

Recent Advances in Enhanced Thermal Property in Phase Change

Materials using Carbon nanotubes: A review

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Abstract

As the demand for renewable and sustainable energy sources rises, considerable efforts have been dedicated to the development of energy storage materials. Phase change materials (PCMs) have garnered significant attention for their potential applications in thermal energy storage (TES) and management systems. However, the low thermal conductivity and potential leakage during phase transition hinder the widespread application of these materials. The integration of carbon nanotubes (CNTs) into PCMs has emerged as a promising strategy to enhance their thermal properties due to their exceptional thermal conductivity and structural characteristics. Herein, we provide an overview of recent advances in the utilization of different types of CNTs to improve the thermal performance of PCMs. We discuss various methods of incorporating CNTs into PCMs, including physical mixing, chemical functionalization, and hybrid nanostructures. Furthermore, we examine the effects of CNT addition on the thermal conductivity, latent heat storage capacity, light-to-thermal conversion efficiency, and thermal stability of PCMs. In addition, we discuss the current challenges and prospects for the practical implementation of CNT-enhanced PCMs in diverse TES applications.

Keywords: Phase change material, carbon nanotubes, thermal conductivity, nanocomposites, thermal energy storage

1. Introduction

In recent years, as the economy has progressed, there has been a consistent rise in energy demand (Figure 1)[1] and nonrenewable resource consumption [2, 3]. Additionally, the use of fossil fuels has led to a surge in greenhouse gas emissions, significantly impacting the global climate. Developing clean, sustainable, and renewable energy sources has thus become increasingly urgent [4-7]. However, renewable sources like solar and wind energy are intermittent, and unavailable at night or during calm weather conditions. Hence, the development of energy storage materials is of paramount importance. Energy storage materials offer a multitude of advantages across various sectors. Firstly, they facilitate the integration of renewable energy sources like solar and wind power by storing surplus energy for use during periods of low generation, thus ensuring a reliable and consistent energy supply [8-10]. Energy storage materials also enable smoother energy distribution by storing excess energy during off-peak hours and releasing it during times of peak demand, consequently reducing energy costs for consumers and utilities [11-13]. Furthermore, these materials enhance energy independence by maximizing the utilization of domestically available renewable resources, reducing dependence on imported fuels. Overall, the adoption of energy storage materials leads to a more resilient, efficient, and sustainable energy infrastructure, benefiting both the environment and society as a whole.

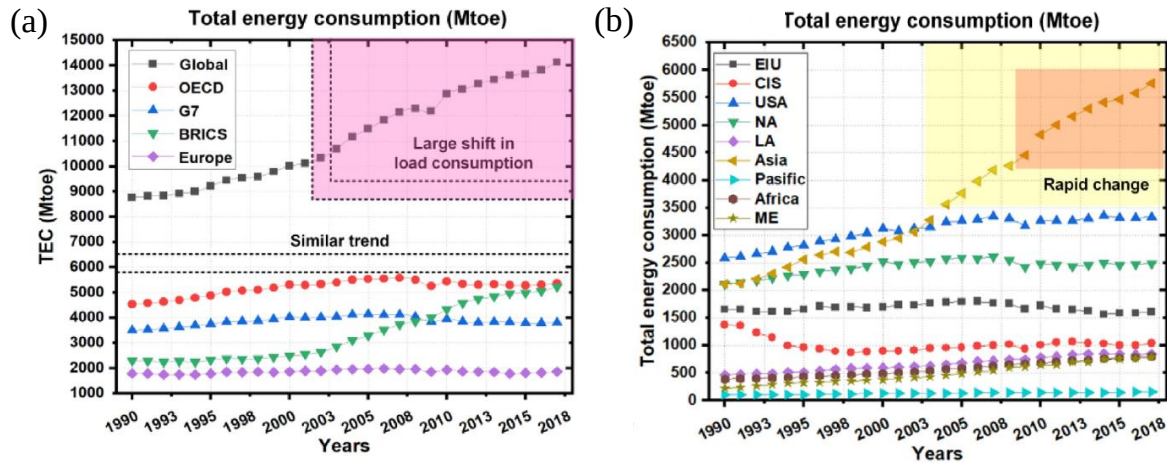


Fig. 1. (a) The overall energy consumption of Europe, Brazil, Russia, India, China, and South Africa (BRICS), The Group of Seven (G7), The Organization for Economic Cooperation and Development (OECD), and (b) the total energy consumption of the European Union (EU), Commonwealth of Independent States (CIS), United States of America (USA), North America (NA), Latin America (LA), Asia, Pacific, Africa and Middle East (ME) regions. Reproduced with permission. [1] Copyright 2020 Elsevier.

In this regard, phase change materials (PCMs) are energy storage materials that can absorb and release large amounts of thermal energy during phase transitions. This property allows PCMs to effectively store thermal energy during one phase and release it when needed, making them valuable for applications requiring temperature regulation or thermal energy storage (TES). TES plays a vital role in enhancing energy efficiency, optimizing renewable energy utilization, and balancing energy supply and demand. By storing excess heat or cold for later use, it adds flexibility to energy systems, helping to reduce dependence on fossil fuels and minimize energy waste. TES has numerous applications, particularly in industrial settings where large amounts of heat are generated and often wasted. Capturing and storing this heat allows for its reuse, improving process efficiency and lowering energy costs. In buildings, TES is commonly used in heating, ventilation, and air conditioning (HVAC) systems to provide more efficient temperature control. It stores thermal energy during off-peak periods and releases it when needed, reducing both energy consumption and peak grid loads. TES is also critical in solar thermal power plants, where solar energy is stored as heat and converted into electricity even when sunlight is unavailable, such as during cloudy days or at night. When PCMs harness their latent heat storage capacity, they contribute to energy efficiency and the integration of renewable energy sources by enabling the storage and utilization of surplus energy generated during peak production periods for later use during peak demand or when renewable sources are not actively generating power [14-16]. Currently, PCMs have been broadly applied in many fields, such as solar energy utilization [17-19], buildings [20-23], electronics [24-26], thermoelectric [27-31], and textiles [32, 33]. PCMs are categorized into four distinct types: solid-gas PCM, liquid-gas PCM, solid-solid PCM, and solid-liquid PCM. While solid-gas and liquid-gas PCMs exhibit high latent heat, their extensive gas production during phase transition restricts their industrial applicability. Comparatively, solid-solid PCMs typically feature higher phase transition temperatures but lower latent heat than solid-liquid PCMs. Solid-liquid PCMs offer several advantages, including minimal volume change during phase transition, a broad temperature range for phase change, and significant latent heat. Consequently, solid-liquid PCMs are widely utilized as heat storage materials. Solid-liquid PCMs can be further classified based on their chemical composition into inorganic and organic PCMs.

While PCMs exhibit excellent heat storage performance, their application is often limited by various drawbacks including low thermal conductivity, volume changes during phase transition, flammability, phase separation, and supercooling phenomenon [34-36]. More importantly, the workability of PCMs is significantly influenced by properties such as thermal conductivity and thermal storage capabilities. To enhance the thermal performances of PCMs, functional additives such as metals, metal oxides, or carbon-based materials are usually added [37-40]. Even though the addition of metals and metal oxides can improve the thermal characteristics of PCMs, they will also increase the overall density of the PCMs, which is not ideal for practical applications with a limit to the mass of the system. Therefore, carbon-based materials such as carbon nanotubes (CNTs), graphene, carbon fiber, and expanded graphite are

usually preferred due to their high thermal conductivity, relatively low density, and stable chemical nature [41-44]. Furthermore, the fabrication of these materials is becoming more affordable with emerging technologies. Amongst these carbon-based materials, the unique combination of high aspect ratio, exceptional thermal conductivity, mechanical reinforcement, flexibility, and tailorable properties of CNTs make them advantageous for incorporation with PCMs.

Several reviews have been published regarding incorporating different carbon nanomaterials into PCMs for enhanced thermal conduction and TES. To the best of our knowledge, no review article has been made available that focuses on nano-enhanced PCMs by adding CNTs. Here, we review the recent developments of PCMs with enhanced properties brought about by the incorporation of CNTs (Figure 2). The characteristics and physical properties of different types of CNTs, namely single-walled CNTs (SWCNTs), multi-walled CNTs (MWCNTs), modified CNTs, and PCMs-encapsulated CNTs are introduced. The relation between the different types of CNTs and the properties of composite PCMs is analyzed. Finally, the current challenges and an outlook of amalgamating CNTs with PCMs are discussed.

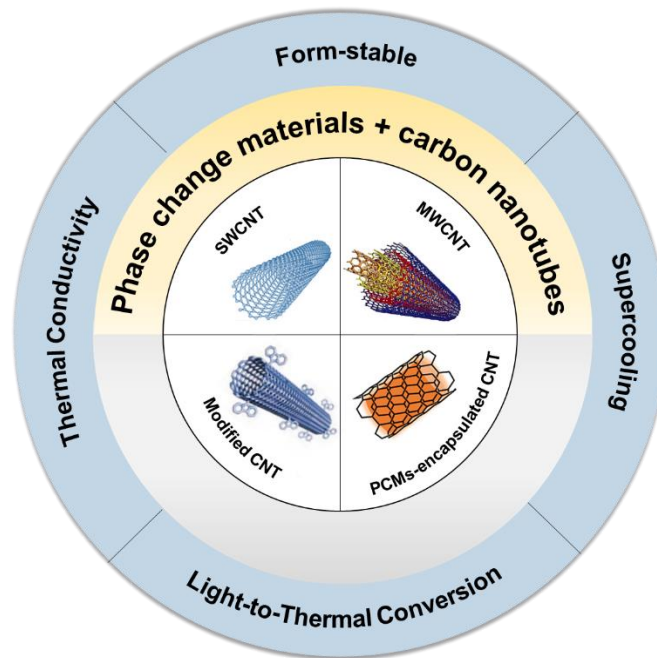


Fig. 2. Overview of PCMs incorporated with CNTs for properties enhancement covered in this work.

2. Single-walled carbon nanotubes

SWCNTs possess a single layer of graphene enclosed in a hexagonally closely wrapped cylindrical architecture, with lengths ranging from a few tens of nanometers up to several centimeters and diameters

from 1 to 3 nm [45, 46]. Additionally, they exhibit a high thermal conductivity of 3500 W/(m·K) at room temperature [47]. Compared to MWCNTs, they have fewer defects while offering greater surface area and uniformity. However, they contain contaminants and therefore need to be purified before usage to prevent altering their characteristics and affecting their potential applications. SWCNTs can be synthesized using approaches such as arc discharge, laser ablation, chemical vapor deposition, and sol-gel [48-51]. Due to the more complicated process involved in synthesizing SWCNTs, they require a catalyst and are more challenging to prepare in bulk compared to MWCNTs.

Yu et al. reported that the thermal conductivity of paraffin was improved by adding SWCNTs [52]. Different mass ratios of SWCNTs (1 and 2 wt%) were used and the results showed that the thermal conductivity of paraffin was increased by 26.1% and 56.5%, respectively. To further enhance the thermal conductivity of the PCM, the authors introduced a porous media such as metal foam, which is known to improve the thermal conductivity of PCMs due to its large specific surface area and high thermal conductivity [53, 54]. Compared with the composite that consisted of only paraffin and metal foam, the thermal conductivity of the composites with the addition of 1 and 2 wt% SWCNTs and metal foam was enhanced by 12% and 21.3%, with only a slight reduction in enthalpy of 3.8% and 4.9%, respectively. Similarly, Xu et al. investigated the effect of combining CNTs with paraffin as a filler compared to thermal grease on the pulsating flow in a microchannel heat sink [55]. Three different mass ratios (5, 10, and 15 wt%) of CNTs/paraffin composites were prepared and it was noted that the addition of CNTs has a significant influence on the thermal conductivity, with an enhancement of 109.7%, 234.6%, and 323.2%, respectively. In addition, the overall thermal performance of the heat sink increased with the loading of CNTs, with a decrease of 3.1 ~ 6.6 °C in the maximum temperature for 15 wt% of CNTs. To enhance the thermal conductivity of paraffin (RT 4) for cooling applications in a vertical open-type refrigerated display cabinet, Wu et al. added four different mass fractions of CNTs and studied their thermo-physical properties [56]. The paraffin/CNTs composite was prepared through dispersant and ultrasonic oscillation with 1, 3, and 5 wt% CNTs. The results revealed that the optimal loading of 3 wt% CNT increased the thermal conductivity by 30.3% (Figure 3a) and the highest thermal conductivity values occurred during the phase transition process due to the increase in temperature which enhances the molecular vibration of the PCM. Meanwhile, the melting phase change latent heat decreases as the mass of SWCNTs increases in the composite. The PCM/CNTs composite was applied to the shelves of the vertical open-type refrigerated display cabinet and results showed that 3 wt% CNTs further reduced the temperature difference of the lengthways and depth-wise of the shelf by 12% and 4.6%, respectively when compared to the shelf being filled with only paraffin.

Besides investigating the effects of SWCNTs on paraffin, which is considered one of the most widely used organic PCMs, molten salts and nanomaterials-based composite PCMs (CPCMs) have been studied as well due to their significant role in concentrating solar power (CSP) systems. Molten salt-based PCMs are often used as the heat transfer fluid and it is desirable to improve their thermal

conductivity and heat capacity to improve the overall efficiency of CSP devices and therefore reduce their usage. With the rapid advancement of computer technology and its broad application in thermal physics, molecular dynamic (MD) simulations, combined with molecular design and modeling, have become a valuable and powerful tool for exploring thermal properties that are difficult or impossible to measure through experimental methods. To reduce experimental costs and gain deeper insights into the mechanisms behind thermal property enhancement, it is crucial to develop simulation methods that can predict and control the thermal behavior of PCMs using MD simulations. To gain an insight into how molten salt-based PCMs can be designed to maximize their effectiveness for solar energy utilization in CSP technology as a heat transfer fluid, Yu et al. studied the thermal properties of NaCl-SWCNTs-based CPCM using MD simulation at a nanoscale level [57]. Different thermal properties of the composites such as the melting point, thermal conductivity, specific heat capacity, and melting enthalpy were predicted using MD simulation. It was found that an increased mass weightage of SWCNTs decreases the melting point of the composites, with 0 (CPCM₀), 3.5 (CPCM₁), and 6.7 (CPCM₂) wt% SWCNTs having a melting temperature of 1220 K, 1096 K, and 1078.6 K, respectively (Figure 3b). On the other hand, the thermal conductivity of the composite was significantly enhanced by 8.75% and 38.59% for CPCM₁ and CPCM₂, respectively while the specific heat capacity also increased by 2.30% and 5.87%. To understand the microscale mechanism of SWCNTs on the thermal performance enhancement of CPCMs, simulation studies were conducted to analyze the microscopic energy components of Na⁺ and Cl⁻ ions and their interaction in the CPCMs. The results revealed that as the mass fraction of SWCNTs increased, the potential and kinetic energy in the solid state increased while the van der Waals energy decreased. This phenomenon indicated that SWCNTs strengthen the interaction between the atoms in the system and facilitate their collisions, thus enhancing heat transfer. However, due to the absorption of Na⁺ and Cl⁻ ions on SWCNTs, a liquid layer was formed around it and the melting enthalpy decreased by 19.84% and 36.37% for CPCM₁ and CPCM₂, respectively (Figure 3c). To further understand the micro mechanism of thermal properties enhancement and regulation of SWCNTs in molten-based PCMs for CSP systems, Yu et al. conducted experimental and MD simulations to study the thermal storage performance of KNO₃-SWCNTs-based CPCM [58]. Through the experiments, similar trends could be observed when the SWCNTs mass fraction increases. It was found that with the addition of 4.43 and 8.48 wt% of SWCNTs, there was an increase in thermal conductivity by 2.26% and 28.01% and an increase in the specific heat capacity by 11.72% and 16.55%, respectively. The melting enthalpy showed a decrease of 32.31% and 45.19%, respectively as well. From the MD simulations and DSC results, the mechanism of thermal storage performance enhancement was revealed and it was noted that SWCNTs facilitate the potential energy interactions between the atoms and promote the thermal link and surface nanolayer formations, increasing specific heat capacity and thermal conductivity. Additionally, when a different surface charge modification method was carried out through simulation to treat the SWCNTs' surfaces, it was noted that surface positive modification of SWCNTs changes the microstructure of the PCM and regulates the spatial

distribution of ions, thus effectively regulating the thermal storage performance. As much as thermal conductivity enhancement is crucial for PCMs, the viscosity of CPCMs is a factor to consider as well. Viscosity plays a significant role in determining the heat transfer performance of PCMs. Even though the thermal conductivity of the CPCMs was enhanced, the viscosity increased by 27.68% and 39.05% for 4.43 wt% and 8.48 wt% SWCNTs, respectively as well. When CNTs are added to PCMs, it will result in an increase in viscosity and this will reduce the CPCM's ability to flow easily, leading to weaker natural or forced convection. As a result, the heat transfer rate by convection will decrease. On the other hand, to determine which carbon nanomaterial can improve the thermal conductivity of high-temperature carbonate eutectic salts the most, Tao et al. utilized five different mass ratios (0.1, 0.5, 1.0, 1.5, and 2.5 wt%) of SWCNTs, MWCNTs, graphene, and fullerene and prepared the CPCMs through the solution evaporation method [59]. The correlation between thermal conductivity and columnar structure was investigated, and it was reported that SWCNTs have the greatest influence on thermal conductivity, followed by MWCNTs, graphene, and fullerene (Figure 3d). The columnar structure nano-layer of SWCNTs allows useful heat conduction links to form between the salts and nanomaterial surface, which enhances thermal conductivity. However, the thermal conductivity of the CPCM with SWCNTs reaches its peak enhancement of thermal conductivity of 56.98% at 1.5 wt% and decreases afterward due to aggregation. It was also concluded that the nanomaterial additives have limited effects on the total TES capacity and that thermal conductivity enhancement plays a more important role. Even though SWCNTs offer a better thermal conductivity enhancement compared to MWCNTs, the latter has a better ease of mass production, lower cost, and higher homogeneous dispersibility in the matrix phase and therefore is mainly utilized in applications instead.

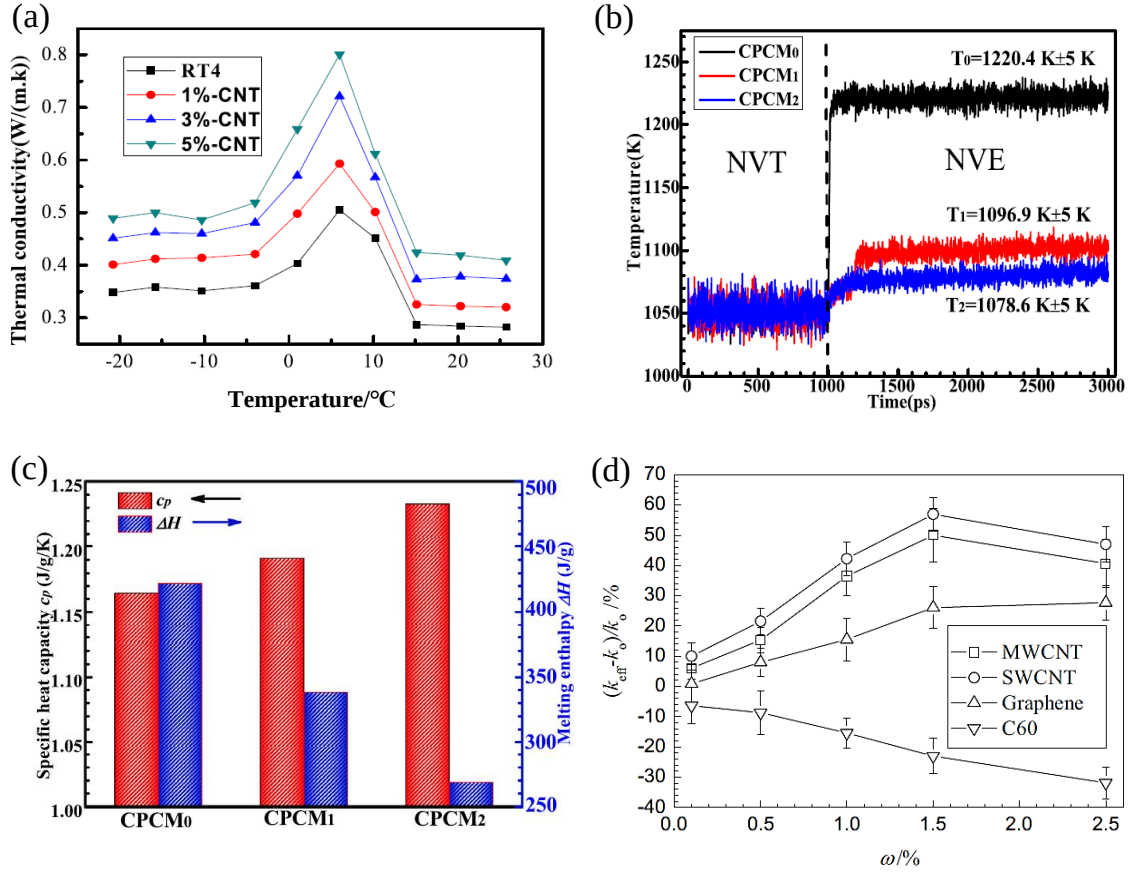


Fig. 3. (a) Thermal conductivity of RT4, 1%-CNT, 3%-CNT, and 5%-CNT composites. Reproduced with permission. [56] Copyright 2019 Elsevier. (b) Melting point, and (c) Melting enthalpy, ΔH , and specific heat capacity, c_p of CPCM₀, CPCM₁, and CPCM₂. Reproduced with permission. [57] Copyright 2021 Elsevier. (d) Effects of nanomaterial microstructure and mass fraction on CPCM thermal conductivity. Reproduced with permission. [59] Copyright 2015 Elsevier.

Form-stable PCMs (FSPCMs) or shape-stabilized PCMs (SSPCMs) play a crucial role in TES as they can undergo phase transition while maintaining their structural integrity [60-63]. When integrated with SWCNTs, these PCMs offer an innovative and promising development. During phase transitions, PCMs can absorb and release large amounts of energy, while SWCNTs provide exceptional thermal conductivity and structural stability. As SWCNTs are incorporated into PCM matrices, they provide a robust support structure, prevent PCM leakage, and enhance thermal conductivity. As a result, the composite material maintains its structural integrity even during phase transitions, ensuring energy storage and release are both efficient and reliable. Qian et al. fabricated a series of form-stable nanocomposites using polyethylene glycol (PEG) and SWCNTs via a facile impregnation method, with the mass ratio of SWCNTs ranging from 2 to 10 wt% [64]. At the time of reporting, they obtained the highest absorption rate of PEG of 98% by SWCNTs and the composites displayed exceptional shape

stability, with no trace of leakage even after 400 thermal cycles. The results showed that SWCNTs enhanced the thermal conductivity of pure PEG significantly, with an enhancement of 1329% from 162% in the solid state and 533% from 21% in the liquid state. In addition, the melting point of the PEG/SWCNTs composites shifted lower while the freezing point shifted higher, which resulted in a reduction of supercooling from 26.4% to 38.8% with the increase of SWCNTs loading. Another FSPCM using PEG and SWCNTs was prepared as well, with expanded vermiculite added as an encapsulation material [65]. The latent heat and heat transfer capability of the composite were measured, and it was found that the latent heat of the composite increased during melting due to the decreasing inhibition effect of expanded vermiculite and increasing absorption capacity of PEG. The heat transfer of the composite was greatly enhanced due to the presence of SWCNTs fillers as the thermal conductivity increased linearly with the increasing content of SWCNTs. The interfacial thermal resistance could be reduced by the interfacial interaction between PEG and SWCNTs network that are responsible for forming rapid thermal conductive pathways that enhance thermal conductivity. The thermal conductivity increases simultaneously via the synergistic effect of SWCNTs and expanded vermiculite, while the latter also acted as a suitable encapsulation material and form stabilizer which prevents the leakage of the PCM during melting. Liu et al. investigated a ternary system of capric acid (CA), diatomite (Dt), and CNT as FSPCM where Dt served as the supporting material to absorb CA [66]. The composite was prepared via the direct melting-impregnation method with the addition of 1, 3, 5, and 7 wt% of CNT. The thermal conductivity of the CA/Dt/CNTs composite with 7 wt% CNT increased by 1.56 times (0.15 to 0.48 W/(m·K)) compared to that of CA/Dt (Figure 4a). This is due to the huge number of heat transfer channels in the Dt porous disk-like structure that creates directionality of the energy wave which improves the heat transfer efficiency of the Dt, leading to lower wave resistance and higher thermal conductivity (Figure 4b). Dt also acted as a support material to prevent leakage and good form stability could be achieved with a loading of 54% CA in the composite. The addition of CNT also improves the Dt transfer channel by further reducing the wave resistance and enhancing the thermal conductivity by 260% (1 wt%), 273% (3 wt%), 306% (5 wt%), and 320% (7 wt%) when compared to pure CA. Li et al. used an interesting approach to integrate CNTs and PCMs to form SSPCMs by using the core-sheath structure of CNT and metal-organic frameworks to enhance the energy storage capacity and heat transfer ability [67]. The porous CNT core sheath was synthesized by carbonation of CNT@ZIF-8, and stearic acid (SA) was impregnated into the structure via the wet impregnation method (Figure 4c). The porous carbon (PC) has high porosity and a large specific surface area which are used to stabilize the PCM as the PCM is absorbed by the capillary force of the PC sheath. The SA/CNT@PC composite showed a 200% increase in thermal conductivity as compared to the pristine PCM. The thickness of the carbon layer did not affect the thermal conductivity of the composite as it showed a stable thermal conductivity value of 1.023 W/(m·K) throughout the increment from 10 nm to 40 nm. Moreover, the porous carbon homogenous coating absorbed more SA compared to that of pure porous carbon due to the *in situ* growing process.

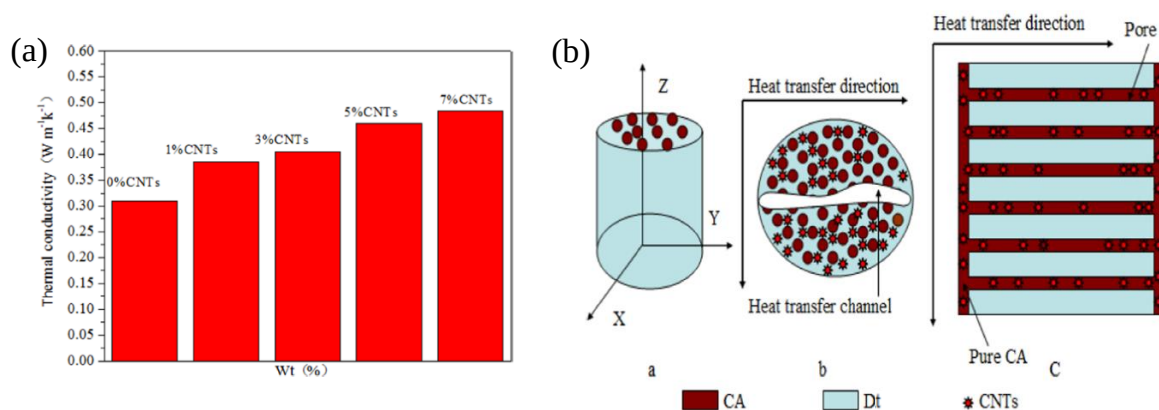


Fig. 4. (a) Thermal conductivity of CA/Dt FSPCMs with different mass fractions of the CNT additive, and (b) Improvement in the mechanism of the thermal conductivity of CA. Reproduced with permission. [66] Copyright 2019 American Chemical Society.

Besides enhancing the thermal conductivity of PCMs, CNTs can be used to improve light-to-thermal conversion and thermoregulated capabilities as well. Li et al. prepared coaxial fibers via electrospinning technology with CNT, poly(tetradecyl acrylate)(PTA) as PCM and core, and poly(ethylene terephthalate)(PET) as sheath [68]. The PET/PTA/CNT composite shows great thermal stability due to its sheath-core structure, where the PET sheath acts as a protection and CNT acts as an excellent thermal conductor. The addition of CNT demonstrated good spinnability and dispersion, which averted mass loss during solution extraction. Meanwhile, the phase transition of PTA and the wide UV-vis light absorbance of CNT contributed to the good thermal storage behavior of the composites. It was also noted that under a simulated solar illumination at 100 W/cm², the maximum temperature of the PET/PTA/CNT composite with 2 wt% CNT was 23.2 °C higher than PET/PTA after 600s illumination (Figure 5a), indicating that CNT is useful for temperature-increasing and light-harvesting. To compare the performance of this composite against PET membrane as a greenhouse film, the authors took a step further by placing these membranes inside a bottle to determine the effect that these two membranes have on the greenhouse temperature. The results showed that the temperature of the bottle covered with the composite was 3 °C higher than the one covered with PET after 900s illumination (Figure 5b). The combined thermoregulated capability and light-to-thermal ability of the sheath-core structure of this composite are desirable for vegetable growth during the application in a greenhouse. To achieve light-to-thermal conversion and enhance the thermal conductivity of CPCMs synchronously, Qian et al. prepared PEG-infiltrated CPCMs by filling the pores of Dt using two different types of carbon fillers, SWCNTs (PEG/DCNs) and carbon nanoparticles (PEG/DCPs) [69]. The temperature variations of PEG, PEG/Dt, PEG/DCNs, and PEG/DCPs composites (Figure 5c) during solar irradiation showed that the temperature of the PEG/DCNs and PEG/DCPs composites rose rapidly, and this is attributed to the enhanced photon absorption capability endowed by the fillers, causing the PEG to heat up faster. In

particular, the temperature of PEG/DCNs rose faster than PEG/DCPs, indicating that SWCNTs is a more attractive photon captor and molecule heater. The light-to-thermal conversion efficiency (Figure 5d) of PEG/DCNs (~87.7%) is also higher than that of PEG/DCPs (~77.3%) and this is because SWCNTs can form a more continuous and denser heat transfer route and ensure that more sunlight is absorbed. In addition, it was noted that the thermal conductivity of PEG/DCNs is 130% higher than that of PEG/DCPs. The more favorable three-dimensional (3D) structure of Dt and SWCNTs compared to carbon nanoparticles further enhanced the thermal conduction due to the larger contact area of SWCNTs. 0D carbon nanoparticles resulted in some disconnected parts in thermally conductive pathways due to aggregation while the nearly continuous 3D network of SWCNTs can be formed from the linking of end-to-end with one-dimensional (1D) SWCNTs.

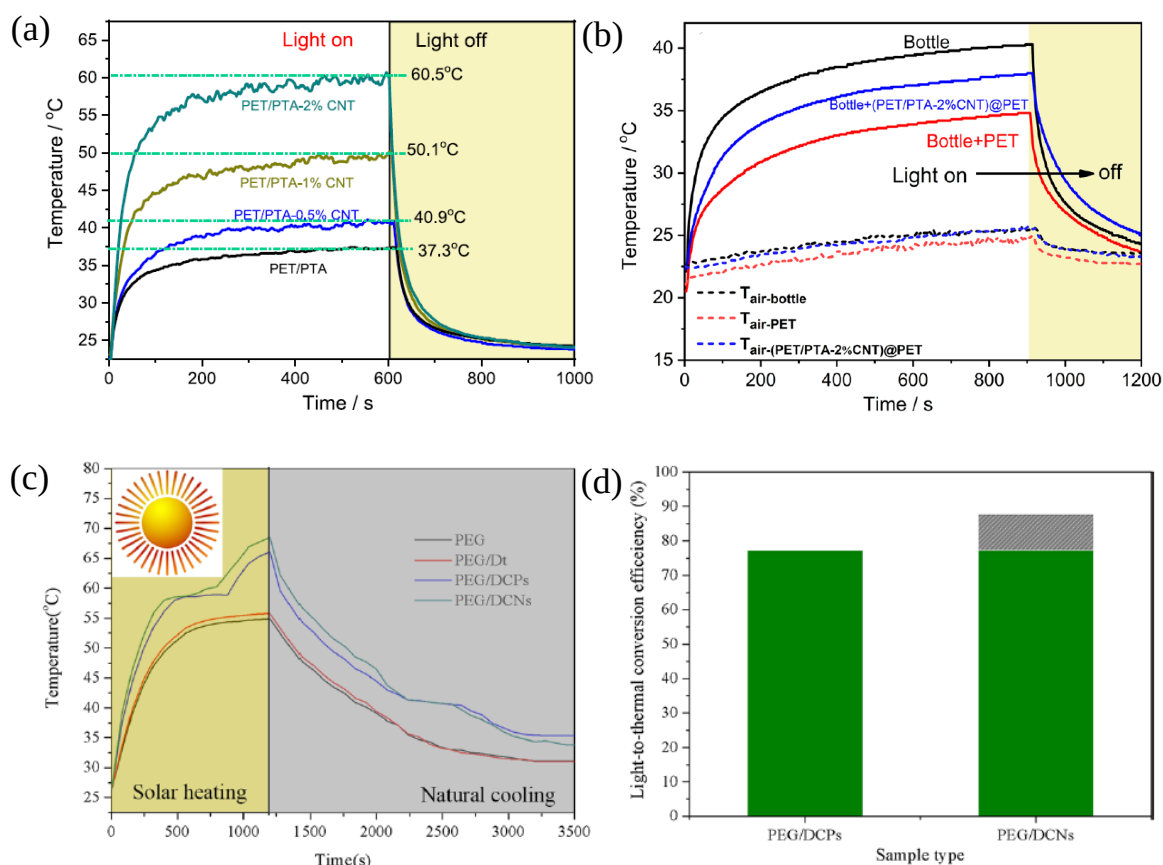


Fig. 5. (a) Light-to-thermal conversion curves of PET/PTA and PET/PTA/CNT coaxial fibers, and (b) Temperature variation process for the covered bottle. Reprinted (adapted) with permission from [68] Copyright 2019 American Chemical Society. (c) Time–temperature curves of PEG, PEG/Dt, PEG/DCPs and PEG/DCNs, and (d) TES efficiency of PEG/DCPs and PEG/DCNs. Reprinted (adapted) with permission from [69] Copyright 2019 American Chemical Society.

3. Multi-walled carbon nanotubes

MWCNTs are coaxial cylinders with a hollow core surrounded by multiple concentric graphene layers. They have an internal diameter of 1-3 nm, with the diameter ranging from 1-100 nm and length spanning up to several micrometers. They were reported to have a thermal conductivity of 3000 W/(m·K) at room temperature [70]. Due to the trap centers that tend to come between the layers, MWCNTs are susceptible to varying defects and hence have lower thermal stability compared to SWCNTs [71]. However, they exhibit higher mechanical stability and resilience compared to SWCNTs due to their multilayered structure. The presence of multiple walls can distribute mechanical stress more effectively, making MWCNTs more resistant to deformation and damage. MWCNTs can be synthesized via the same methods aforementioned as SWCNTs and the methods used depend on the desired scalability, properties, and production efficiency.

To regulate the temperature of a photovoltaic (PV) cell, Abdulmunem et al. explored the possibility of using a passive cooling method by using paraffin wax (PW), copper foam matrix (CFM), and MWCNTs [72]. Different mass ratios (0.05%, 0.1%, 0.15%, and 0.2%) of MWCNTs were used, and scanning electron microscopy (SEM) images revealed that as the mass ratio increased, the number of tangled tubes' bundles increased as well. During the charging process, the PV cell with PCM/CFM reduced the maximum cell temperature by 5.93% compared to the PV cell with PCM only. The presence of the CFM eliminated the internal convection in the PCM and channeled them into the CFM instead, resulting in an accelerated and homogenous melting process and more heat absorbed by the PV cell. By adding 0.2 wt% MWCNTs, the thermal equilibrium between the CFM and PCM was enhanced. The tangled tubes' bundles of MWCNTs further increased the thermal conductivity and specific heat capacity, leading to higher heat absorption from the cell and reduced the cell temperature by a further 2.86%. The electrical performance of the PV cell was improved with the addition of CFM and MWCNTs, which resulted in an enhancement of the electrical efficiency of the PV cell by 5.68%. Dinesh et al. investigated the heat transfer behavior of a ternary eutectic PCM based on oleic acid, isopropyl palmitate, and butyl stearate and added 0.02, 0.04, 0.06, 0.08, and 0.1 wt% of MWCNTs [73]. It was found that the pure eutectic PCM took 1200s for the solidification process while this rate was enhanced by 34.45% for 0.1 wt% MWCNTs (Figure 6a). This enhancement is due to MWCNTs being a heterogeneous nucleating agent that serves as a core for the PCM and facilitates the crystallization process. The uniform dispersion of MWCNTs without aggregation and non-settling behavior also led to quicker charging and discharging rates. In addition, the thermal conductivity of the composites showed an increasing trend with the mass % of MWCNTs. It reached a maximum increment of 67.28% with 0.10 wt% of MWCNTs, with only a slight reduction in latent heat storage capacity and phase change temperature. MWCNTs enhanced the thermal stability of the composites as well, where the decomposition temperature was increased from 260 °C for the pure eutectic PCM to 335 °C with the addition of 0.10 wt% MWCNTs (Figure 6b). On the other hand, the viscosity of the composites was

found to decrease linearly with the increase in temperature and this is attributed to the increased kinetic energy of the PCM and the reduced shear force between the MWCNTs and PCM (Figure 6c). Even though the viscosity increased with increasing mass % of MWCNTs added, the maximum increment was noted to be 77% and further addition of MWCNTs does not exceed this value by more than 1%. The melting time of CA incorporated with different volume fractions of MWCNTs (0.01, 0.02, and 0.025%) alongside other thermal properties such as thermal conductivity, and c_p was also investigated by Yadav et al. [74]. The variation of liquid fraction with time for CA and the different composites was monitored and it was found that 0.02 vol% of MWCNTs is the optimum loading to reduce the melting time, which is 66.66% lower than CA. Besides, optimum thermal conductivity and c_p were achieved as well with this loading compared to the others. Moreover, the energy and exergy of the storage medium had an enhancement of 61.9% and 70.92% as compared to that of CA due to the hexagonal structure of MWCNTs which improves the interaction between the composite due to the surface resistance.

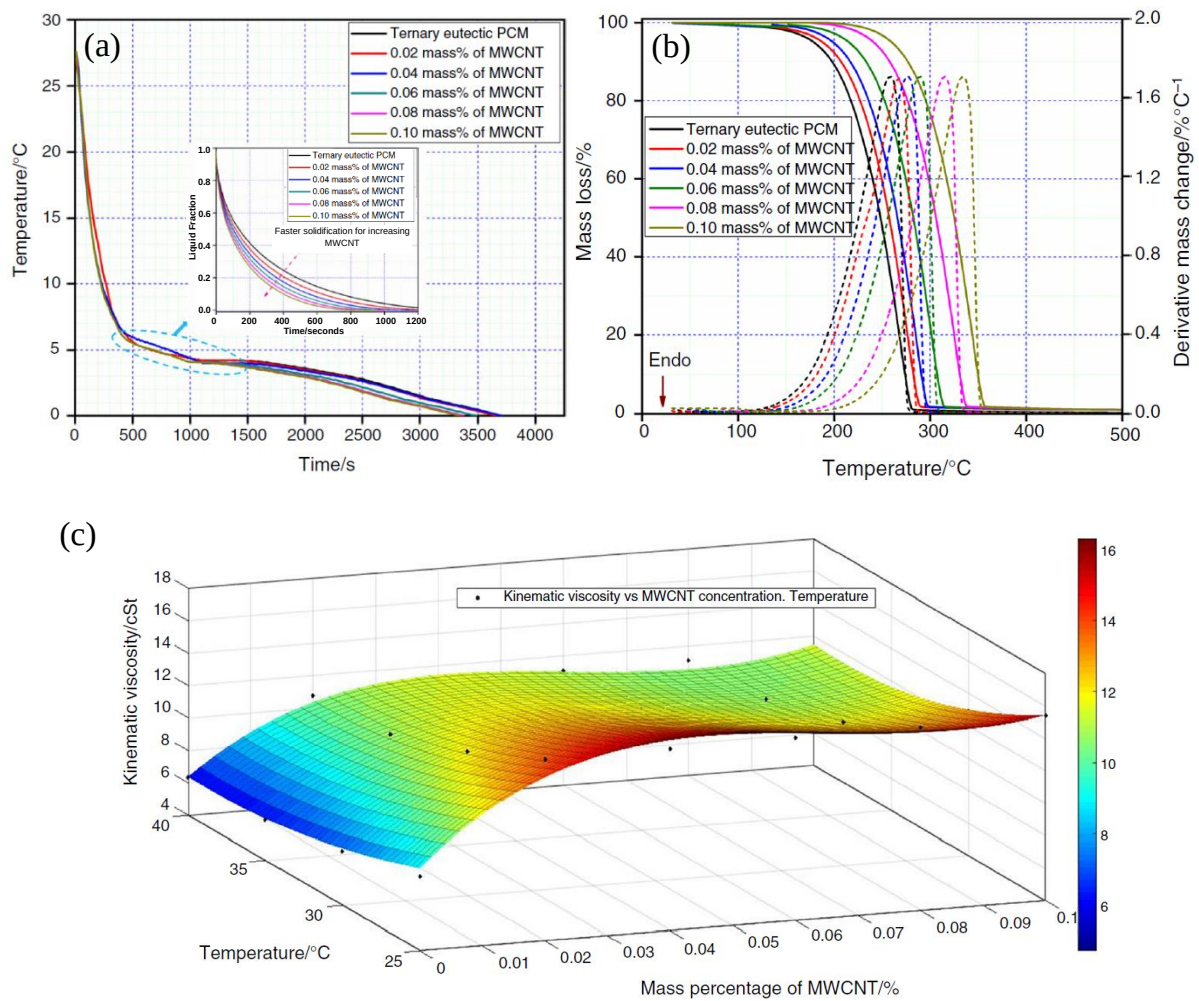


Fig. 6. (a) Solidification curves of the spherical encapsulated pure and composite ternary eutectic PCM for increasing MWCNT content, (b) TG-DTG curves of the pure and composite ternary eutectic PCM,

and (c) Viscosity variation of pure and composite ternary PCM with respect to MWCNT concentration and temperature. Reproduced with permission. [73] Copyright 2021 Springer Nature.

Similar to SWCNTs, MWCNTs can be incorporated into PCMs to confer enhanced thermal conductivity and structural stability. In a study conducted by Zhang et al., FSPCMs were prepared using MWCNTs, paraffin, and expanded perlite (EP) via the vacuum impregnation method [75]. Different MWCNTs mass fractions (0, 1.83, 3.62, and 5.27 wt%) were used and a thermal conductive network with a high length-width ratio was obtained. An increasing mass fraction of MWCNTs resulted in a more compact thermal network and subsequently a more enhanced thermal conductivity. Additionally, a higher mass fraction of MWCNTs also led to a faster thermal storage and release rate, where pure paraffin took 290s for solidification while the composite with 5.27 wt% MWCNTs took 190s (Figure 7a). The composite also exhibited good thermal reliability after 200 thermal cycles (Figure 7b). Sari et al. impregnated PEG with raw Dt and MWCNTs to prepare SSPCMs and the thermal conductivity of the raw Dt/MWCNTs/PEG composites were enhanced between 73% to 93% with 0.57, 1.70, and 2.50 wt% MWCNTs [76]. The leakage test showed that there was no leakage of PEG up to 51.0 wt% due to the structural skeleton of MWCNTs (Figure 7c), which allowed PEG to be absorbed into the raw Dt/MWCNTs composite while maintaining its structural stability. Not only do MWCNTs increase the absorption ratio of PEG, but they also reduce the heat-releasing/storing times as well. On the other hand, Luo et al. encapsulated paraffin and MWCNTs into polypropylene hollow fiber membranes to fabricate flexible SSPCMs [77]. A maximum encapsulation capacity of 80.97% with good shape stability was achieved and the addition of MWCNTs enhanced the thermal conductivity by 100% with just 0.93 wt% of MWCNTs. Polymeric materials encompass a broad range of substances prized for their versatility [78, 79], durability [80], and electrochromic capabilities [81], finding utility across numerous applications [82-86]. However, polymer-based SSPCMs have low thermal conductivity and inferior mechanical properties. To leverage the excellent mechanical and thermal properties of MWCNTs, Mu and McNally mixed them with paraffin and high-density polypropylene (HDPE) and prepared SSPCMs by the extrusion method [87]. 0.1, 0.5, 1, 2, and 3 wt% MWCNTs were used and SEM images indicated that they were evenly dispersed in the SSPCM matrix. However, it was noted that the 0.5 wt% MWCNTs blend appeared isolated whilst an interconnected network was formed for the 2 wt% MWCNTs blend. The thermal conductivity of the composites increased with an increase in mass ratios of MWCNTs and more importantly, their mechanical properties were enhanced as well. In general, the mechanical properties (e.g., yield stress and flexural modulus) increased with increasing MWCNTs ratios due to the excellent interfacial interactions between paraffin, HDPE, and MWCNTs, where MWCNTs acted as enhanced fibers. However, for Young's modulus and flexural stress, the composite without MWCNTs has higher values compared to those with a lower MWCNTs loading (<1 wt%). It

was proposed that the low loading of MWCNTs (0.1 and 0.5 wt%) could not form an inter-connected network and therefore does not significantly affect the matrix modulus.

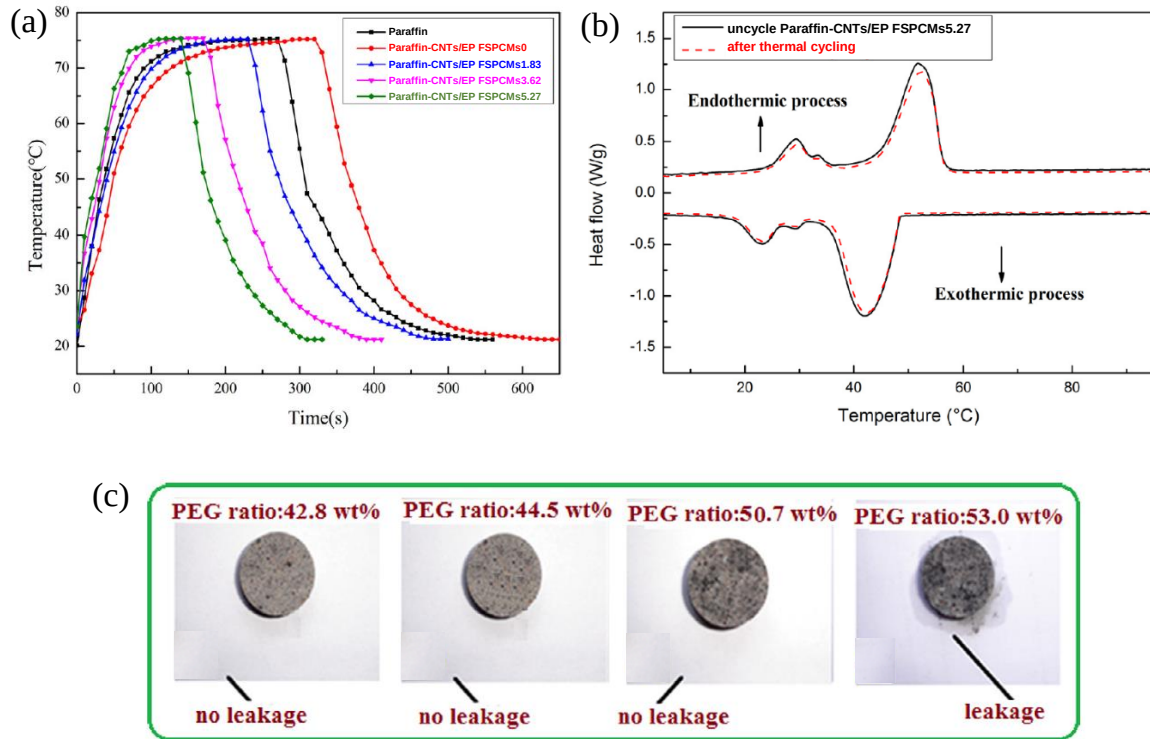


Fig. 7. (a) Melting and freezing temperature curves of paraffin and paraffin–CNTs/EP FSPCMs, and (b) DSC curves of paraffin and paraffin–CNTs/EP FSPCMs before and after 200 thermal cycles. Reproduced with permission. [75] Copyright 2017 Elsevier. (c) The leakage test results obtained at 20 °C depending on the PEG ratio in the SSPCM with CNTs. Reproduced with permission. [76] Copyright 2018 Elsevier.

CNTs possess outstanding thermal, electrical, and mechanical characteristics, which make them highly desirable for numerous applications. When paired with other nanomaterials or additives, CNTs frequently exhibit synergistic effects, where the composite material outperforms the individual components. This enhanced performance is often due to the complementary physical, chemical, and structural properties of CNTs and their counterparts, leading to multifunctional composites with superior capabilities. To overcome the intrinsically low thermal conductivity of PW, Hu et al. deposited a continuous diamond film on copper foam via chemical vapor deposition and MWCNTs were catalytically grown on the surface of the diamond film [88]. To disperse the MWCNTs into the diamond-coated copper foam, vacuum impregnation was carried out at 100 °C for 3 h. With this stable hierarchical structure of continuous diamond-carbon nanotube foam, it was revealed that the diamond

film with superior thermal conductivity acts as the first main channel for thermal conduction while the CNT networks act as the secondary channels for uniform thermal conduction within the composite. As a result, the thermal conductivity was enhanced from 0.105 W/(m·K) for PW to 9.72 W/(m·K), one of the highest enhancements ever reported at that time. Nevertheless, the cost of nanodiamonds used in the preparation of the CPCMs might impede its use for future applications. A 3D interpenetrating network PCM composite was fabricated by Bao et al. by incorporating graphene oxide aerogel (GA), MWCNTs, and carbon spheres (CSs) with PEG via vacuum impregnation [89]. To prepare the CPCMs, GA or B-GA (comprised of graphene oxide, MWCNTs, and CSs) aerogels were mixed with PEG to get the aerogels fully submerged. Thereafter, the vacuum impregnation process was conducted at 80 °C for 30 min. Graphene oxide has a much lower absorbance compared to MWCNTs and CS and the addition of the latter two components resulted in a higher overall absorbance in the composite (B-GA)(Figure 8a). Under simulated solar radiation, the temperature of B-GA rose more rapidly compared to PEG and GA (Figure 8b) due to the enhanced light absorption capacity and thermal conductivity, having an enhancement in heating rate by 766.7% compared to PEG (Figure 8c). The thermal conductivity of B-GA improved by 181.58% compared to PEG as well and the photothermal conversion efficiencies of GA and B-GA are 53.2% and 89.3%, respectively (Figure 8d). To simultaneously enhance the thermal conductivity and solar absorption capacity of PW, Lin et al. added expanded graphite (EG) and MWCNTs, where EG served as the porous matrix and MWCNTs as the light absorbent [90]. MWCNTs were added to melted PW and the solution was sonicated for 15 min. EG was added to the mixture and stirred at a temperature of 80~90 °C, followed by a vacuum process for 20 min. Based on the diffuse reflectance spectra, the addition of 5 wt% MWCNTs (5CNTs-PW) decreased the reflectance from ~60% for PW to ~3-4% as compared to ~10% for 5 wt% EG (5EG-PW)(Figure 8e), indicative of strong light absorption across a wide spectral range by MWCNTs. However, increasing the mass fraction of the additives did not enhance the solar absorption any further (Figure 8f). The addition of 2 wt% MWCNTs and 5 wt% EG resulted in the maximum solar absorption capacity of 94.2% and increasing the loading of MWCNTs was found to decrease this value due to possible aggregation. Meanwhile, Kardam et al. investigated the heat transfer characteristics of nano graphite and MWCNTs with magnesium nitrate hexahydrate using both conventional heating and solar illumination setups for solar TES [91]. The path of lower resistance in these carbon materials allows the phonons to travel more easily and at the same time, their aspect ratio and interfacial contact area with the PCM facilitate the heat transfer rate. As a result, the composites with these carbon nanofillers exhibited higher thermal conductivity and a faster heat rate compared to pristine PCM in both experimental setups. In particular, there is a significant reduction in melting time for the solar illumination setup, where the pristine PCM took 45 min to melt while the composite with nano graphite and MWCNTs took 1 min and 30 s, respectively. MWCNTs being a molecular heater and effective photon captor, absorbs solar energy and converts it into thermal energy via plasmonic heating in addition to conventional thermal diffusion. The incident solar energy accelerates the free electrons in MWCNTs, causing them to oscillate at their resonant frequency and the

energy gained will be turned into heat energy [92]. Thermal diffusion will then carry this localized energy into the surrounding medium.

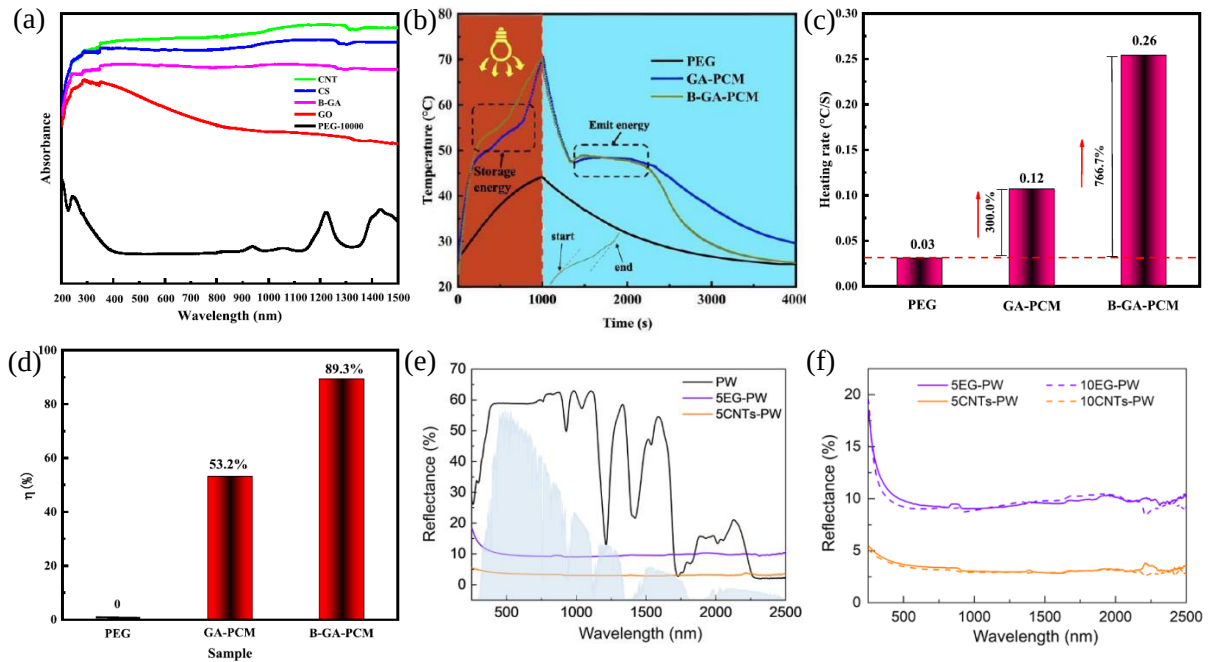


Fig. 8. (a) UV–vis–NIR absorption spectrum of MWCNTs, CS, GO, B-GA, and PEG, (b) photothermal conversion curves of GA-PCM, B-GA-PCM, and PEG, (c) heating rate of PEG, GA-PCM, and B-GA-PCM, and (d) photothermal energy conversion efficiency of GA-PCM and B-GA-PCM. Reprinted (adapted) with permission from [89] Copyright 2021 American Chemical Society. Diffuse reflectance spectra of (e) PW and the composite PCMs with 5 wt.% additives, and (f) composite PCMs with additives of different mass fractions. Reproduced with permission. [90] Copyright 2022 Elsevier.

CNTs reduce supercooling in PCMs by enhancing thermal conductivity and acting as nucleation sites. Their ability to efficiently distribute heat minimizes temperature gradients, ensuring the PCM cools uniformly and reaches crystallization temperature more consistently. Additionally, CNTs lower the energy barrier for nucleation by providing surfaces that facilitate the solidification process. This combination of improved heat transfer and increased nucleation points allows CNTs to significantly reduce supercooling, promoting more efficient and reliable phase transitions. However, it should be noted that poor dispersion of CNTs within the PCMs might result in reduced nucleation efficiency while a low chemical compatibility with PCMs might hinder their effectiveness in promoting nucleation. Wang et al. prepared CPCMs using erythritol (ER), graphite foam (GF), and MWCNTs with different mass ratios of 0 (GF₀), 1 (GF₁), 2 (GF₂), and 4 wt% (GF₄) [93]. The water contact angle results indicated that the surfaces of the composites have water contact angles larger than 90° and hence are hydrophobic. The water contact angle increased with an increase in the mass fraction of MWCNTs and this

corresponds to greater surface roughness according to the Wenzel wetting model [94]. A rougher surface can provide more nucleation sites for heterogeneous nucleation thereby increasing the probability of nucleation [95]. As a result, the supercooling degree of ER/GFs is reduced, where GF₄ has the lowest supercooling degree of 46.9 °C compared to ER's 93 °C. Muthoka et al. investigated the supercooling effect of MWCNTs by adding different mass fractions (0.2-1 wt%) to barium chloride dehydrate (BaCl₂·2H₂O) [96]. It was found that BaCl₂·2H₂O has a lower supercooling degree when it was cooled at a lower temperature and vice versa as a faster heat removal/charging rate will enhance the rate of nucleation. The addition of 1 wt% MWCNTs reduced the supercooling degree by 92% and it was noted that MWCNTs facilitated the nucleation in two ways. Firstly, the larger size of MWCNTs compared to BaCl₂·2H₂O provided a nucleus for the latter's molecules to accumulate and form crystals. Secondly, the high thermal conductivity of MWCNTs allows a faster heat removal rate during crystallization and therefore accelerates the growth of crystals. Similarly, Sathishkumar and Cheralathan added different mass ratios of MWCNTs (0.25, 0.5, and 0.75 wt%) to deionized water (as a base PCM) to enhance the thermal properties under different heat transfer fluid temperature conditions [97]. The degree of supercooling was noted to decrease with increasing MWCNTs concentrations and at a mass ratio of 0.75 wt% MWCNTs, supercooling was eliminated. Wu et al. added MWCNTs and nucleating agents such as CaCl₂ and Ca(OH)₂ to MgCl₂ eutectic solution and interestingly, there is a synergistic effect between these materials on the supercooling degree [98]. Xanthan gum (XG) was added as a thickener, stabilizer, and suspending agent as it can prevent phase separation of inorganic salt solutions and precipitation of nucleating agents [99]. Additionally, a gel-like network structure can be formed to support the MWCNTs suspended in the salt solution. 23 wt% MgCl₂ solution has a supercooling degree of 16.562 °C and the addition of 0.5 wt% XG did not have any effect, with a supercooling degree of 16.292 °C. When 1, 2, and 3 wt% of MWCNTs were added, the supercooling degree was reduced to 10.269, 13.414, and 11.436 °C. The large specific surface area of MWCNTs provided more nucleation sites for the PCM, reduced the potential barrier of nucleation, and promoted the non-uniform nucleation process. On the other hand, 1 wt% of CaCl₂, 0.25 wt% Ca(OH)₂, and 0.5 wt% XG resulted in a supercooling degree of 7.726 °C. The lowest supercooling degree was achieved at 2.167 °C when the nucleating agents and 1 wt% MWCNTs were added together and this indicated that they have a synergistic effect in promoting nucleation.

4. Modified carbon nanotubes

Due to the large surface area, high aspect ratio, and strong van der Waals interactions among CNTs, they have a high tendency to aggregate and therefore difficult to disperse in organic and inorganic solvents [100]. To overcome this problem, modifications to the surface of CNTs can be carried out to facilitate their dispersion in the target medium. The commonly used methods for surface modification

of CNTs that serve to alter their surface energy include covalent and non-covalent modification [101]. In covalent modification, functional groups such as carboxyl, hydroxyl, and hydrocarbyl can be introduced to the surface of CNTs and the most adopted form of covalent modification is oxidation. The ratio of sulfuric acid to nitric acid determines the degree of oxidation [102], whereby sulfuric acid affects the surface morphology and nitric acid attaches the functional groups to the surface. Even though covalent modification improves the chemical compatibility of CNTs with PCMs, the use of acids at high temperatures might result in structural defects in CNTs [103]. In this regard, non-covalent modifications have the advantage of introducing different groups on the surface of CNTs without disturbing the π - π system of the CNT layers. In this method, surfactants or polymers are usually physically adsorbed or wrapped onto the surface of CNTs [104].

4.1. Modified single-walled carbon nanotubes

To improve the uniformity of CNTs in PEG and enhance the overall thermal conductivity of the as-prepared SSPCMs, Yang et al. made use of hydroxylated CNTs (HO-CNTs) and added γ -glycidyl ether oxypropyl trimethoxysilane (KH-560) as a cross-linking agent [105]. Due to the good water solubility and reactive groups of KH-560, only deionized water was used and no organic solvents were involved in the synthesis of the SSPCMs. The HO-CNTs were successfully grafted onto PEG and no aggregation was observed as they were attached to the molecular structure of PEG. SEM images also revealed that the HO-CNTs were uniformly distributed and an interconnected network was formed, resulting in an enhancement of thermal conductivity of 2.2 times for 9.0 wt% HO-CNTs compared to PEG. This work presented a novel green synthesis method to prepare SSPCMs using CNTs and has good potential for temperature regulation in TES applications. To overcome the leakage issue and low heat harvesting and releasing rate of stearyl alcohol (SAL), Göksu et al. prepared SSPCMs by crosslinking polyacrylic acid (PAA) with SAL and thermally enhanced the PCMs with acid-treated SWCNTs (A-SWCNTs) [106]. SWCNTs were modified with concentrated sulfuric acid/nitric acid, which facilitates the grafting with PAA with its carboxyl groups via hydrogen bonding. The A-SWCNTs/PAA/SAL composites with mass ratios of 1:1:2, 1:3:4, and 1:5:6 were named NCPCM1, NCPCM2, and NCPCM3, respectively. The addition of the A-SWCNTs not only prevented the leakage of SAL but also increased the thermal degradation temperatures of the composites due to the enhanced activated SWCNTs-PAA structure (Figure 9a). Furthermore, NCPCM1 has significantly enhanced heat harvesting and releasing performance, with a reduction in heat harvesting and releasing time of 51.96% and 44.26%, respectively compared to SAL (Figure 9b). Through the self-assembly of 1D SWCNTs and two-dimensional boron nitride nanosheets (BNNSs), Wu et al. constructed CPCMs with a 3D bulk structure with PEG uniformly dispersed in the network [107]. To achieve this, SWCNTs were modified using nitric acid while hexagonal boron nitride was modified using sodium lignosulfonate before being peeled off into nanosheets. Meanwhile, PEG was reacted with 4,4'-diephenylmethane diisocyanate to introduce isocyanate groups at the end of the prepolymer, which facilitated the self-assembly with SWCNTs and

BNNs via hydrogen bonding (Figure 9c). With PEG dispersed homogeneously into a 3D structure, a continuous thermal conductive network can be formed while eliminating the issue of aggregation. The synergistic assembly between the highly thermal conductive SWCNTs and BNNs not only enhances the thermal conductivity by 178.8% but also provides excellent structural stability with no leakage of PEG. Furthermore, the light-thermal conversion efficiency of the CPCMs increased from 45.24 to 97.15%, indicating that this method can be used to achieve good thermal performances and light-to-thermal conversion. On the other hand, Du et al. introduced amino-functionalized SWCNTs (SWCNTs-NH₂) into PEG-based polyurethane PCM to develop organic PCMs with good shape stability and photothermal conversion efficiency as well [108]. Nitric acid oxidation and amidation treatment were carried out to introduce amino groups onto the surface of SWCNTs, which enhanced the dispersibility and reactivity of SWCNTs. The results indicated that the introduction of SWCNTs-NH₂ does not affect the degree of crystallinity and phase change enthalpy of the CPCMs. Additionally, the thermal conductivity was significantly enhanced and the solar-thermal conversion and storage efficiency was improved from 44.8 to 89.3%.

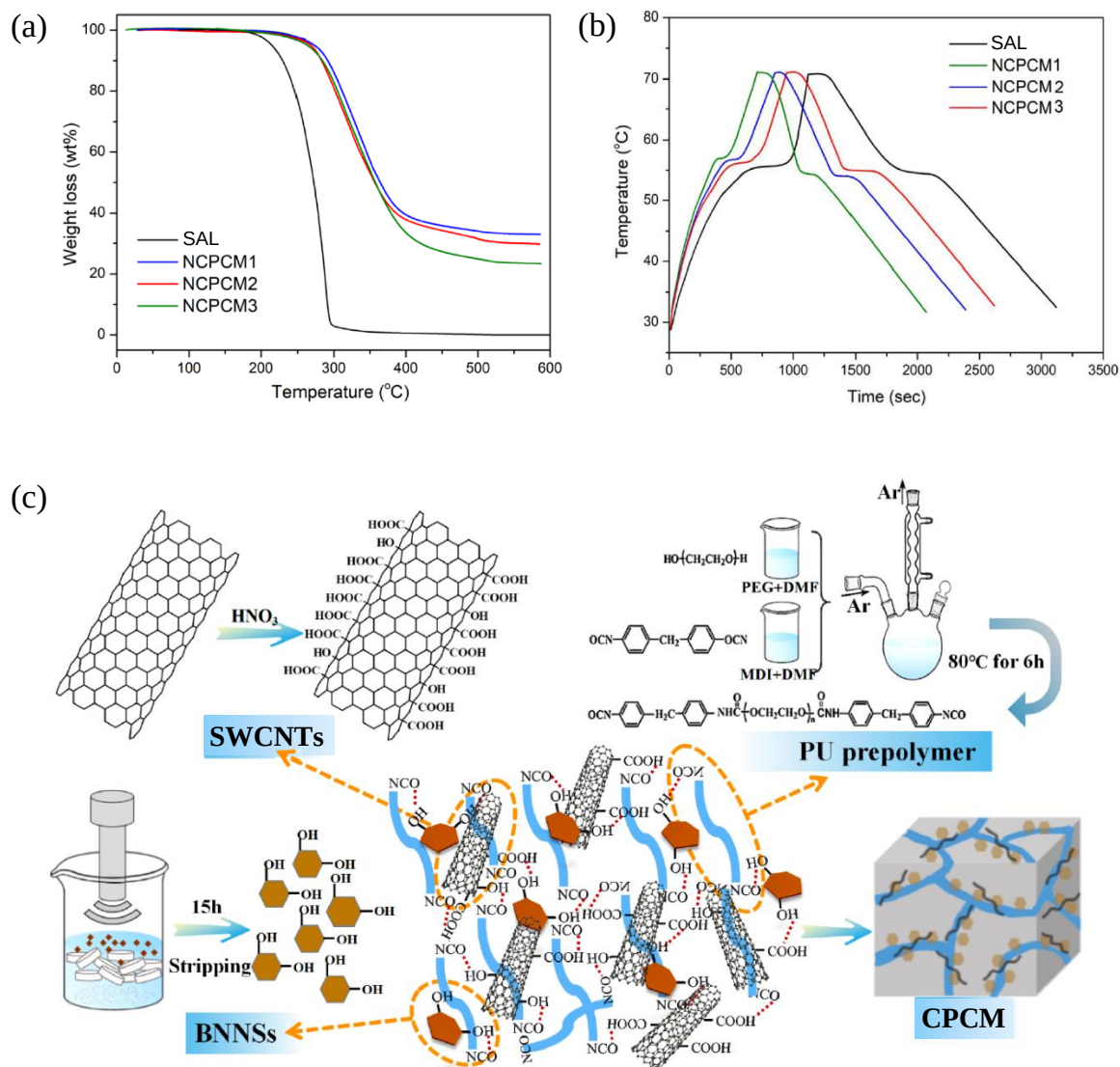


Fig. 9. (a) TGA curves of SAL, NCPCM1, NCPCM2, and NCPCM3, and (b) Heat harvesting and releasing performance of SAL, NCPCM1, NCPCM2, and NCPCM3. Reproduced with permission. [106] Copyright 2022 Elsevier. (c) Schematic illustration for the fabrication mechanism of the novel CPCMs. Reproduced with permission. [107] Copyright 2022 Elsevier.

Encapsulation is a widely used technique that can be applied in different applications [109-112]. The encapsulation of PCMs within a core-shell structure offers both shape stability and leakage prevention. Nevertheless, the drawback of this approach is the typically poor mechanical strength, low thermal conductivity, and thermal stability associated with the core-shell. Furthermore, there is poor compatibility and density difference between CNTs and most organic PCMs. When PCMs undergo a solid-liquid phase transition, their viscosity decreases and the CNTs in the core of the microencapsulated PCM will settle down at the bottom and agglomerate. To overcome these issues, Meng et al. added octadecyl isocyanate-grafted CNTs (OICNTs), where a *n*-octadecane/*n*-octacosane

(C18/C28) mixture served as the core and melamine-formaldehyde (MF) resin as the shell [113]. The SEM images revealed that the OICNTs were uniformly distributed both in the core and on the surface of the shell, providing numerous thermal conductive paths for heat transfer. As a result, the thermal conductivity of the microencapsulated PCM with 3 wt% OICNTs increased by 71.4% compared to that without OICNTs, and a relatively high phase change enthalpy of 187.9 J/g was obtained as well.

4.2. Modified multi-walled carbon nanotubes

Zhao et al. prepared a eutectic PCM made up of decyl alcohol and lauryl alcohol and added hydroxylated MWCNTs to develop a low-temperature CPCM [114]. Sodium dodecyl benzene sulfonate was added as a dispersant as well to further improve the dispersibility of the MWCNTs, in addition to other commonly used techniques such as applying an external magnetic field, ultrasonic waves, and magnetic stirring [115-117]. Different mass ratios of the eutectic PCM were first investigated and it was found that decyl alcohol/lauryl alcohol with a mass ratio of 76:24 has the lowest freezing point of -2.0 °C and no supercooling. This mass ratio was later used to investigate different mass ratios of hydroxylated MWCNTs. The results indicated that the thermal conductivity was enhanced by 15.8% with 0.15 wt% hydroxylated MWCNTs compared to pure eutectic PCM, with only a slight reduction in phase change temperature and enthalpy. Nevertheless, the addition of the hydroxylated MWCNTs resulted in supercooling as the MWCNTs tend to agglomerate due to their weak attraction. This reduced the nucleation matrix and caused the solution to nucleate on the agglomerated substrate. Liu et al. prepared a FSPCM by using a polymer gel as the supporting material and investigated the effects of added hydroxylated MWCNTs [118]. Glauber's salt (GS) was used as the PCM while poly (acrylate sodium) copolymer (PAAS) served as a supporting material. PAAS also functioned as a thickening agent and it can increase the viscosity of the solution and prevent phase separation of GS. Nevertheless, once again striking a balance between thermal conductivity and viscosity is crucial for the optimization of heat transfer performance in PCMs. Due to the abundance of hydrophilic hydroxyl groups on the surface of the MWCNTs, they are well embedded into the hydrate salt hydrogel matrix (Figure 10a). The interaction between GS and PAAS resulted in a higher thermal stability and the introduction of MWCNTs further enhanced this stability due to the formation of more hydrogen bonds between GS and MWCNTs (Figure 10b). Additionally, this stronger hydrogen bond network increased the phase change temperature of the GS/PAAS/MWCNTs composite compared to GS/PAAS as well. Based on the results revealed in this work, the thermal storage properties and stability of such composites could be adjusted and possibly controlled by carrying out surface functionalization on different nanomaterials containing different hydrophilic groups. On the other hand, Zhang et al. introduced HO-CNTs into microencapsulated PCMs prepared via *in situ* polymerization, with *n*-hexadecanol as the core and MF resin as the shell [119]. SEM images revealed that the HO-CNTs do not affect the morphology of the microcapsules and some of these are found to extend out of the core and adhere to the shell (Figure 10c). These observations were confirmed by the transmission electron microscopy (TEM) images

(Figures 10d and e), indicating that they are well-distributed in the microcapsules. This distribution of the HO-CNTs, coupled with their high aspect ratio and high number of -OH groups on the surface helps to reduce the interfacial thermal resistance by establishing chemical bonds or polar interactions with the shell. This effect is evident in the thermal conductivity results, where 0.6 wt% of HO-CNTs increased the thermal conductivity from 0.3002 to 0.3598 W/(m·K). It was also noted that when the HO-CNTs are less than 0.2 wt%, the latent heat of the microencapsulated PCMs remains stable at around 180 J/g and it decreases after exceeding this amount. Nevertheless, the latent heat and energy storage efficiency still has a relatively high value of 155.6 J/g and 64.51%, respectively.

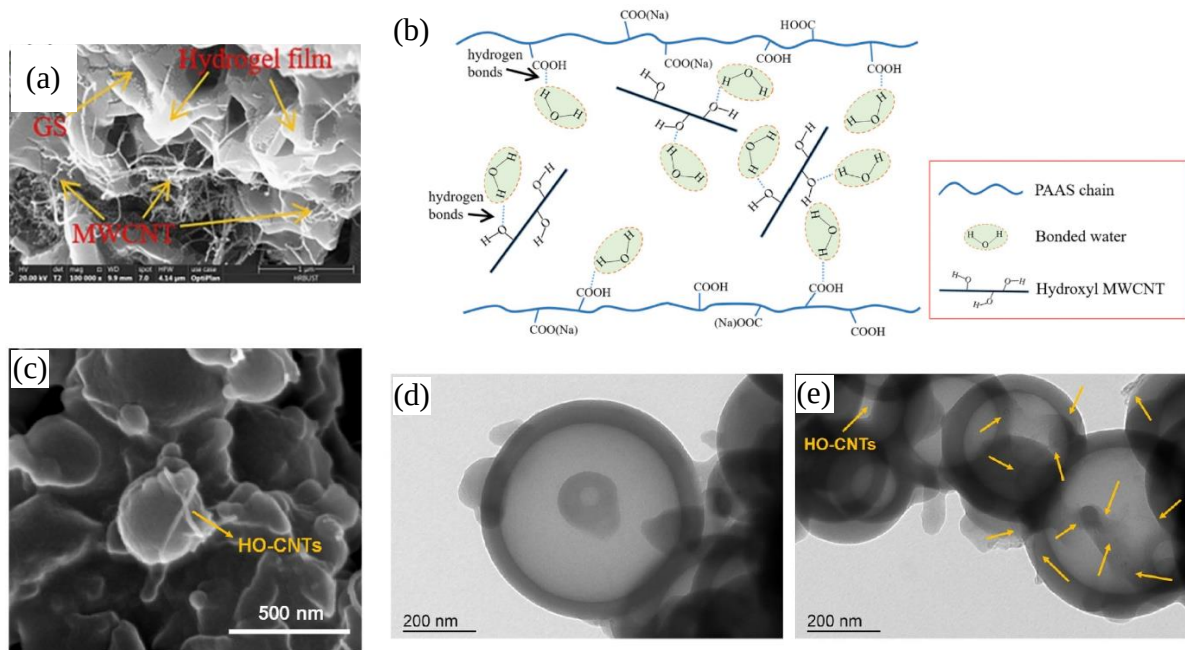


Fig. 10. (a) SEM images of MWCNTs-GS/PAAS composite, and (b) Schematic illustration of hydrogen bonds formed by the interaction of bonded water in GS with PAAS and hydroxyl MWCNTs, respectively. Reproduced with permission. [118] Copyright 2020 Elsevier. (c) SEM images of microencapsulated PCMs with 0.6 wt% HO-CNTs, TEM images of (d) Microencapsulated PCMs without HO-CNTs, and (e) Microencapsulated PCMs with 0.6 wt% HO-CNTs. Reproduced with permission. [119] Copyright 2022 Elsevier.

Besides hydroxylated MWCNTs, carboxylated MWCNTs have been used by researchers in the field of TES as well. Li et al. prepared SSPCMs with tailored phase transitions by integrating SA and hydroxylated MWCNTs [120]. To achieve this, a hydroxylated MWCNTs/ethanol suspension was mixed with an SA/ethanol solution and the mixture was first sonicated for 30 min, followed by stirring at 70 °C for 4 h. Thereafter, an impregnation process was carried out at 80 °C for 24 h. The results

indicated reversible phase transitions from high melting temperatures (74 °C) for SA to room temperature (~30 °C) for the SSPCMs, due to a strong interface confinement effect. Furthermore, the phase transitions could be driven by sunlight for TES and thus can potentially be used for solar applications. Using carbonized bamboo parenchyma cells (CPC) as supporting materials, Zheng et al. prepared SSPCMs using PEG and added oxidized MWCNTs to improve their dispersibility and interfacial affinity with CPC [121]. Biomass carbon materials such as bamboo parenchyma cells are highly porous and contain hydroxyl groups on their surface, making them conducive for the encapsulation of PEG. Furthermore, they have good mechanical properties and can enhance the thermal conductivity and photo-thermal conversion efficiency of CPCMs. The carboxylated MWCNTs and CPC are bound by a myriad of forces such as hydrogen bonds, capillary force, surface tension, and van der Waals force, with the former forming a continuous and dense network on the surface of the CPC. Consequently, the CPCM with 3 wt% carboxylated MWCNTs increased the thermal conductivity by 128% compared to PEG. Additionally, the photo-thermal conversion efficiency was enhanced from 81.3 to 87.2%. Compared to the other samples, the CPCM with 3 wt% carboxylated MWCNTs has a higher surface (Figure 11a) and internal (Figure 11b) temperature after 1200s of heating, indicative of the fastest heat transfer. This result was supported by the UV-vis-NIR analysis (Figure 11c), where the CPCM with 3 wt% carboxylated MWCNTs has the highest absorbance. Conversely, Yan et al. synthesized CPCMs using PEG, silicon dioxide network grafted aminopropyl group, and carboxylated MWCNTs via the sol-gel method [122]. SEM images showed that PEG was distributed and adsorbed into the mesoporous structure of the gel, resulting from the hydrogen bond interactions with the amino and silicon hydroxyl groups of the functionalized silica gel. Meanwhile, the carboxylated MWCNTs were well-dispersed in PEG, leading to significant enhancements in thermal conductivity (43.5 to 108.7%) of the CPCMs compared to PEG. Moreover, the CPCMs possessed high light-to-heat conversion and storage efficiency (85.8 to 88.3%) and the degree of supercooling decreased as well.

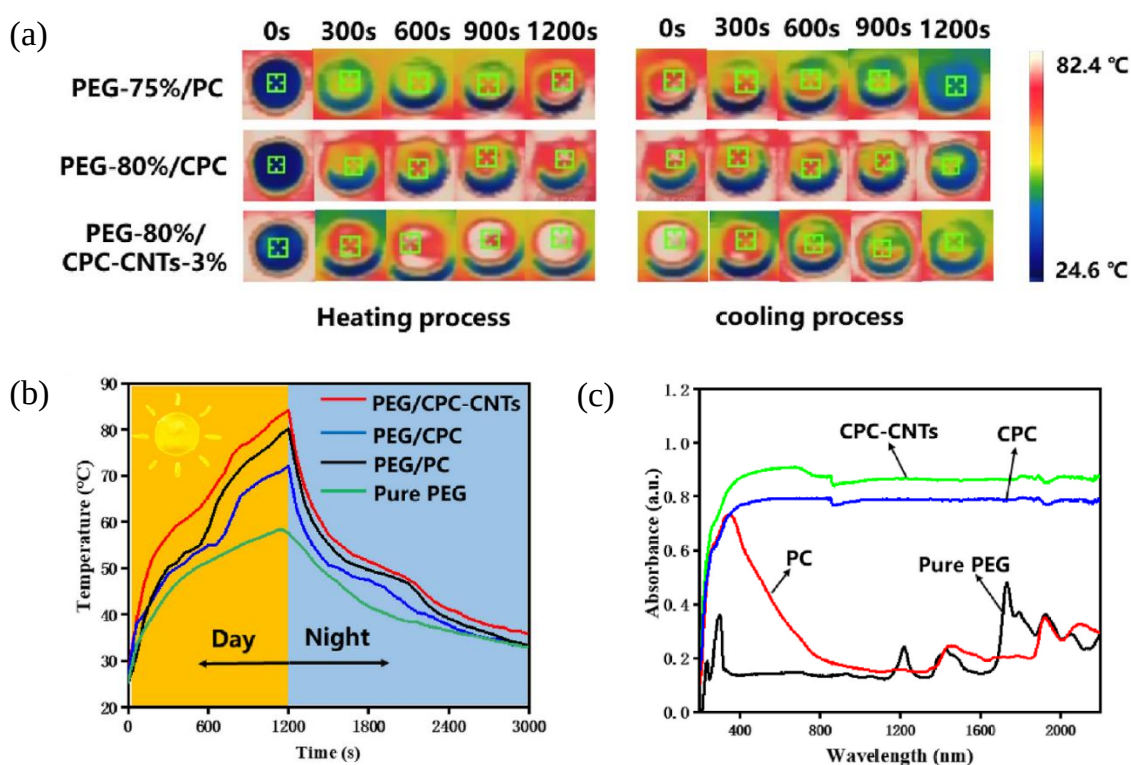


Fig. 11. (a) Infrared thermography photos showing the surface temperature variation of different samples at the same heating and cooling times, (b) Temperature variation plots of PEG, PEG/PC, PEG/CPC, and PEG/CPC-CNTs with or without simulated solar illumination, and (c) UV-vis-NIR patterns of pure PEG, PC, CPC, and CPC-CNTs. Reproduced with permission. [121] Copyright 2022 Elsevier.

Besides the commonly used acid treatment to introduce oxygen-containing functional groups such as hydroxyl and carboxyl onto the surface of MWCNTs, other unconventional methods for surface modification of MWCNTs could be carried out as well. In some of these cases especially, they are tailored to meet specific application requirements. Metal-organic frameworks (MOFs) are a class of crystalline materials constituted by inorganic metal ions and organic linkers. They have extremely high surface areas with 3D porous structures and are therefore widely used in a range of different applications such as drug delivery and heterogeneous catalysis [123, 124]. Even though they have low thermal conductivity, their high surface area, high pore volume, and tunable nature make them a viable alternative supporting material for the preparation of SSPCMs. Wang et al. developed a highly thermal conductive supporting material with an interpenetrating network structure by coating the surface of MWCNTs with amino-functionalized MOFs nanoparticles (Cr-MIL-101-NH₂) [125]. The TEM images showed that the shell structures of Cr-MIL-101-NH₂ were homogeneously distributed on the surfaces of MWCNTs. With the porous nature of the MOF, PEG can be impregnated into it and the leakage of PEG can be prevented due to surface tension force and capillary action. Additionally, PEG is stabilized

within the pores via hydrogen bonding with the amino groups of the MOF. Furthermore, the close integration between the MOF nanoparticles and MWCNTs bestows higher mean free paths and continuous heat transfer paths for photon transmission. With the addition of 5.16 wt% MWCNTs, the thermal conductivity of the CPCMs was enhanced by 100.9% compared to the composite without MWCNTs. This work sheds light on improving the heat transfer performance of SSPCMs through the integration of porous materials such as MOFs on the surface of highly conductive nano additives. Wang et al. on the other hand, functionalized MWCNTs using sodium dodecyl sulfate (SDS) and dispersed them in PEG with different mass ratios (0.5, 1, 2, and 5 wt%) to investigate the form-stability and thermal behavior of the CPCMs [126]. Due to their hydrophobic nature, the MWCNTs could not be dispersed well in PEG solution even in low mass fraction loadings. Their functionalization with SDS, however easily overcame this issue and resulted in a stable dispersion. As the mass ratios of the MWCNTs increased, the melting temperature and phase change enthalpy decreased gradually from 63.9 to 61.9 °C and 165 to 150 J/g, respectively. The thermal conductivity of the CPCMs increased from 0.295 to 0.531 W/(m·K) with the addition of 0 to 5 wt% MWCNTs while excellent form stability was achieved for the CPCMs with 2 wt% MWCNTs as well. For microencapsulated PCMs to potentially be used for building applications, they need to have excellent temperature regulation and flame retardant capabilities as organic PCMs are inherently flammable [127]. To develop a microencapsulated PCM that can meet both criteria, Niu et al. grafted 9,10-dihydro-9-oxa-10 phosphaphenanthrene-10-oxide (DOPO) onto the surface of MWCNTs and synthesized the microencapsulated PCM via *in situ* polymerization, with CA as the core and poly(methyl methacrylate) (PMMA) as the shell [128]. Due to the synergistic effect of MWCNTs and DOPO, the thermal stability of the microcapsules was enhanced and it was noted that the flame retardant MWCNTs did not affect the encapsulation efficiency. The microcapsules were added to rigid polyurethane foam to determine their temperature regulation and flame retardancy performance and the results showed that DOPO and MWCNTs can increase carbon residue and effectively inhibit heat release during foam combustion. Additionally, smaller fluctuation in indoor temperature was attained as the modified MWCNTs can improve the heat control ability of the microcapsules.

There are many different ways to modify the surface of MWCNTs and different functionalization methods can result in MWCNTs with varied properties such as stability, dispersibility, and thermal conductivity. Therefore, comparing the different functionalization methods of MWCNTs will give us a better understanding of their advantages and limitations and how we can develop CPCMs with these tailored materials for specific applications. Shen et al. employed three different methods, namely acid treatment, mechanochemical process, and ball milling to modify MWCNTs, and the influence of the different MWCNTs on ER-based CPCMs was investigated [129]. From the TEM images, the walls of pristine MWCNTs are smooth and complete while ball-milled MWCNTs (B-MWCNTs) and mechanochemical-treated MWCNTs (BB-MWCNTs) have similar outcomes and were not affected by

the mechanical treatment (Figures 12a-c). However, the acid-treated MWCNTs (A-MWCNTs) were partially destructed as several broken spots (circled in red) were seen along the tube walls (Figure 12d), implying the harshness of this method. Nonetheless, A-MWCNTs displayed superiority over B-MWCNTs and BB-MWCNTs in the dispersion stability test (Figures 12e-l). After 6 h, the pristine MWCNTs completely precipitated while B-MWCNTs and BB-MWCNTs yielded significant precipitation as well. A-MWCNTs showed no signs of precipitation as they can form strong hydrogen bonds with ER. A largely similar study conducted by Putra et al. to investigate the form stability of beeswax-based CPCM also revealed that A-MWCNTs can form strong bonds with beeswax with their functional groups. They concluded that the ball milling process does not alter the chemical structure of MWCNTs and instead only increases their surface area [130]. On the other hand, Li et al. modified MWCNTs with acid treatment, silicon coating, and ethylene grafting, respectively, and compared their dispersion stability with paraffin [131]. The silicon-coated MWCNTs were prepared via the sol-gel process (Figure 12m) while the ethylene-grafted MWCNTs were prepared via the reaction of acylated MWCNTs and diallyl amine (Figure 12n). The results showed that the silicon-coated MWCNTs have better dispersibility compared to A-MWCNTs as the silicon coating can decrease the van der Waals force between the MWCNTs while the ethylene-grafted MWCNTs have the best dispersibility due to the good compatibility between paraffin and the carbon chains of the MWCNTs. The thermal conductivity of paraffin was enhanced by 42.3, 84.6, and 83.5% with the addition of unmodified MWCNTs, silicon-coated MWCNTs, and ethylene-grafted MWCNTs, respectively. The greater enhancements for the modified MWCNTs arise due to the lower interface thermal resistance and higher heat transfer efficiency. To try to understand the stability of MWCNTs in PCMs better, Ranjbar et al. studied the effect of PCM polarity on the stability by using PCMs with three different polarity – nonpolar (PW), partially polar (SA), and polar (PEG) [132]. Additionally, the MWCNTs were modified using both SA and hexadecyl amine. It was found that the polarity of the PCM and MWCNTs are the dominant factors in determining the stability as compared to factors like using a surfactant or surface modification. Pristine MWCNTs have better stability with nonpolar PCMs like PW and SA due to their nonpolar-nonpolar interaction while modified MWCNTs displayed higher stability with PEG arising from their polar-polar composition.

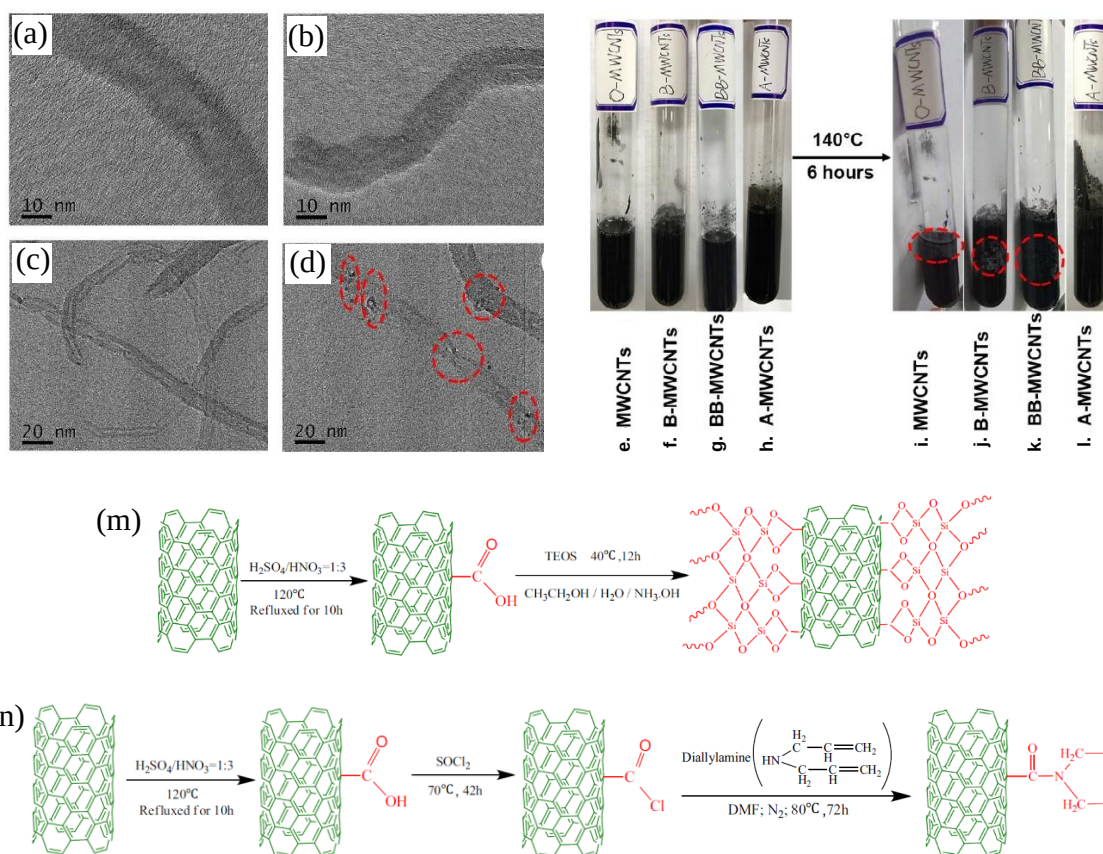


Fig. 12. TEM images of (a) Pristine MWCNTs, MWCNTs that undergo (b) ball milling process, (c) mechanochemical process, (d) Acid treatment, and (e-l) Digital photos showing the dispersion stability of MWCNTs in ER. Reproduced with permission. [129] Copyright 2018 Elsevier. (m) Schematic diagram of the silicon-coated MWCNTs, and (n) Schematic diagram of the ethylene-grafted MWCNTs. Reproduced with permission. [131] Copyright 2022 Elsevier.

5. PCMs-encapsulated carbon nanotubes

Nanomaterials have unique optical, electrical, and reactive properties that differ from bulk materials [133-137]. Unlike bulk materials, materials that are encapsulated in nanoscaled structures exhibit different behaviors, such as improved resistance to environmental and decomposition degradation and confined phase transitions [138, 139]. As mentioned previously, PCMs can be encapsulated in micro or nanocapsules to prevent their leakage but the shell of microencapsulated PCMs is usually made of polyurethane or urea-formaldehyde resin, both of which have low thermal conductivity [140, 141]. Besides the high thermal conductivity of CNTs, another important feature that they possess is their hollow cylindrical structure. By leveraging this characteristic, various molecules or particles such as organics, water, and metals can be used to fill up the hollow structure [142-146], whereby the confined molecules are bound by van der Waals interactions within the CNT wall [147, 148]. Various

encapsulation methods have been reported, such as capillary action, arc discharge, wet chemical techniques, and chemical vapor deposition [149-152]. Nevertheless, highly pure CNTs are required for such a process to ensure that there are no internal blockages.

Wu et al. used a combination of experiments and MD to gain insight into encapsulated PCM properties from a microcosmic perspective, where the nano-encapsulated PCM was formed by the vacuum infiltration of lauric acid (LA) into CNT [153]. MD simulation was carried out by constructing an MD model of the nano-confined PCM, whereby the structure of LA in CNT, end-to-end distance, and torsion angle were studied. TEM images showed the open ends of CNTs and the presence of LA, indicating the successful filling of LA in the CNT with a degree of filling of 10.02%. Simulation studies indicated that the molecular distribution of LA in a freestanding environment has a chaotic structure as opposed to a circular and orderly arrangement in the nano-confined CNT due to their restricted torsion and the possible van der Waals interaction with CNT. This highly ordered structure and nano-confined effect enhanced the heat and mass transfer of the system and improved the thermal conductivity by 20 times compared to LA. The distance between the wall of CNT and LA is determined to be 4.8 Å and this is in the range of van der Waals force, thus preventing the outflow of LA from the inner space of CNT during phase transition. Nie et al. conducted a similar study on the physical encapsulation of paraffin in CNT via the vacuum infiltration method as well [154]. TEM images revealed that the tips of the CNTs were removed after acid treatment and that the molten paraffin was infiltrated into the CNTs. Similar observations of paraffin molecules with an orderly orientation along the CNT walls were observed, with a decreasing degree of order as the temperature increased. The degree of filling of paraffin in the CNT is 26.27%, which is twice as high as what Wu et al. have achieved. However, these results still leave much to be desired in terms of achieving a high filling ratio of the CNT. Using a slightly different approach, Feng et al. were able to improve the filling ratio even further [155]. The authors used a vacuum-based infiltration-purification method to fill LA into MWCNTs, where deionized water was added to the mixture and subjected to high pressure and temperature after the vacuum infiltration process. The highest filling ratio of the LA/MWCNTs composites was 54.8%, which is considerably higher compared to the aforementioned works. To determine the relationship between the structural distribution of LA confined in CNTs and the phase change properties, MD simulations were conducted and it was noted that LA molecules tend to form a liquid layer near the CNT walls and crystallization only took place for the LA molecules dispersed in the central region. When the filling ratio was reduced to 15.5%, no phase change occurred and it was postulated that the LA molecules attached to the inner walls of the CNT do not undergo crystallization. Composites with high CNT loading and enhanced thermal conductivity show great potential as SSPCMs for TES. The above studies offer valuable insights into the phase change behavior of organic PCMs within confined spaces, which is crucial for future research on complex PCMs incorporated into porous materials. On the other hand, Jeong et al. are cognizant of the fact that having highly pure CNTs without any internal blockages is crucial for the

encapsulation of nonmetallic materials such as paraffin and polymers as they can open up new doors of opportunity for developments in nanotechnology and materials science. Therefore, the authors fabricated ultrapure CNTs with high crystallinity to encapsulate PW with a high filling yield [156]. The pure CNTs were prepared in a reactor maintained at 1 Pa, with temperatures between 1400-1800 °C for 5 h under H₂ atmosphere. The vacuum-based infiltration-purification process for the infiltration of CNTs is shown in Figures 13a-d. TEM and DSC analysis confirmed that paraffin was efficiently filled in the CNT with a high yield. Nevertheless, the synthesis procedure for the CNTs is slightly complex and hence might not be practical for real-life applications.

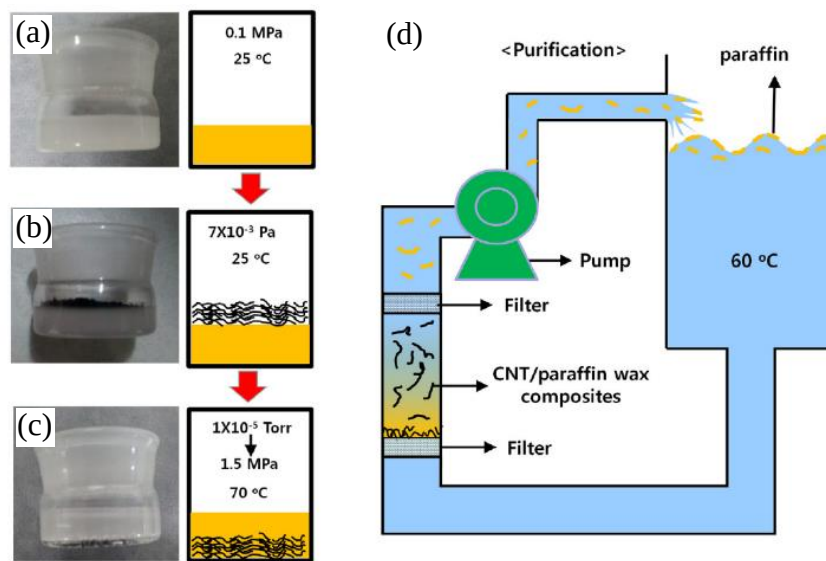


Fig. 13. (a-c) Vacuum-based infiltration process to fill the internal spaces of the CNTs with paraffin wax, and (d) purification process of the paraffin wax/CNTs composite. Reproduced with permission. [156] Copyright 2013 Elsevier.

Sinha-Ray et al. intercalated two different types of paraffin (W1 and W2) into CNTs using self-sustained diffusion and this method allows the tailoring of the working temperature range of the encapsulated PCMs [157]. The working temperature range of paraffins W1 and W2 are 35-52 °C and 36-62 °C, respectively and the intercalated blend of both PCMs in a 1:1 ratio resulted in a wider range from 35-62 °C, indicating that the working temperature range of the intercalated PCMs blend can be expanded. Furthermore, this method can be scaled up easily for mass production. However, the latent heat of the PCMs is relatively small at 26.52 J/g and the filling ratio was not determined. Therefore, further optimizations and studies should be carried out before consideration for wide use. Abbaspour et al. investigated the thermal properties of two different types of paraffins (octadecane and icosane) as well using simulation studies by varying different parameters such as CNT length, diameter, and

paraffin length [158]. According to the results, the encapsulation of octadecane increased the melting temperature, which concurred with the findings by Akbarzadeh et al. [159]. Moreover, a longer CNT resulted in a decrease in the melting temperature of the encapsulated PCM system due to a larger accessible volume for the confined octadecane molecules while a larger CNT diameter and longer icosane molecule resulted in a higher diffusion coefficient.

Apart from using encapsulated CNT to enhance the TES capability, Li et al. explored the flame-retardant ability by encapsulating SAL with phosphorus oxychloride (POCl_3) in SWCNTs [160]. SAL was modified with POCl_3 to reduce its flammability and SWCNTs were used to improve the thermal conductivity and leakage issue of the formed flame-retardant FSPCM film. The modified SAL exhibits greater fire retardancy as compared to pure SAL as it resulted in a reduction of 24.4% for the peak of the heat release rate and 24.3% for the total heat release. Encapsulation of the modified SAL lowered these values even further due to the synergistic effect between the SWCNTs and the phosphorus group of POCl_3 . The phosphorus groups promoted char formation to reduce the contact of the material with the heat source, and the dense SWCNTs layer has a self-carbonization effect that stops the transfer of volatile products into the matrix. Thermal conductivity and electrical properties were also significantly improved as well. These data suggest that the interaction of both phosphorus-containing groups and SWCNTs could be used to expand the application of organic PCMs in flame-retardant materials. A summary of previously reported studies related to the incorporation of PCMs with different types of CNTs is presented in Table 1 below.

Table 1. Summary of thermal properties of PCMs with different types of CNTs.

Phase Change Composite	Methodology/ conditions	Loading of CNTs (wt%)	Melting Temperature (°C)	Melting Enthalpy (J/g)	Thermal Conductivity ((W/(m·K))	Enhancement in Thermal Conductivity (%)	Reduction in supercooling (%)	Light-to-thermal conversion efficiency (%)	Application	Ref.
Single-walled carbon nanotubes										
Paraffin	Melt impregnation/ 60°C for 1 h	-	46.8	247.0	0.23	-	-	-	-	[52]
Paraffin/ SWCNTs		1	46.7	239.6	0.29	26.1	-	-	-	
		2	46.6	237.2	0.36	56.5	-	-	-	
Paraffin/metal foam/SWCNTs		-	-	177.4	1.83	695.7	-	-	-	
		1	-	170.6	2.05	791.3	-	-	-	
		2	-	168.7	2.22	865.2	-	-	-	
Paraffin	Direct mixing/60°C for 2 h	-	46 ~ 63	251.0	0.19	-	-	-	Micro-channel heat sink	[55]
Paraffin/ SWCNTs		5	45 ~ 59	232.0	0.39	109.7	-	-		
		10	42 ~ 58	208.0	0.62	234.6	-	-		
		15	42 ~ 52	193.0	0.78	323.2	-	-		
Paraffin	Dispersant and ultrasonic oscillation/25°C for 75 min	-	-	151.8	0.35 (s), 0.37 (l)	-	-	-	Vertical open refrigerated display cabinet	[56]
Paraffin/ SWCNTs		1		146.3	0.41 (s), 0.41 (l)	16.6 (s), 12.0 (l)	-	-		
		3		137.0	0.46 (s), 0.47 (l)	30.3 (s), 28.6 (l)	-	-		
		5		128.2	0.49 (s), 0.52 (l)	39.5 (s), 40.3 (l)	-	-		
NaCl	MD simulation	-	947.3	422.2	-	-	-	-	CSP systems	[57]
NaCl/SWCNTs		3.5	823.8	338.4		8.8	-	-		
		6.7	805.5	268.6		28.6	-	-		
KNO ₃	-MD simulation -Direct mixing+ ultrasonic oscillation/60°C for 1 h	-	608.3	113.8	-	-	-	-	CSP systems	[58]
KNO ₃ /SWCNTs		4.43	607.9	111.9		2.3	-	-		
		8.48	607.8	100.3		28.0	-	-		
Eutectic salt	Solution evaporation/3 h	-	486.4	331.5	-	-	-	-	-	[59]
Eutectic salt/SWCNTs		1.0	485.1	307.7		42.3	-	-		
		1.5	484.2	300.3		57.0	-	-		

		2.5	484.2	293.9		47.0				
PEG	Melt-mixing/ 80°C for 50 min	-	59.1	183.4	0.24 (s)	-	-	-	Building	[64]
PEG/SWCNTs		6	56.0	172.4	-	-	32.8			
		8	55.8	169.4	-	-	34.4			
		10	55.3	165.4	3.43 (s)	1329.2 (s), 533 (l)	38.8			
PEG	-Physical blending/ 70°C for 2 h	-	53.3	176.7	0.25	-	-	-	Building	[65]
PEG/expanded vermiculite/ SWCNTs	-Impregnation/ 70°C for 4 h	3.30	42.8	25.6	0.40	58.6				
		5.20	45.1	45.1	0.43	70.9				
		7.09	48.0	83.9	0.51	103.0				
CA	Melt- impregnation/ 50°C for 10 min	-	30.9	163.4	0.15	-	-	-	Building	[66]
CA/Dt		-	31.0	87.6	0.31	106.7				
CA/Dt/SWCNTs		3	-	-	0.41	173.3				
		5	-	-	0.46	206.7				
		7	31.4	79.1	0.48	220.0				
SA	-Carbonation/ 1000°C for 6 h	-	72.5	-	0.34	-	-	-	Waste heat utilization	[67]
SA/SWCNTs	-Wet impregnation/ 90°C for 1 h	25	70.7	155.7	1.02	200.0				
PTA	Electrospinning/ 15 cm at 10 kV	-	28.6	108.0	-	-	-	-	Greenhouse film	[68]
PTA/PET		-	28.8	37.0	0.03	-				
PTA/PET/ SWCNTs		0.5	30.4	47.0	-	-				
		1.0	29.0	41.0	-	-				
		2.0	29.9	38.0	0.04	48.3				
PEG	Impregnation/30 min	-	59.1	183.4	0.22	-	-	-	Solar	[69]
PEG/Dt		-	58.9	94.4	0.29	31.8	-	-		
PEG/DCPs		-	57.7	87.7	0.66	200.0	-	77.3		
PEG/DCNs		3.2	56.8	107.4	1.52	590.9	25.8	87.7		
Nextek 24D	Mechanical stirring/2400 rpm for 40 min	-	25.6	198.0	0.17	-	-	-	-	[161]
Nextek 24D/SWCNTs/ unsaturated polyester resin		0.005	25.6	19.6	0.20	17.6				

Multi-walled carbon nanotubes										
PW/CFM/MWCNTs	Direct mixing	0.2	59.0	154.42	0.2	-	-	-	Photovoltaic panel	[72]
Eutectic PCM	Direct mixing+sonication/1 h	-	5.1	104.1	-	-	-	-	Cold storage	[73]
Eutectic PCM/MWCNTs		0.06	5.1	96.0		24.4				
		0.08	5.1	92.4		47.5				
		0.10	5.1	90.7		67.3				
PW	-Direct mixing/20°C for 1 h	-	44.3	177.5	0.20	-	-	-	Building, domestic hot water, and heating systems	[75]
PW/expanded perlite	-Vacuum impregnation/80°C for 1 h	-	44.2	103.9	0.11	-47.5				
PW/expanded perlite/MWCNTs		1.83	44.3	100.3	0.21	4.4				
		3.62	45.4	97.7	0.38	83.8				
		5.27	43.7	96.0	0.52	152.9				
PEG	Vacuum impregnation/90 min at 65 kPa	-	-	-	0.22	-	-	-	Building	[76]
PEG/Dt		-	7.9	51.4	0.15	-				
PEG/Dt/MWCNTs		0.57	7.3	53.8	0.26	73.3				
		1.70	6.9	55.9	0.27	80.0				
		2.50	7.2	62.9	0.29	93.3				
PW	-Physical blending/70°C for 2 h	-	54.1	140.0	0.23	-	-	-	-	[77]
PW/polypropylene hollow fiber membrane/MWCNTs	-Injection/70°C	0.47	53.6	107.4	0.37	60.9				
		0.71	53.6	107.6	0.41	78.3				
		0.93	53.6	103.3	0.46	100.0				
PW/HDPE	Melt-mixing	-	56.0, 122.0	60.5, 63.0	-	-	-	-	-	[87]
PW/HDPE/MWCNTs		1.5	58.0, 124.0	59.6, 68.0	-	-				
		2	-	-	0.39	-				
		3	-	-	-	29.0				
PW	Vacuum impregnation/100°C for 3 h	-	-	172.5	0.11	-	-	-	-	[88]
PW/copper foam				-	1.69	1436.4				
PW/diamond-coated copper foam				-	2.74	2390.9				

PW/copper foam-CNT				-	3.43	3018.2				
PW/diamond-coated copper foam				-	9.72	8736.4				
PEG	Vacuum impregnation/80°C for 30 min	-	63.2	154.8	0.38	-	-	-	Solar	[89]
GA			62.7	150.2	-	81.6		53.2		
MWCNTs/GA			-	-	-	144.7		-		
B-GA			62.1	148.4	-	181.6		89.3		
PW	-Ultrasonic/15 min	-	51.4	178.0	-	-	-	-	Solar	[90]
PW/MWCNTs/EG	-Mechanical stirring/80-90°C	2	49.3	159.0	2.05					
	-Vacuum/20 min									
ER	Vacuum impregnation/150°C for 1 h	-	-	334.4	0.66	-	-	-	Solar	[93]
ER/GF		0		184.4	56.93	8525.8	36.6	-		
		2		124.4	-	-	-	-		
		4		103.6	27.18	4018.2	49.6	85.4		
Deionized water	Ultrasonication/1 h	-	1.6	302.9	-	-	-	-	Building	[97]
Deionized water/MWCNTs		0.25	7.1	325.5		-	-			
		0.50	9.5	338.8		-	-			
		0.75	10.8	349.7		23.0 (s), 11.2 (l)	94.0			
MgCl ₂	-Mechanical stirring/2 h	-	-	153.3	-	-	-	-	Cold storage	[98]
MgCl ₂ /XG/	-Ultrasonic oscillation/30 min	0.5	-35.5	146.7	-	-	-	-		
Ca(OH) ₂ /		1	-36.5	146.9	0.53	-	86.9			
CaCl ₂ /MWCNTs	-Mechanical stirring/2 h	2	-36.1	142.3	-	-	-			
		3	-35.6	139.8	0.56	10.7	-			
N-octadecane	-Direct mixing/50°C for 40 min	-	30.9	265.7	0.48	-	-	-	-	[162]
N-octadecane/octadecanol/MWCNTs	- Ultrasonic oscillation/1 h	0.1	27.6	35.2	-	-0.91 (s), -0.84 (l)	36.4			
PEG	Direct mixing/60°C for 3 h	-	62.7	158.5	0.25	-	-	-	Building, furniture,	[163]

Reactive PEG/MWCNTs		1.0 1.3 2.0	63.5 64.0 64.5	131.8 124.2 120.9	0.36 - 0.44	44.0 - 76.0				wearable devices	
Sodium acetate trihydrate (SAT)/sodium monohydrogen phosphate dodecahydrate (DSP)	Direct mixing/80°C	-	-	263.5	0.46	-	-	-	-	-	[164]
SAT/DSP/MWCNTs		2 4 6 8		258.0 253.9 249.0 244.0	0.66 0.87 1.08 1.29	43.5 89.1 134.8 180.4					
PEG PEG/MWCNTs	-Direct mixing/70°C for 1 h -Ultrasonic oscillation/1.5 h	- 0.5 1.0 2.0	66.2 69.7 67.4 72.9	190.4 161.0 150.4 136.1	0.20 0.23 0.28 0.31	- 15.0 40.0 55.0	-	-	-	Electronics	[165]
Phase Change Composite	Methodology/ conditions	Type of CNT	Loading of CNT (wt%)	Melting Temperature (°C)	Melting Enthalpy (J/g)	Thermal Conductivity ((W/(m·K))	Enhancement in Thermal Conductivity (%)	Reduction in supercooling (%)	Light-to-thermal conversion efficiency (%)	Application	Ref.
Modified carbon nanotubes											
PEG PEG/HO-CNTs	Chemical grafting/626°C for 5 h, 20 h	Hydroxylated SWCNTs	- 5.0 7.0 9.0	58.9 57.8 57.9 57.2	185.3 127.9 127.0 123.7	0.32 0.62 0.67 0.71	- 90.8 109.4 119.8	- 30.0 8.4 7.9	-	-	[105]
SAL SAL/PAA/a-SWCNTs	Direct mixing/24°C for 30 min, 100°C for 3 h, 100°C for 24 h	Hydroxylated SWCNTs	- 25 12.5 8.3	54.9 54.9 55.2 54.8	271.2 134.6 134.7 134.4	-	-	-	-	Electronics, automobile modules, food carriers, solar PV panels	[106]

PEG PEG/SWCNTs/ BNNSs	Self- assembly/24 h	Hydroxylated SWCNTs	- 0 4 8	62.1 60.4 60.4 57.5	182.5 149.3 149.1 126.6	0.33 0.52 0.75 0.95	- 57.6 127.3 187.9	-	45.2 79.3 86.5 97.2	Solar	[107]
PEG PEG/ SWCNTs-NH ₂	Polymerization/ 80°C for 4 h, 40°C for 2 h	Amino- functionalized SWCNTs	- 0.5 2 4	65.9 55.7 54.6 53.9	179.1 125.8 124.7 125.1	- 0.29 0.37 0.42	- 11.1 42.7 60.7	- 0.9 9.7 13.4	44.8 67.5 81.7 89.3	Solar	[108]
C18 C28 C18/C28 C18/C28/MF/ OICNT	<i>In-situ</i> polymerization/ 1.5 h	Octadecyl isocyanate- grafted SWCNTs	- - - 0 1 2 3	29.5 62.3 29.1, 52.8 28.6, 51.9 29.2, 51.9 28.9, 51.9 28.7, 51.9	240.2 258.6 124.7, 128.5 100.2, 99.7 101.6, 94.9 95.8, 96.6 89.9, 98.0	- - - 0.19 0.26 0.29 0.33	- - - - 34.9 50.0 71.4	-	-	Electronics, air conditioning systems	[113]
Decyl alcohol Lauryl alcohol Decyl alcohol/ lauryl alcohol Decyl alcohol/ lauryl alcohol/ MWCNTs	Ultrasonic oscillation/45°C for 1 h	Hydroxylated MWCNTs	- - - 0.15	6.0 23.8 -3.2 -3.1	205.2 179.1 179.7 178.4	- - 0.29 0.33	- - - 15.8	-	-	-	[114]
GS GS/PAAS GS/PAAS/ MWCNTs	Direct mixing/60°C for 2 h	Hydroxylated MWCNTs	- - 3	34.7 36.7 38.5	235.7 202.8 187.2	0.57 0.74 1.38	- 30.0 141.7	-	-	Solar	[118]
<i>n</i> -hexadecanol <i>n</i> -hexadecanol/ MF <i>n</i> -hexadecanol/ MF/HO-CNTs	<i>In-situ</i> polymerization/ 70°C for 3 h	Hydroxylated MWCNTs	- - 0.2 0.4 0.6	51.2 44.0, 51.2 43.8, 51.5 44.0, 52.1 42.5, 51.1	243.1 182.5 183.4 169.2 157.3	- 0.30 0.32 0.34 0.36	- - - - -	-	-	Solar	[119]

SA SA/MWCNTs	-Direct mixing/ 70°C for 4 h -Impregnation/ 80°C for 24 h	Carboxylated MWCNTs	- 3	74.0 29.0	140.9 111.8	-	-	-	-	Solar	[120]
PEG PEG/CPC PEG/CPC/ MWCNTs	Vacuum impregnation/ 80°C for 1 h	Carboxylated MWCNTs	- - 1 3 5	65.0 62.9 64.0 64.1 63.7	187.5 145.3 148.2 149.4 149.1	0.22 0.37 0.47 0.49 0.50	- 69.6 118.0 127.6 128.6	-	- 81.3 - 87.2 -	Building	[121]
PEG PEG/MWCNTs	Sol-gel/60°C for 70 min	Carboxylated MWCNTs	- 1 2 3	61.4 59.0 58.5 59.5	212.8 129.3 125.2 116.3	0.23 0.39 0.44 0.48	- 69.6 91.3 108.7	-	- 86.6 87.5 88.3	Solar	[122]
PEG Cr-MIL-101-NH ₂ PEG/ Cr-MIL-101-NH ₂ PEG/CNT@ Cr-MIL-101-NH ₂	Physical blending+ impregnation/ 4 h	MOFs- functionalized MWCNTs	- - - 5.16	54.8 - 50.6 52.4	172.2 - 83.1 96.2	0.30 0.20 0.23 0.46	- -32.6 -22.5 55.7	-	-	-	[125]
PEG PEG/SDS/ MWCNTs	Solution blending/80°C for 1 h	SDS- functionalized MWCNTs	- 1 2 5	63.9 62.9 62.8 61.9	165.0 165.3 156.6 150.0	0.30 0.43 0.45 0.53	- 45.4 52.9 80.0	-	-	-	[126]
CA CA/PMMA/ MWCNTs	Direct mixing	DOPO- functionalized MWCNTs	-	32.6 33.5	155.5 103.2	-	-	-	-	Building	[128]
Beeswax Beeswax/ MWCNTs	-Direct mixing/70°C for 4 h - Impregnation/80 °C for 24 h	Hydroxylated MWCNTs	- 5	60.6 60.2	153.2 115.5	0.25 0.46	- 84.0	-	-	-	[130]
Palmitic acid Palmitic acid/Ag- MWCNTs/	Vacuum impregnation/80 °C for 12 h	Ag-MWCNTs	-	65.2 65.5	188.0 155.9	0.22 0.65	- 202.3	-	-	Electronics	[166]

MXene											
Paraffin Paraffin/SiO ₂ - MWCNTs	Direct mixing/1h + 50°C for 3 h	SiO ₂ - MWCNTs	- 3	27.8 26.6	99.3 95.4	0.18 0.30	- 63.7	-	-	-	[167]
PW PW/MWCNTs	Direct mixing/180°C for 3.5 h	CuS- MWCNTs	- 4.1 6	61.1 60.4 60.9	173.1 147.1 145.8	-	- 13.1 28.2	-	-	Solar	[168]
Phase Change Composite	Methodology/ conditions	Degree of filling (%)	Melting Temperature (°C)	Melting Enthalpy (J/g)	Thermal Conductivity (W/(m·K))	Enhancement in Thermal Conductivity (%)	Light-to- thermal conversion efficiency (%)	Application	Ref.		
PCMs-encapsulated carbon nanotubes											
LA LA/CNT	-Direct mixing/80°C for 20 min -MD simulation	- 10.0	43.4 39.7	198.2 16.8	0.20 3.91	1855.0	-	-		[153]	
PW PW/CNT	-Direct mixing/80°C for 20 min -MD simulation	- 26.3	54.0 49.0	123.9 20.1	-	-	-	Micro- electronics		[154]	
LA LA/CNT (5- 10 nm)	-Direct mixing/20 min -MD simulation	- 54.8 33.1 15.5	43.1 37.8 34.5	175.0 54.6 22.1	0.28 - -	- - -	-	-		[155]	
LA/CNT (2-5 nm)		41.0	32.0	22.6	1.00	257.1					
PW PW/SWCNTs	Vacuum-based infiltration- purification/ 70°C 1.5 MPa for 5 min	-	55.1 51.3	144.8 67.5	-	-	-	Drug delivery		[156]	
PW (W1) PW (W1)/CNT PW (W2)	Direct mixing+ ultrasonic oscillation/30 min	-	47.2 44.2 56.7	140.9 10.5 147.5	-	-	-	Solar, electronics		[157]	

PW (W2)/CNT			52.6	17.7					
PW (W1+W2)/ CNT			46.9	26.5					
SAL	MD simulation	-	60.6	227.5	0.50	-	-	-	[160]
Modified SAL			80.0	191.3	0.18	-64.0			
SAL/SWCNTs			66.1	121.9	2.51	402.0			
			69.0	139.0	1.99	298.0			
			71.2	167.9	1.69	238.0			

6. Conclusion and outlook

In conclusion, this review has explored the recent advancements in leveraging CNTs to enhance the thermal properties of PCMs. The amalgamation of CNTs with PCMs offers a multitude of advantages that significantly enhance their thermal properties and overall performance. Firstly, CNTs possess exceptional thermal conductivity, which when integrated into PCMs, facilitate rapid and efficient heat transfer, thereby improving the overall thermal conductivity of the composite material. Moreover, the high aspect ratio and large surface area of CNTs enable enhanced interfacial interactions with PCM molecules, promoting better dispersion and compatibility within the composite matrix. Additionally, modified CNTs, functionalized with different chemical groups or compounds, can further tailor the properties of the CNT-PCM composite, such as improving dispersion, stability, and compatibility with specific PCM matrices. These modifications can also impart additional functionalities, such as enhanced thermal stability, mechanical strength, and even tunable phase transition temperatures, making them suitable for a broader range of applications. Other than experimental methods, simulation studies are an invaluable tool for investigating CNT-PCM composites as they allow researchers to explore various PCM-CNT combinations and optimize their properties without the need for expensive materials and extensive laboratory equipment. They also provide a detailed understanding of how CNTs interact with PCMs at the molecular level, offering insights that are difficult to achieve through experiments alone. The incorporation of both CNTs and modified CNTs into PCMs holds tremendous potential for various applications, including TES, thermal management systems, and temperature regulation in diverse industries.

Despite their many advantages, the incorporation of CNTs and modified CNTs into PCMs presents several limitations and challenges that need to be addressed.

- Achieving uniform dispersion of CNTs within the PCM matrix is a significant challenge. Agglomeration of CNTs can lead to sedimentation and this will result in phase separation, ineffective utilization of their beneficial characteristics, and reduced material efficiency over time. To mitigate sedimentation, surfactants or dispersants can be added to enhance the wettability of the additives and stabilize the dispersion. Encapsulating CNTs along with PCMs inside microcapsules can also ensure a uniform distribution of the CNTs within the PCMs. Increasing the viscosity of the PCM matrix by incorporating thickeners or gelling agents can also slow down the sedimentation.
- Producing CNTs in large quantities and at low cost remains a challenge, which affects the scalability and commercial viability of CNT-enhanced PCMs. To lower the cost of producing CNTs for large-scale applications, several strategies can be used. For example, instead of using catalysts like iron, nickel or cobalt for CNTs production, alternative catalysts with lower costs can be explored. Chemical vapor deposition is one of the most widely used methods to produce CNTs and therefore optimizing

the reactor design and enhancing the throughput can lower costs. In addition, implementing renewable energy sources like solar or wind in the production plants can also result in lower energy costs.

- Although CNTs enhance thermal conductivity, their incorporation can sometimes reduce the latent heat storage capacity of the PCM as they do not contribute to the phase change process. Hence, the overall PCM content may be reduced, leading to a decreased amount of latent heat stored per unit weight or volume. Therefore, considerations must be given to balance improved thermal conductivity and maintaining adequate latent heat capacity based on the intended application.
- The repeated solid-liquid phase transitions in PCMs can cause mechanical fatigue and degradation of the material, thus affecting their cycling stability. Furthermore, if CNTs are not well integrated, they may exacerbate this effect, causing cracks or voids over multiple thermal cycles. Several strategies can be employed to enhance cycling stability. Besides the aforementioned introduction of stabilizing additives or encapsulation techniques to prevent sedimentation that will affect thermal cycling performance, CNTs-enhanced PCMs can be embedded in porous structures such as silica aerogels or metal foams to enhance the mechanical stability and prevent structural degradation over time. The polymer chains of organic PCMs can be cross-linked to create a more rigid structure that helps to trap CNTs in place, thus reducing the risk of material separation during thermal cycling. By implementing a combination of these strategies, the cycling stability of CNTs-enhanced PCMs can be significantly improved.

Looking forward, there are promising opportunities to overcome these challenges and unlock the full potential of CNT-enhanced PCMs. Future research efforts should focus on developing scalable and cost-effective production methods for CNTs, refining dispersion techniques, and exploring synergistic combinations with other nanomaterials or additives. Furthermore, advancements in sustainability and recyclability will be essential to promote the widespread adoption of CNT-enhanced PCMs across various industries. One key development is the sustainable sourcing and production of CNTs. Researchers are exploring bio-based CNTs derived from renewable feedstocks or waste materials to reduce dependence on fossil fuels. Green synthesis methods such as chemical vapor deposition processes powered by renewable energy, are being refined to minimize harmful by-products. Additionally, efforts are underway to create PCM matrices that can be recycled alongside the embedded CNTs. Sustainable alternatives to traditional materials, such as bio-based PCMs like fatty acids, are gaining traction. Efficient methods for recovering CNTs from PCM matrices, including mechanical and chemical separation, enable their reuse without compromising their thermal performance, enhancing the eco-friendliness of the materials throughout their lifecycle.

Abbreviations

1D One-dimensional

3D	Three-dimensional
A-MWCNTs	Acid-treated MWCNTs
A-SWCNTs	Acid-treated SWCNTs
B-MWCNTs	Ball-milled MWCNTs
BB-MWCNTs	Mechanochemical-treated MWCNTs
BNNSs	Boron nitride nanosheets
CA	Capric acid
CFM	Copper foam matrix
CNTs	Carbon nanotubes
CPC	Carbonized bamboo parenchyma cells
CPCMs	Composite PCMs
CSP	Concentrating solar power
CSs	Carbon spheres
Dt	Diatomite
EG	Expanded graphite
EP	Expanded perlite
ER	Erythritol
FSPCMs	Form-stable PCMs
GA	Graphene oxide aerogel
GF	Graphite foam
GS	Glauber's salt
HDPE	High-density polypropylene
HO-CNTs	Hydroxylated CNTs
LA	Lauric acid
MD	Molecular dynamic
MF	Melamine-formaldehyde
MOFs	Metal-organic frameworks
MWCNTs	Multi-walled CNTs
OICNTs	Octadecyl isocyanate-grafted CNTs
PAA	Polyacrylic acid
PAAS	Poly (acrylate sodium) copolymer
PC	Porous carbon
PEG	Polyethylene glycol
PET	Poly(ethylene terephthalate)

PCMs	Phase change materials
PMMA	Poly(methyl methacrylate)
POCl ₃	Phosphorus oxychloride
PTA	Poly(tetradecyl acrylate)
PV	Photovoltaic
PW	Paraffin wax
SA	Stearic acid
SAL	Stearyl alcohol
SDS	Sodium dodecyl sulfate
SEM	Scanning electron microscopy
SSPCMs	Shape-stabilized PCMs
SWCNTs	Single-walled CNTs
SWCNTs-NH ₂	Amino-functionalized SWCNTs
TEM	Transmission electron microscopy
TES	Thermal energy storage
XG	Xanthan gum

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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