thermal cycles, mostly attributable to differing testing methodology, heating-cooling rates, and performance evaluation criteria employed in various studies. The variations hinder direct comparisons among published research and emphasize the need for standardized testing protocols and reporting criteria to facilitate more reliable benchmarking of future CA-PCM systems.

5.7.7. Application-oriented materials selection

From an engineering perspective, CA-PCM design should be dictated by application requirements rather than specific material performance. High latent heat storage, dimensional stability, and long-term cycling endurance are essential for thermal regulation, whereas rapid heat dissipation and latent heat storage are necessary for battery thermal management. Flexibility, low weight, and mechanical endurance are essential for wearable thermal management, while efficient photothermal materials like as graphene or MXene are preferred for solar thermal energy conversion. Consequently, the future advancement of CA-PCM is expected to transition from enhancing individual characteristics to application-specific optimization grounded in balanced thermophysical performance.

5.7.8. Future design principles

The existing research suggests that the future development of CA-PCMs will be progressively influenced by the incorporation of rational pore-structure engineering, multifunctional hybrid fillers, scalable eco-friendly manufacturing, and computationally aided materials design. Establishing measurable structure-property-performance relationships across diverse design parameters would enable the transition of CA-PCMs from laboratory demonstrations to commercial thermal energy storage applications.

The aforementioned engineering trends lay the foundation for computationally assisted CA-PCM design, in which multiscale modeling and data-driven optimization can predict the effects of pore architecture, filler distribution, and interfacial interactions on thermal performance. Consequently, the subsequent section addresses the growing importance of computational methodologies in accelerating the rational design of next-generation CA-PCMs.

6. Computational approaches for design of CA-PCMs

As discussed in Sections 4 and 5, significant progress has been achieved in developing high-performance cellulose aerogel-based phase change materials (CA-PCMs) through the incorporation of functional fillers and optimization of fabrication strategies. However, the development of efficient CA-PCMs still relies largely on trial-and-error experimentation to identify suitable cellulose matrices, PCM compositions, filler loadings, and hierarchical pore architectures. Such an approach is time-consuming, resource-intensive, and often requires extensive experimental validation [14]. Consequently, computational approaches have emerged as powerful complementary tools for optimizing CA-PCMs. These approaches enable the prediction of molecular interactions, interfacial compatibility, heat-transfer behavior, phase transition characteristics, and structural stability across multiple length scales prior to material synthesis. By complementing experimental investigations, computational methods reduce the number of experimental iterations, accelerate material discovery, improve prediction accuracy, and facilitate the development of high-performance CA-PCMs with tailored thermal energy storage properties [212]. Among the available computational techniques, molecular dynamics (MD) [213] and density functional theory (DFT) [214] provide atomistic insights into cellulose-PCM interactions, finite element modeling (FEM) [215] predicts heat transfer, phase transition behavior, and thermomechanical performance at the device scale, while machine learning (ML) enables

data-driven optimization of material compositions and processing parameters. When integrated with experimental validation, these complementary approaches establish a computational–experimental framework that can significantly accelerate the development of next-generation CA-PCMs for thermal energy storage.

Together, molecular dynamics (MD), density functional theory (DFT), finite element modeling (FEM), and machine learning (ML) constitute a multiscale materials-by-design framework for CA-PCMs. Rather than functioning as independent computational techniques, they provide complementary information across different length scales, enabling predictive optimization of material composition, pore architecture, interfacial interactions, and device-level thermal performance prior to experimental validation.

Despite the growing importance of computational materials science, its application to CA-PCMs remains in its early stages. Existing computational studies have primarily focused on understanding intermolecular interactions, optimizing filler dispersion and hierarchical pore architectures, predicting thermal transport and phase transition behavior, and evaluating material performance under realistic operating conditions [75]. At the atomistic level, DFT and MD simulations provide complementary insights into cellulose–PCM interactions. DFT is primarily employed to investigate binding energies, adsorption behavior, and interfacial interactions, whereas MD simulations elucidate hydrogen-bonding networks, PCM diffusion, and molecular mobility, thereby guiding the selection of suitable cellulose matrices, phase change materials, and functional additives [213,214]. At the continuum scale, FEM enables the prediction of heat transfer, melting–solidification

behavior, temperature distribution, and thermomechanical responses of CA-PCMs under practical operating conditions, facilitating the design and optimization of thermal energy storage systems [216]. Furthermore, ML algorithms are increasingly being employed to establish structure–property relationships, optimize material compositions and processing parameters, and accelerate the discovery of high-performance CA-PCMs using experimental datasets [217]. Collectively, these computational approaches reduce reliance on empirical trial-and-error experimentation while providing a scientific basis for the rational design and optimization of advanced CA-PCMs. Representative computational studies illustrating these capabilities and their experimental validation are discussed below. For clarity, the computational approaches discussed in this section are presented according to their complementary roles in the CA-PCM design process, spanning atomistic-scale simulations (DFT and MD), continuum-scale modeling (FEM), and data-driven optimization (ML), thereby establishing a multiscale computational framework for rational materials design.

Machine learning (ML) has emerged as a powerful data-driven tool for analyzing complex datasets and optimizing experimental parameters. In CA-PCMs, ML can predict the relationship between PCM content and the resulting thermal and mechanical properties, thereby reducing the need for extensive trial-and-error experimentation (Fig. 16a) [98]. In particular, the authors used a Gaussian predictor to perform Bayesian optimization in estimating nonlinear material parameters such as viscoelastic transition temperature (T_t) and Young's modulus (E). The results suggested a correlation between PEG content in the precursors and PEG's influence on the rheological properties of the precursors and

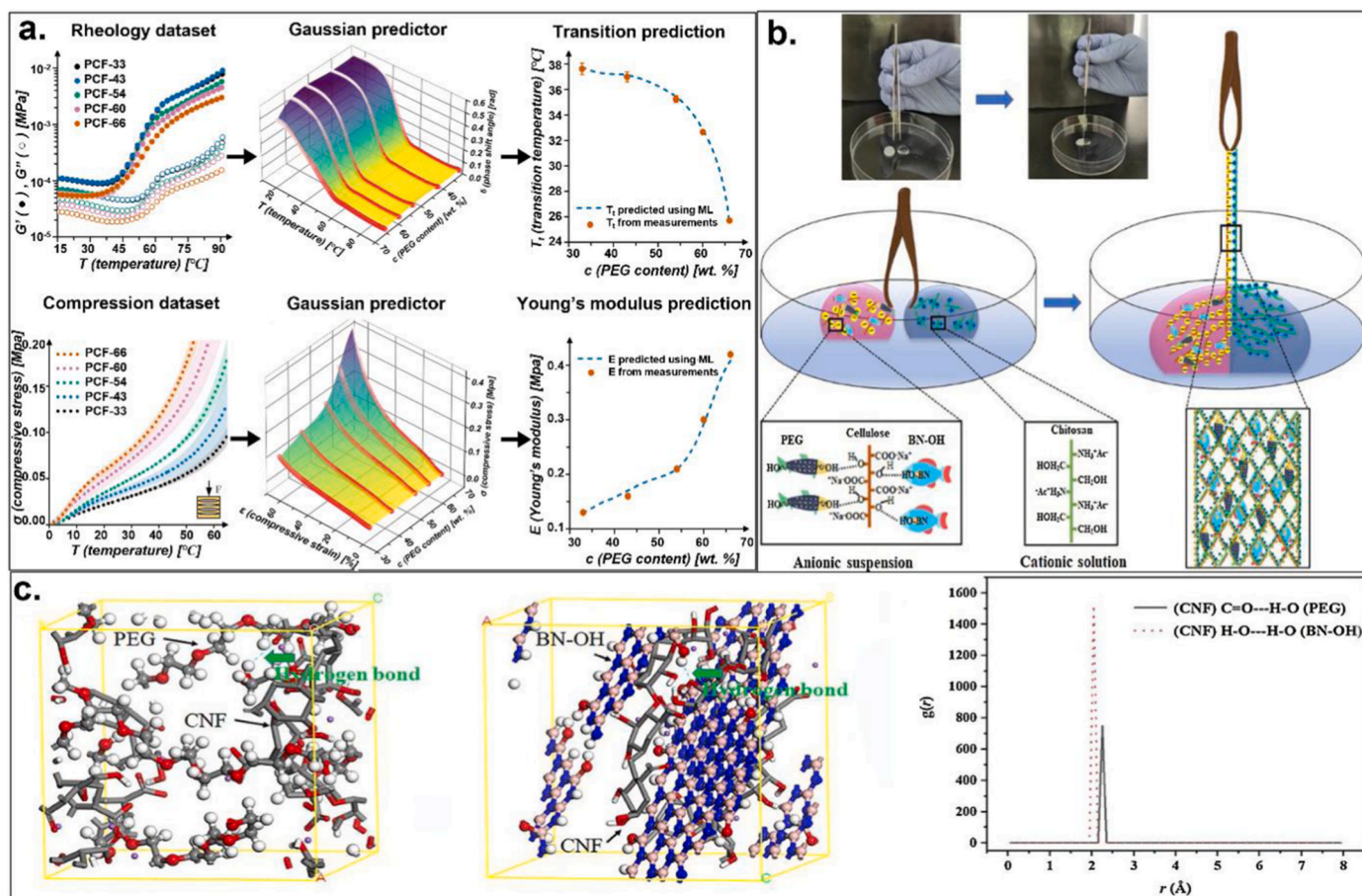


Fig. 16. (a.) Machine learning framework to forecast the viscoelastic gel transition of methylcellulose based on polyethylene glycol content and to estimate Young's modulus of phase change cellulose foams [98]. Adapted from Ref. [98] with permission from Elsevier, copyright 2023 (b.) Process of fabricating PEG/BN/CNF/CS macrofibers using interfacial polyelectrolyte complex spinning. (c.) Molecular dynamics simulation of intermolecular hydrogen bonding interactions between CNF and polyethylene glycol [18]. Adapted from Ref. [18] with permission from Elsevier, copyright 2020.

the final mechanical and thermal properties of the processed cellulose foams. These predictions helped the authors to select the best low-viscosity PEG concentrations that would result in substantial and thermally stable blends. Moreover, ML helped to speed up CA-PCM formation, allowing the characterization of non-linear behaviours in

the experiments, like the effect of PEG on the viscoelastic properties, and determining the latent heat and temperature changes of phase transition. Overall, the study illustrates how computational devices can assist in realizing the research goals by reducing time, improving accuracy, and speeding up the engineering of new eco-composite materials with

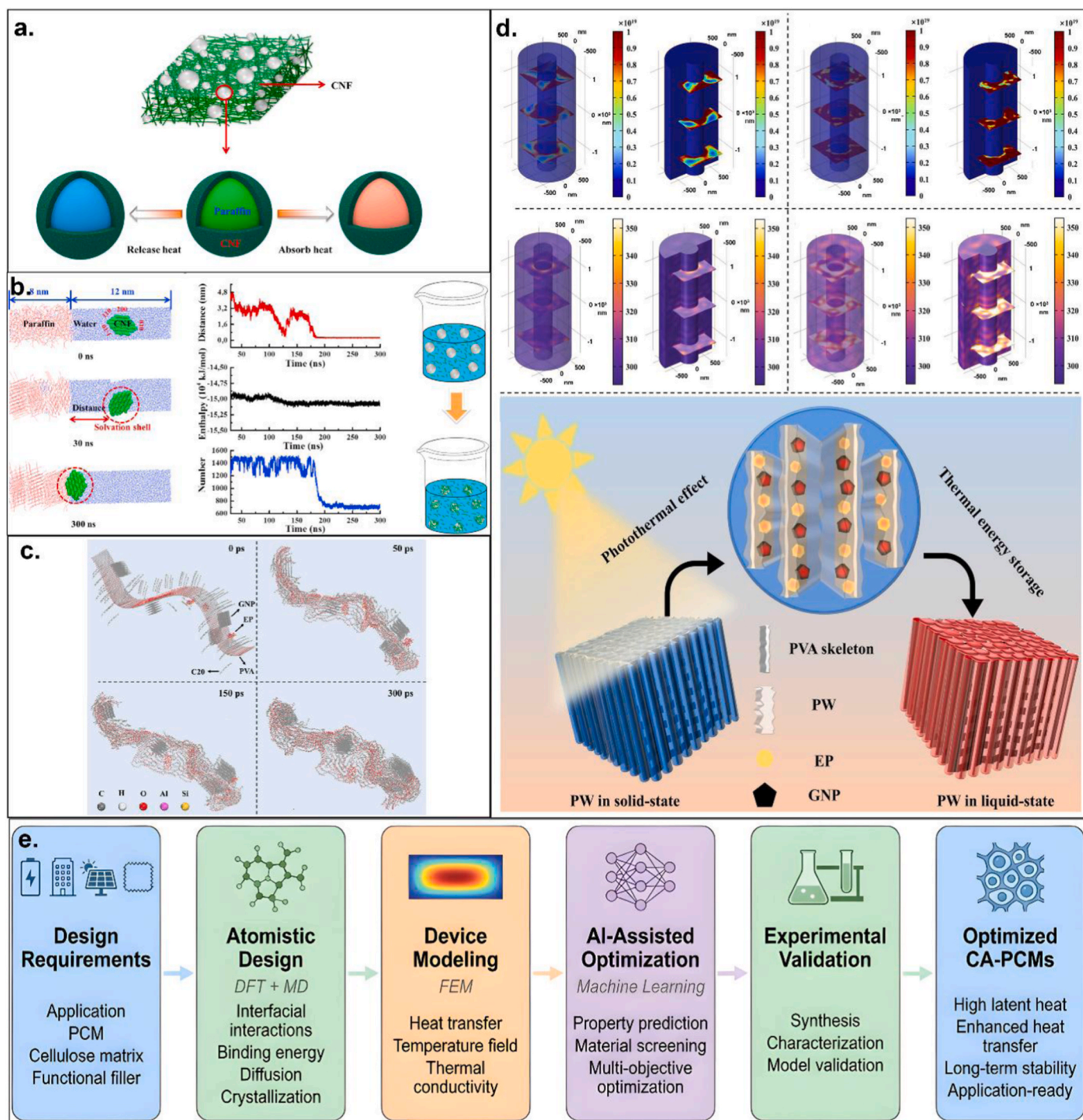


Fig. 17. (a.) Representation of the three-dimensional structure of form-stable thermal control biocomposites, including paraffin-core and CNF-shell capsules. (b.) Molecular modeling and diagram depicting the production of paraffin capsules [55]. Adapted from Ref. [55] with permission from Elsevier, copyright 2017 (c.) Results of molecular dynamics simulations regarding the interactions between PEPG and C20 molecules over time. (d.) Theoretical simulation study and electric field distribution utilizing radio frequency interface in COMSOL Multiphysics program [219]. Adapted from Ref. [219] with permission from the American Chemical Society, copyright 2023. (e.) Integrated materials-by-design framework illustrating how computational approaches support the rational development of cellulose aerogel-based phase change materials (CA-PCMs). Density functional theory (DFT) and molecular dynamics (MD) provide atomistic insights into interfacial interactions, diffusion, and crystallization, finite element modeling (FEM) predicts device-scale thermal behavior, and machine learning accelerates property prediction and multi-objective optimization. Experimental validation verifies computational predictions and enables the development of high-performance CA-PCMs for advanced thermal energy storage applications.

specific properties.

Although ML applications in CA-PCMs are still limited, successful implementation in other PCM systems further demonstrates their potential. For example, bhatasana et al. employed neural-network surrogate models integrated with genetic algorithms to optimize PCM design parameters and thermophysical properties, reducing the maximum temperature rise by 19% and temperature fluctuations by up to 93% while significantly reducing computational cost [218]. These findings suggest that similar ML-assisted optimization strategies could be extended to optimize the composition of cellulose aerogels, PCM loading, filler content, and hierarchical pore architectures, thereby advancing experimental development of next-generation CA-PCMs.

MD simulations have been widely employed to elucidate intermolecular interactions governing the stability and thermal performance of CA-PCMs. In contrast, density functional theory (DFT) is still rarely applied to CA-PCMs. Nevertheless, DFT remains a valuable complementary tool for quantifying adsorption energies, charge distribution, and interfacial interactions between cellulose, PCM molecules, and functional nanofillers, thereby providing atomistic guidance for selecting compatible material combinations prior to experimental synthesis. A representative MD study investigated the intermolecular interactions among CNF, hydroxylated boron nitride (BN-OH), and PEG using hydrogen-bond analysis and radial distribution function (RDF) calculations (Fig. 16b and c) [18]. The simulations demonstrated that strong hydrogen-bonding interactions between PEG, CNF, and BN-OH restricted the mobility of PEG chains, thereby improving structural stability during repeated melting–solidification cycles. RDF analysis further confirmed these intermolecular interactions and revealed homogeneous dispersion of BN-OH within the cellulose matrix, promoting the formation of continuous heat-transfer pathways. These molecular-level interactions enhanced thermal conductivity, shape stability, and leakage resistance of the composite PCM. Importantly, the computational predictions were validated experimentally, demonstrating a 22.56-fold enhancement in thermal conductivity together with excellent form stability, highlighting the capability of MD simulations to guide the rational selection of thermally conductive fillers and optimize interfacial interactions in CA-PCMs.

MD simulations were also used to understand paraffin encapsulation in CNF, demonstrating that CNF-shell modeled capsules take advantage of the amphiphilic character of the CNFs, which allows them to make stable contact at the oil-water interface, as shown in Fig. 17a [55]. The simulations in Fig. 17b showed that a system enthalpy reduction and an increase in solvent entropy induce the bulked water CNFs to the interphase through diffusion. Such a dynamic stabilization of the encapsulated material, in essence, guarantees robust encapsulation while at the same time enabling robust thermal regulation as the paraffin droplets contained in the capsule are surrounded by tightly packed CNFs, preventing them from leaking. The computational analysis identified minimal thermodynamic parameters governing capsule formation, including CNF interaction energies and entropy contributions of CNF. The MD simulations predicted the interface's stability, which is necessary to understand heat's effect on the capsules' structure properly, and were confirmed by experiment.

In another study, simulations with COMSOL, Multiphysics, and MD were used to help understand why improvements in photothermal conversion and storage of CA-PCMs were observed (Fig. 17c and d) [219]. COMSOL simulations showed that adding paraffin wax (PW) into the composite system locked sunlight to the top surfaces of graphite nanoplates, thus increasing their ability to absorb and convert sunlight to heat through lattice oscillations. This increases resistance losses in the graphite backbone and processes favoring heat generation and storage in the molten PW. The MD simulations also highlighted the stabilizing aspects within the composite matrix by recognizing van der Waals and Lewis acid-base interactions as fundamental to the stability and performance of PW and the aerogel scaffold. Measurements validated the computational conclusions with 95.2% photothermal conversion

efficiency under sunlight and remarkable CA matrix stability across 2000 thermal cycles. This study demonstrates how multiscale computational approaches (MD and COMSOL Multiphysics) can bridge molecular interactions with macroscopic thermal performance, thereby providing valuable guidance for optimizing high-performance photothermal CA-PCMs.

At the mesoscale and continuum scales, finite element analysis (FEA) has emerged as an effective computational tool for predicting the macroscopic thermal behavior of CA-PCMs, including temperature distribution, melting–solidification behavior, and thermal management performance. For example, Shang et al. employed finite element modeling to evaluate the chip-level thermal management performance of a biomass carbon-reinforced PEG/PVA/CNF aerogel. At full load of 1.8 W, the bare chip reached the critical temperature of 85 °C within 200 s. In contrast, the chip coated with PVA/CNF and P50C5/700 reached only 72.7 °C and 68.4 °C, respectively, at the same time. The time required to reach 85 °C increased from 200 s for the bare chip to 325 s for PVA/CNF and 370 s for P50C5/700, corresponding to approximately an 85% increase in safe operating time. These FEM results demonstrate that the CA-PCM delayed overheating by combining latent heat absorption with improved heat conduction through the biomass carbon network [220]. More broadly, a 2026 review of FEA of composites for latent heat storage highlights the increasing use of numerical modeling to resolve temperature fields, phase transition behavior, and geometry-dependent thermal performance in PCM-based systems [216]. Moreover, mixed-dimensional finite element modeling has been recently shown to lower the computational cost while maintaining good predicted accuracy, with the results confirmed against experiments for latent heat storage-related systems. These advances are particularly pertinent to CA-PCM systems, where the porous scaffold design, PCM confinement and transient temperature response are strongly related.

Recent studies demonstrate that computational tools such as MD simulations and finite element modeling can provide mechanistic hydrogen bonding, interfacial stabilization, heat transfer, and photothermal behavior in CA-PCMs. Experimental results verify improvements in PCM confinement, thermal conductivity, conversion efficiency, and cycling stability. However, the present computational investigations are limited to isolated case studies, with a lack of cross-verification between different cellulose matrices, PCM systems and filler types, and the model assumptions and parameter sensitivity are generally not critically examined. Future research should transition from isolated computational studies toward integrated multiscale design frameworks that combine DFT, MD, FEM, and ML with systematic experimental validation, uncertainty quantification, and application-oriented performance evaluation. As summarized in the proposed materials-by-design framework (Fig. 17e), DFT, MD, FEM, and ML operate as complementary computational tools across multiple length scales, collectively enabling predictive optimization of CA-PCMs prior to experimental validation. This multiscale computational–experimental strategy establishes a conceptual roadmap for integrating computational predictions with experimental validation, thereby reducing trial-and-error experimentation and accelerating the rational design of next-generation CA-PCMs.

7. Challenges and future aspects of CA-PCMs

Despite the impressive developments in CA-PCMs, their translation from laboratory-scale demonstrations to commercially viable thermal energy storage solutions is restricted by multiple interconnected scientific, engineering and economic hurdles. While recent literature has mostly investigated the enhancement of thermophysical properties using sophisticated fillers and hierarchical pore engineering, the practical application of aerogels has become more reliant on manufacturability, long-term durability, sustainability, and cost-effectiveness. Unlike many conventional stabilizing materials, cellulose itself is

abundant, renewable, biodegradable, and very cheap. Therefore, the key constraints to commercialization are not the availability of raw materials but rather the processing technology needed to manufacture highly porous aerogel structures with structural integrity and multifunctional performance. Hence, future research should not only focus on material optimization, but also take an integrated strategy, considering performance, manufacturability, sustainability and economic feasibility simultaneously. [15,96,221]. Fig. 18 summarizes the properties, applications, challenges, and prospects of CA-PCMs. Among the various future research directions, two themes are expected to play particularly important roles in accelerating industrial adoption of CA-PCMs. The first is the development of green, scalable manufacturing strategies capable of reducing production cost and environmental impact without compromising material performance. The second is the integration of multifunctionality, whereby thermal energy storage is combined with self-healing, sensing, electromagnetic shielding, or other intelligent functionalities to expand the application space of CA-PCMs beyond conventional latent heat storage. These two directions collectively provide the foundation for the next stage of CA-PCM development and are discussed in the following subsections.

7.1. Green preparation processes and sustainable manufacturing

From a commercialization perspective, the environmental and economic impacts of CA-PCMs are largely determined by the manufacturing method rather than the cellulose precursor per se. Cellulose is renewable, biodegradable and abundantly available yet the synthesis of high-performance aerogels frequently involves energy-intensive drying, multiple solvent-exchange stages and the addition of expensive functional nanomaterials. Thus, future manufacturing strategies should aim to reduce environmental impact, simplify processing, increase production throughput, and reduce manufacturing cost while retaining the

hierarchical porous architecture responsible for high PCM loading and superior thermal performance.

From an industrial perspective, the various PCM stabilization systems currently exhibit differing degrees of technological advancement. Owing to advanced production techniques, traditional microencapsulation, expanded graphite composites, and mineral-supported phase change materials (PCMs) are currently utilized in many heat management applications at commercial or pilot scales. Nonetheless, the majority of CA-PCM systems remain in laboratory or first pilot phases, mostly due to challenges associated with continuous aerogel synthesis, drying methods, and reproducible large-scale processing. Consequently, subsequent endeavors should concentrate on improving material performance and the technological preparedness of CA-PCMs via scalable production techniques, standardized quality control measures, and pilot-scale demonstrations to confirm long-term operating reliability in realistic situations.

The conventional preparation of CA-PCMs has depended on energy-intensive drying techniques such as freeze-drying and supercritical drying. A more pragmatic approach would be to promote processing pathways closer to industrial adoption, while simultaneously minimizing environmental load. In this respect, ambient pressure drying (APD) is a promising short-term alternative, provided that solvent exchange and surface modification are properly done to avoid pore collapse. Ambient temperature drying (ATD) might also be applicable for some CA-PCM systems that involve hydrophobic alteration or in situ PCM integration to maintain structural integrity.

Several developing manufacturing technologies, along with drying at ambient pressure, can fill the gap between lab-scale fabrication and industrial production. Spray drying provides a continuous process with much higher production throughput compared to conventional freeze-drying, but it is still problematic to preserve the highly interconnected porous architecture required for high PCM loading. Microwave-assisted

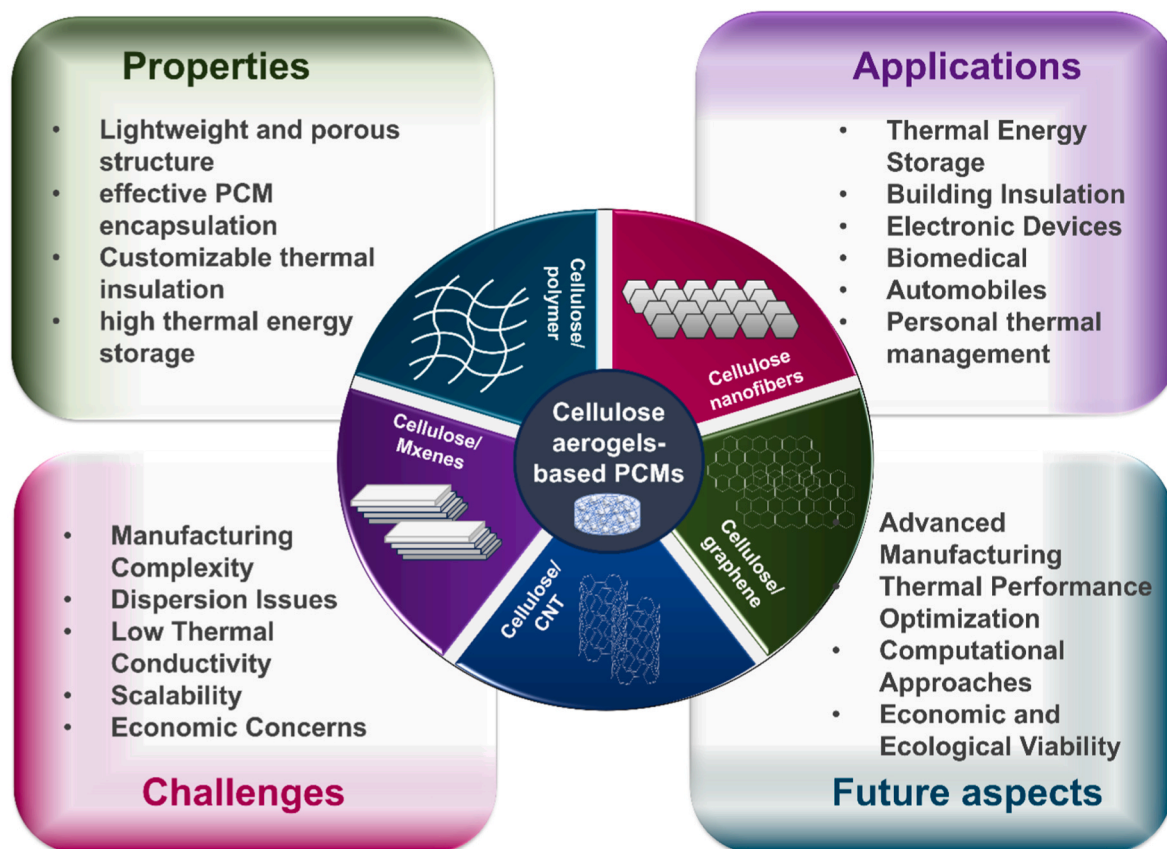


Fig. 18. Summary of CA-PCMs: properties, applications, challenges, and future aspects.

drying can cut drying time and energy consumption considerably by improving the internal heat transmission. However, an optimization of microwave power and solvent removal is required to avoid structural collapse of the aerogel network. Therefore, the drying technology should be selected based on the processing speed and cost, but also on the preservation of the pore morphology, the retention of the PCM, and the long-term thermal reliability, which ultimately define the practical performance of CA-PCMs. There are many process-level modifications that can make products more environmentally benign besides drying methods: (1) In-situ PCM loading during aerogel synthesis either through direct mixing or in-situ polymerization. This eliminates the need for a separate impregnation step, thereby reducing processing time and material losses by 20 to 30%; (2) Solvent recovery and recycling systems can reclaim around 90% of the organic solvents employed in gel preparation and solvent exchange, leading to a reduction in waste and material costs, and (3) A closed-loop manufacturing processes that incorporate water and solvent recycling can decrease freshwater consumption by 80 to 90% and organic solvent emissions by ~95%. (4) Implication of bio-based precursors, such as bacterial cellulose from agricultural or industrial waste streams, as well as bio-derived cross-linkers, can be used instead of petroleum-based materials. These changes will make it easier to make raw materials with less carbon footprint.

Consequently, future computational studies should use life cycle assessment (LCA) and process optimization algorithms to identify the best mix of solvent systems, drying technologies, and preparation techniques that minimize environmental impact while meeting performance requirements of CA-PCMs systems. Machine learning models that use experimental data can quickly look through the huge data and parameters for greener preparation pathways. They can assist in adopting smart processes by predicting the ideal circumstances for various CA-PCM compositions. Ultimately, successful commercialization of CA-PCMs will require simultaneous optimization of material performance, manufacturing scalability, environmental sustainability, and technology readiness rather than improvements in thermophysical properties alone.

7.1.1. Integration of multifunctionality in the CA-PCMs

The integration of multifunctionality into CA-PCMs should be regarded as an evolutionary engineering strategy, rather than the concurrent incorporation of many distinct functions. From an industrial perspective, additional features should be integrated solely when they yield quantifiable benefits without substantially compromising thermal energy storage capacity, mechanical reliability, or manufacturability. Multifunctional development can be categorized into three distinct practical issues: (i) functions aimed at directly enhancing performance to improve thermal storage capabilities (e.g., enhancing thermal conductivity, photothermal conversion), (ii) functions focused on reliability enhancement to increase durability and operational safety (e.g., self-healing, leakage detection), and (iii) functions designed for intelligence realization to facilitate autonomous monitoring, electromagnetic shielding, and adaptive thermal management. Conversely, this hierarchical development approach provides a more pragmatic pathway for transitioning multifunctional CA-PCMs from laboratory demonstrations to actual engineering applications.

While CA-PCMs have been developed primarily for thermal energy storage, their integration with additional functionalities can open substantially new application domains. This can ultimately result in dramatic increase market value and societal impact of CA-PCMs. Strategic incorporation of self-healing, sensing, and electromagnetic (EM) shielding characteristics in CA-PCMs represents a transformative research direction.

Self-healing is of particular significance since structural defects, such as cracks, micro-delamination or rupture of the aerogel matrix, can compromise mechanical integrity and lead to PCM leakage. The integration of self-healing mechanisms is able to self-repair damages during the use of CA-PCMs, leading to an increase in the service life and

reliability of the material. Embedding reversible cross-links into the cellulose network permits self-healing via dynamic bond breakage and reformation, enables at increased temperatures during regular phase change cycling. Also, embedding liquid precursors in microcapsules within the aerogel can induce healing when microcapsules burst upon damage, releasing reactive components that polymerize and fill cracks of CA-PCMs. Alternatively, one can include ion-containing polymers to allow for electrostatic self-assembly of the fractured surfaces, leading to quick recovery of mechanical and thermal properties. Self-healing integration is of particular importance in aerospace and automotive applications where maintenance access is limited or expensive. Research can be focused on such procedures that do not compromise the thermal cycle stability, do not impair the latent heat capacity, and can operate in the range of realistic temperatures.

The integrated sensing capabilities enable CA-PCMs to switch from passive thermal energy storage materials to smart systems for thermal management, which are capable of performing continuous monitoring of structural integrity and operational conditions. Embedded sensing technologies should not be considered as stand-alone sensing devices but rather as a tool to deliver engineering information that will immediately improve the thermal energy storage system dependability, safety and maintenance efficiency. From a practical point of view, sensing functions are categorized into four classes in terms of the information they provide: (i) temperature monitoring, (ii) structural health monitoring, (iii) leakage detection and (iv) environmental monitoring. Thermal monitoring can be conducted using thermochromic dyes, resistance temperature detectors or thermistors to map the temperature distribution during charge and discharge cycles. Piezoresistive materials are used for structural health monitoring to detect the presence of stress accumulation, deformation or microcrack formation during repeated temperature cycling. The PCM migration due to structural degradation can be detected by leakage detection based on the use of conductive networks inserted in the cellulose matrix. Any unusual change of electric resistance can indicate the PCM migration. Environmental monitoring, such as humidity sensing, gives more information about external circumstances that may accelerate material degradation or degrade thermal management performance. Collectively, these intelligent sensing capabilities are anticipated to enable predictive maintenance and autonomous operation of future smart buildings, wearable electronics, battery thermal management systems and aerospace thermal protection structures.

The major challenges in the sensor integration to CA-PCMs system includes preventing sensor degradation during thermal cycling, maintaining sensor functionality across wide temperature ranges, and avoiding heat-leak pathways created by sensor leads or housings. Future research can focus on prioritizing minimally invasive, distributed sensor arrays embedded within the bulk aerogel matrix rather than external sensors.

Combining electromagnetic interference (EMI) shielding and thermal energy storage is an emerging area for multifunctional thermal management materials for next-generation electronic devices. As electronic devices become smaller and power density increases, thermal management and electromagnetic protection need to be considered simultaneously to ensure operational stability, signal integrity and device dependability. Therefore, CA-PCMs with dual functions of latent heat storage and EMI shielding offer great prospects for aerospace electronics, military systems, wearable gadgets, medical instruments, and advanced communication technologies. For CA-PCMs, integration of EM-shielding functionality can occur through conductive filler networks like graphene, carbon nanotubes, or metallic nanoparticles creates percolating conductive pathways that absorb and dissipate incident EM energy. The conductivity must be sufficient for EMI shielding (typically $\sigma > 10^{-2}$ S/cm) while not so high as to create excessive thermal conductivity that reduces insulation capability. Magnetic nanoparticles can also be integrated to CA-PCM network to provides magnetic loss absorption which reduces transmitted EM radiation.

Moreover, hybrid approaches like combining conductive and magnetic phases can enable multi-mechanism EM attenuation with broader frequency range.

7.1.2. Implementation roadmap

A defined roadmap is required to achieve these advanced research areas.

Short-term (1–3 years): Focus on improving certain green preparation techniques and verifying proof-of-concept for individual multifunctions (self-healing OR sensing OR shielding). Create Life Cycle Assessment frameworks to evaluate the environmental benefits of different drying techniques. **Intermediate period (3–5 years):** Create coherent production procedures that combine green drying with concurrent PCM loading and the application of beneficial fillers. Display systems having dual functionality (for example, thermal storage combined with self-healing or thermal storage merged with sensing). **Extended duration (5–10 years):** obtain fully integrated, multifunctional CA-PCMs that use green manufacturing methods to store thermal energy, fix themselves, have sensors built in, and block electromagnetic interference (EMI). Build pilot-scale factories to show that the technologies can be used in real life in aerospace, building, and wearables and that they can be made to make money.

In the future, researchers may focus on multiscale models, predictive analytics, and the real-time improvement of material effectiveness. To sum up, the progress of CA-PCMs relies on overcoming the technical, economic, and environmental challenges through research and cross-disciplinary innovation. As renewable energy technologies like solar thermal energy storage improve, they can make it easier to use CA-PCMs in sustainable energy settings by addressing the issues we discussed in this part.

8. Concluding remarks

CA-PCMs have evolved from basic shape-stabilized thermal storage substances to multifunctional platforms for sustainable thermal regulation. This review exceeds a mere synopsis of recent advancements and establishes a materials-by-design framework that links the manufacturing method, pore architecture, interfacial interactions, and hybrid filler selection to the thermophysical performance of CA-PCMs. The comparison reveals that no single material system optimizes latent heat storage, thermal conductivity, mechanical robustness, multifunctionality, and scalability, emphasizing that material selection should be guided by application-specific performance criteria rather than the optimization of a singular property.

The research indicates that future progress relies more on the rational engineering of structure-property-performance relationships through optimal fabrication, hierarchical pore design, multifunctional integration, and scalable manufacturing, rather than on new fillers. Despite significant advancements, widespread implementation is constrained by challenges related to long-term durability, commercialization, sustainable processing, and uniform performance assessment. Future development should integrate experimental research, computational materials design, and application-focused optimization to expedite the practical implementation of next-generation CA-PCMs for improved thermal energy storage applications.

Author contributions

SB: Conceptualization; Data curation; Formal analysis; Roles/Writing - original draft; and Writing - review & editing, **SS:** Conceptualization; Data curation; Formal analysis; Roles/Writing - original draft; and Writing - review & editing, **PD:** Visualization; Roles/Writing - original draft; and Writing - review & editing, **IIS:** Writing & editing; Supervision; Validation; Visualization, **PKM:** Resources; Software; Supervision; Validation; Visualization; Roles/Writing - original draft; and Writing - review & editing.

Declaration of Competing interests

The authors declare no conflict of interest.

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Data availability

Data will be made available on request.

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