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One-pot synthesis of carbon nanotube /graphene composite phase change materials based on Pickering emulsion for solar thermal storage

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Abstract

The practical deployment of phase change materials (PCMs) is frequently constrained by two intrinsic drawbacks. respectively low thermal conductivity and susceptibility to leakage during solid–liquid phase change. To address these issues, this study reports the synthesis of composite PCMs comprising hexadecanol encapsulated within a three-dimensional carbon nanotube (CNT)/reduced graphene oxide (RGO) network, fabricated via a one-pot Pickering emulsion method using graphene oxide (GO) as the stabilizer. The resulting CNT/RGO scaffold concurrently enhances thermal transport and structural integrity. Specifically, the thermal conductivity of the composite reaches 125.40% of that of pristine hexadecanol, while the latent heat capacity is substantially retained. Furthermore, the composite exhibits improved photothermal conversion efficiency. These characteristics render the developed material a promising candidate for solar thermal storage applications .

Highlight

- A one-pot Pickering emulsion strategy enables simultaneous construction of 3D CNT/RGO network and encapsulation of hexadecanol.
- CNT/RGO framework provide anti-leakage confinement and thermal conduction enhancement.
- Synergistic effect of CNT and RGO boosts thermal conductivity and improves photothermal conversion.
- The one-pot approach simplifies preparation while ensuring uniform dispersion.

Key words: Graphene oxide; Phase change materials; Pickering emulsion; Thermal properties

1.Introduction

The escalating intensity of global energy shortages and associated environmental pressures has intensified research interest in renewable energy sources. Latent heat thermal energy storage (LHTES) based on phase change materials (PCMs) stands out for its capacity to store vast thermal energy within a narrow temperature range near the material's phase change point ^[1]. Phase change materials can absorb substantial thermal energy during the phase change process and release it through the reverse transformation under specific conditions. These materials are capable of storing significant thermal energy within a relatively compact volume ^[2], and are widely applied in thermal regulation systems for solar and wind energy storage ^[3], temperature control in electronic devices ^[4], and building energy efficiency ^[5], among other fields. Therefore, it is essential to investigate composite phase change materials with enhanced thermal transfer and energy storage performance. Although organic PCMs are abundant in variety and offer advantages such as non-toxicity, non-corrosiveness, and excellent chemical stability ^[6], their low thermal conductivity and susceptibility to leakage hinder their practical application ^[7].

To address PCM limitations like low thermal conductivity and leakage during phase change, 3D matrices such as foam ^[8] or aerogel ^[9] can encapsulate them. Graphene, valued for its high thermal conductivity, chemical stability, and mechanical strength ^[10], is increasingly used to enhance PCM composites ^[11]. Jiang et al.^[12] encapsulated hexadecylamine (HDA) into graphene aerogel (GA) via impregnation method. The load capacity of GA for HDA is as high as 5600%, and the resulting composite achieved a thermal conductivity of 0.238 W/(m·K), corresponding to a 1.62 times relative to that of pure HDA. After 50 repeated heating–cooling cycles, the latent heat of the composite phase change material decreased by only 6.5%, indicating excellent thermal stability.

While graphene addition solves leakage issues, extends service life ^[13], and partially improves thermal conductivity, the conductivity remains insufficient for practical applications ^[14]. To further enhance it, researchers often incorporate nanofillers with high thermal conductivity into three-dimensional graphene frameworks ^[15], such as boron nitride (BN) ^[16], silver nanoparticles (Ag NPs) ^[17], and carbon nanotubes (CNTs) ^[18].

Chen et al. ^[19] created a 3D porous network by crosslinking carbon nanotubes (CNT) with reduced graphene oxide (rGO). The thermal conductivity of composite phase change material achieved 1.27 W/(m·K) (552% of pure PW), 90.3% photothermal conversion efficiency, high phase-change enthalpy, and excellent stability. Rajamony et al. ^[20] incorporated binary carbon nanofillers

(graphene/MWCNT) into salt hydrates, increasing thermal conductivity by 160% ($1.2 \text{ W/m}\cdot\text{K}$) versus pure salt hydrates. The composite also enhanced optical absorption (reducing transmittance by 0.07%) while maintaining high heat storage capacity and thermal/chemical stability. He et al. [21] synthesized elastic hybrid aerogels from graphene oxide, carboxylated CNT, and polyvinyl alcohol. These exhibited 169.6 J/g phase-change enthalpy, 88.9% solar-thermal efficiency, and $0.640 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ thermal conductivity. Leveraging the aerogel's thermal buffering, they designed a double-layer structure (DPCG@PEG) enabling infrared stealth for high-temperature equipment.

Combining encapsulation materials with high-thermal-conductivity nanofillers enhances both stability and thermal conductivity in composite phase change materials. These nanofillers can additionally impart photothermal conversion or magnetic properties [22]. However, current 3D matrix preparation methods—primarily sol-gel [23] and vacuum freeze-drying [24,25]. Compared with other methods, the Pickering emulsion method has significant advantages. This method is an innovative preparation technology that uses solid particles as emulsifiers. These particles can form stable films at the oil-water interface, thereby preparing stable emulsions [26]. This method is simple in steps and effectively solves the dispersion problem of graphene oxide in the polymer matrix [27].

This study successfully fabricated a carbon nanotube/graphene-based composite phase change material via a one-step Pickering emulsion approach. This method offers a straightforward, efficient, and controllable route for synthesizing such composite PCMs. The three-dimensional framework structure of graphene prevents the leakage problem of hexadecanol (HD) during the phase transition process. Carboxylated multi-walled carbon nanotubes with high thermal conductivity and mechanical properties are introduced as thermal conductive fillers to further enhance the thermal conductivity, thermal stability and photothermal conversion performance of the material. The carbon nanotube/graphene composite phase change material developed in this work exhibits excellent heat storage capacity and stability. Meanwhile, certain photothermal conversion performance broadens the practical application of the composite material.

2 Materials and Methods

2.1 Materials

Graphene oxide powder (GO) required for raw material preparation was purchased from XFNANO Materials Technology Co., Ltd. (XFNAN) in Jiangsu. Graphitized carboxylated multi-walled carbon nanotubes (MWCNTs-COOH) were supplied by Shanghai Macklin Biochemical Technology Co., Ltd. Hexadecanol (HD) was supplied by Sinopharm Chemical Reagent Co., Ltd., ascorbic acid (VC) was obtained from Tianjin Damao Chemical Reagent Co., Ltd, anhydrous ethanol was purchased from Tianjin Fuyu Fine Chemical Co., Ltd., and hydrochloric acid was purchased from

Tianjin Chemical Reagent Factory.

All purchased reagents were of analytical purity and could be used without further purification.

2.2 Preparation of CNT/GO Composite Phase Change Materials

Initially, an appropriate amount of graphene oxide (GO) powder was introduced into deionized water, followed by ultrasonic treatment for 30 min to produce a 2 mg/mL aqueous GO dispersion. Then, different amount of carboxylated multi-walled carbon nanotubes (MWCNTs-COOH) was added to the GO aqueous dispersion (with the mass ratios of carbon nanotubes being 0 wt.%, 5 wt.%, 10 wt.%, and 20 wt.%, respectively) and then sonicated for 60 min.

Hexadecanol was first heated to 80 °C to achieve complete melting. The molten hexadecanol was then combined with carboxyl-functionalized multi-walled carbon nanotubes (MWCNTs-COOH) and graphene oxide (GO) under acidic conditions (pH = 2), and the mixture was subjected to vigorous mixing using an IKA Vortex device at 2500 rpm for 5 min to form a stable Pickering emulsion. Ascorbic acid (VC) was introduced into the emulsion and dispersed by stirring at 500 rpm for an additional 5 min.

Finally, the carbon nanotube/graphene composite phase change material emulsion was placed in a hydrothermal reactor, and the temperature was set at 95 °C for a reaction duration of 12 hours. The carbon nanotube/graphene composite phase change material was one-pot synthesized. The material was then dried in a vacuum drying oven.

For convenience, PCM0, PCM1, PCM2, and PCM3 represent the composite PCMs with carbon nanotube mass fractions of 0 wt.%, 0.3 wt.%, 0.6 wt.%, and 1.2 wt.% of, respectively.

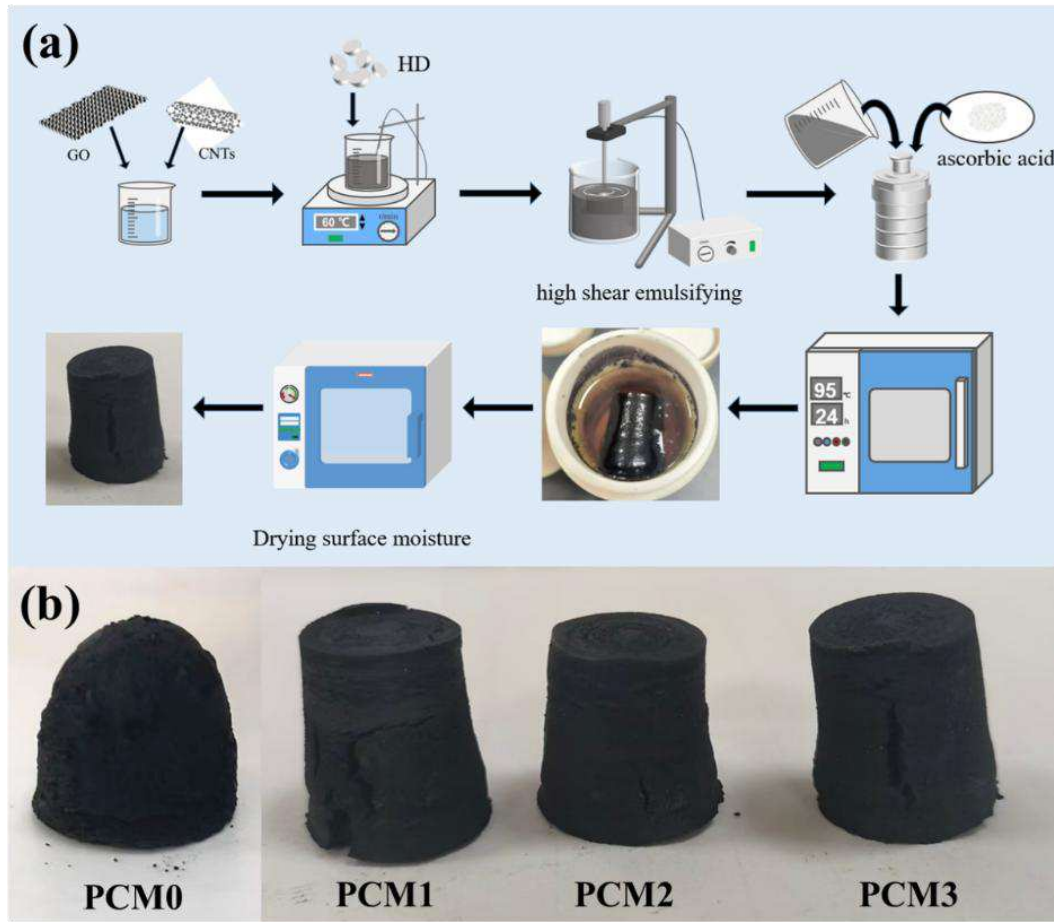


Fig.1 (a) Preparation process diagram of composite phase change materials;(b)Dried image of carbon nanotube/graphene composite shaped phase change material;

2.3 Characterization

The microstructure of the composites were characterized by SEM (TESCAN MIRA4, Czech Republic). FTIR spectroscopy (Smartlab, Rigaku, Japan) was employed to identify characteristic peaks and analyze whether chemical reactions occurred during the material preparation process. The thermal energy storage properties of the materials were investigated by DSC (TA DSC25, USA). The thermal stability of the prepared materials were measured using a thermogravimetric analyzer (TA TGA55, USA) under N_2 atmosphere, while the shape stability of the composites was evaluated through a heating platform. The thermal conductivity of the samples was analyzed with a thermal conductivity analyzer (Hot Disk TPS2500S, Sweden). The photothermal conversion performance of the carbon nanotube/graphene oxide composite phase change material was examined using a thermal imager (HM-TPK20-3AQF/W) and a simulated light source.

3 Results and Discussion

3.1 Characterization of carbon nanotubes, graphene oxide and lotion

Figure 2(a) presents the Fourier transform infrared (FT-IR) spectra of both carbon nanotubes (CNTs) and graphene oxide (GO). The GO spectrum exhibits several characteristic bands: a broad absorption centered at 3399 cm^{-1} , assignable to O–H stretching vibrations; distinct peaks at 1750 cm^{-1} and 1610 cm^{-1} , corresponding to C=O and C=C stretching modes, respectively, with the latter likely originating from the sp^2 -hybridized carbon domains within the graphene structure; and additional bands at 1225 cm^{-1} and 1040 cm^{-1} , attributed to C–O–C and C–O stretching vibrations, respectively. These features collectively confirm the presence of hydroxyl, carboxyl, and epoxide functional groups on the GO surface. In the CNT spectrum, a corresponding O–H stretching absorption appears at 3411 cm^{-1} . Additionally, weaker characteristic peaks are observed at 1628 cm^{-1} and 1118 cm^{-1} relative to those in GO. These results demonstrate that both the carbon nanotubes and graphene oxide contain the above-mentioned functional groups.

Figure 2(b) presents an optical microscopy image of the carbon nanotube/graphene composite phase change material emulsion. The image reveals distinct spherical droplet structures. Indicated that the graphene, carbon nanotubes, and hexadecanol retain their structural integrity, and are uniformly dispersed within the system.

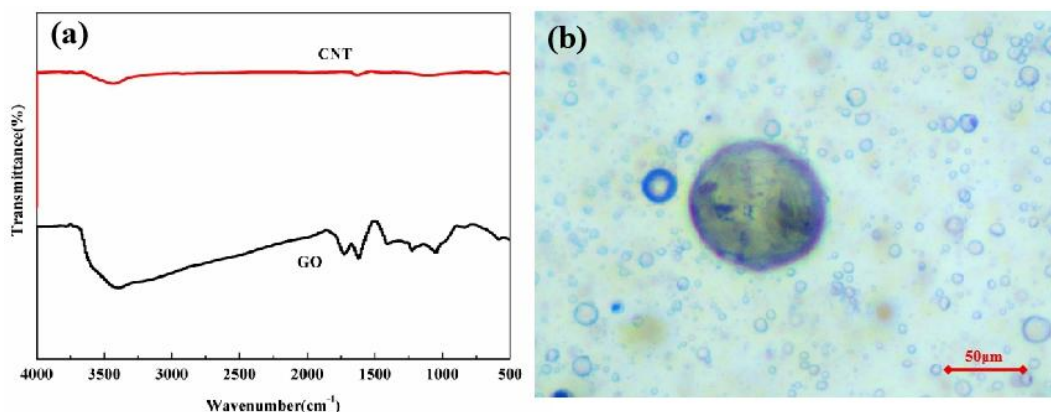


Fig.2 (a) FT-IR spectra of carbon nanotubes and graphene oxide;(b) Optical microscope image of graphene composite phase change material emulison

3.2 Structure of Composite Phase Change Materials

The composite phase change material was soaked in anhydrous ethanol and washed to remove hexadecanol, which was done to better observe the three-dimensional structure of the resulting carbon nanotubes/graphene (Figure 4-4). As the figure shows, a distinct, uniformly porous structure is visible under the electron microscope. The absence of obvious agglomeration between the graphene layers

indicates that a structurally uniform three-dimensional architecture was constructed during the hydrothermal reduction process. The surface-active properties of GO facilitate its dispersion with carbon nanotubes, allowing them to adhere to the GO layers. This interaction then drives the co-coating of GO and carbon nanotubes onto the hexadecanol surface during emulsification. Subsequently, the GO undergoes a reduction reaction, thereby constructing the three-dimensional structure. Furthermore, the carbon nanotubes adhering to the GO nanosheets help build an enhanced thermal conduction network, while their capillary and surface tension enhance the adsorption capacity of graphene for hexadecanol. By effectively preventing hexadecanol leakage during the solid-liquid phase change, this three-dimensional structure can prolong the composite's service life and reduce the corrosion of thermal storage devices.

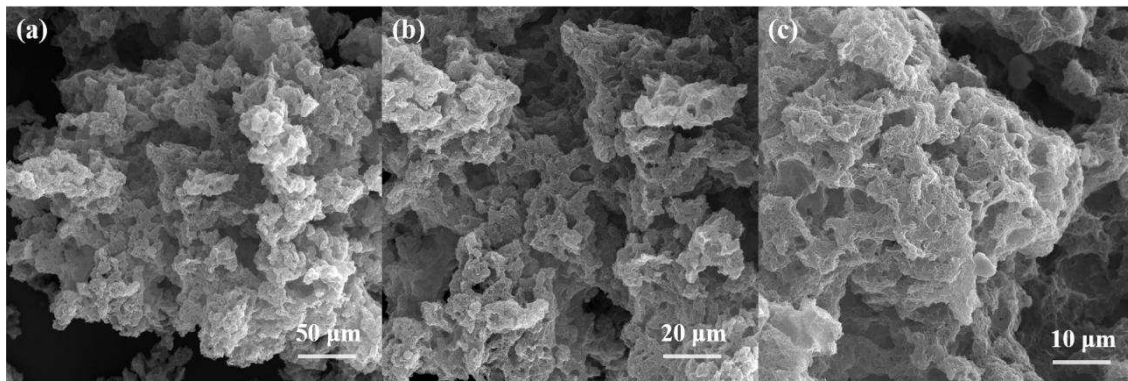


Fig. 3 SEM image of carbon nanotube/graphene three-dimensional framework

Fig.4 shows the Fourier transform infrared (FT-IR) spectra of carbon nanotube/graphene composite shaped phase change materials and pure hexadecanol. The characteristic peaks of hexadecanol are as follows: the CH₂ stretching vibration peaks are observed at 2912 cm⁻¹ and 2856 cm⁻¹, a C-H bending vibration peak is present near 1500 cm⁻¹, a C-H stretching vibration peak is evident at 752 cm⁻¹, and a C-O stretching vibration peak is visible near 1180 cm⁻¹. There are no additional peaks in the FT-IR spectra of PCM0, PCM1, PCM2, and PCM3, indicating that carbon nanotubes, graphene, and hexadecanol are only physically combined without any chemical reactions.

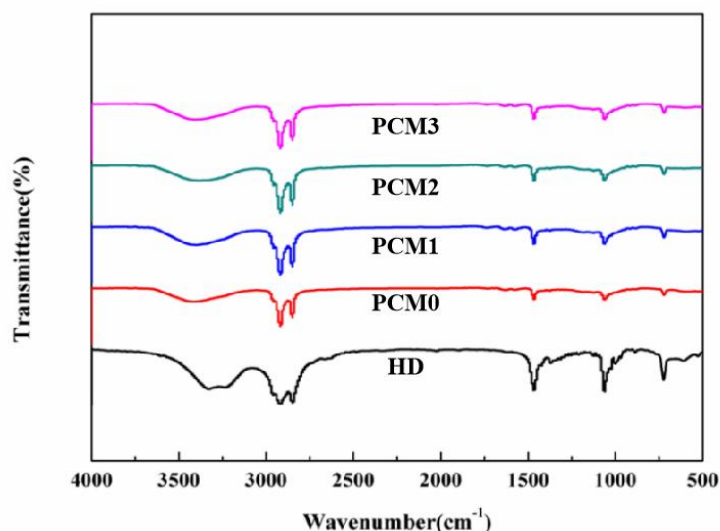


Fig. 4 FT-IR spectra of carbon nanotube/graphene composite shaped phase change materials

3.3 Thermal storage performance

Differential scanning calorimetry (DSC) was employed to measure the corresponding enthalpy values and phase change temperatures of the carbon nanotube/graphene composite PCMs, with the resulting thermograms presented in Figures 5(a) and 5(b). In this study, the thermal storage capacity of the composites is primarily derived from the hexadecanol, while the carbon framework does not participate in the phase change process. Fig. 5(a-b) displays the DSC curves of pure hexadecanol and the composite phase change materials PCM0, PCM1, PCM2, and PCM3. On the DSC curve, according to the solid-liquid phase transition law of hexadecanol, only observe one absorption peak during the endothermic process and two release peaks during the exothermic process. This is attributed to the formation of a metastable structure in hexadecanol as it transitions from liquid to solid state. The number of absorption and release peaks in the presence of carbon nanotubes and graphene exhibits variations compared to that of pure hexadecanol. This situation confirming that only hexadecanol participates in the energy absorption and release process during the phase transition. However, the shape and position of the absorption and release peaks in PCM0, PCM1, PCM2, and PCM3 differ from those of pure hexadecanol. Indicating that carbon nanotubes and graphene influenced the phase change temperature and enthalpy of the composite phase change materials. In the cooling stage, an additional endothermic peak is observed around 25-30°C in the composite phase change materials compared to pure hexadecanol. This is mainly because the cooling rate of the three-dimensional framework of carbon nanotubes and graphene is faster than that of hexadecanol. Resulting in a slightly higher internal temperature of hexadecanol than both the composite surface and the three-dimensional framework.

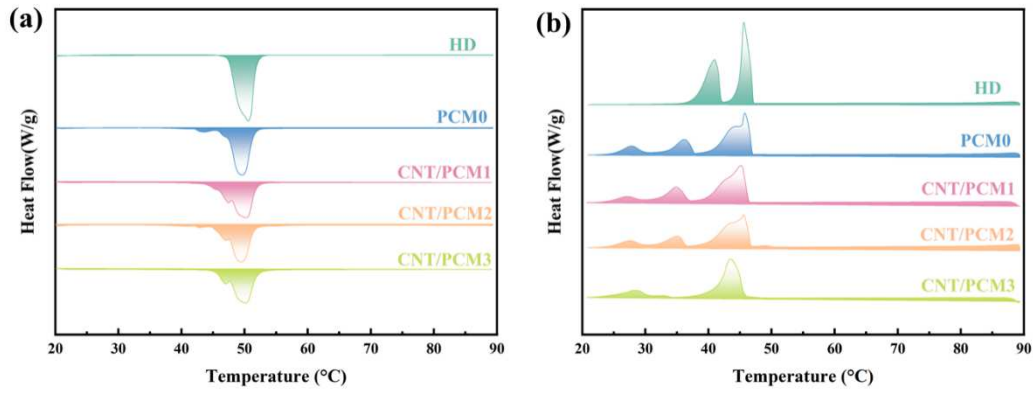


Fig. 5 DSC image of composite phase change material endothermic(a) exothermic(b)

Table 1 DSC data of carbon nanotube/graphene composite shaped phase change materials

	$\Delta H_m(\text{J/g})$	$T_m(^{\circ}\text{C})$	$\Delta H_f(\text{J/g})$	$T_f(^{\circ}\text{C})$
HD	232.05	49.32	229.49	46.42
PCM0	202.38	49.23	195.72	46.15
PCM1	199.16	49.72	189.63	45.45
PCM2	193.18	48.84	158.44	45.04
PCM3	167.51	49.72	135.19	44.08

Table 1 summarizes the phase transition properties for the composite materials, including the enthalpies of melting and solidification, s. Pure hexadecanol exhibits melting and solidification temperatures of 49.32 °C and 46.42 °C, with corresponding enthalpies of 232.05 J/g and 229.49 J/g, respectively. While the phase transition temperatures of the composite materials exhibit only minor differences from pure hexadecanol, the latent heat of phase change decreases notably. This reduction is attributed to the presence of the 3D graphene framework, which lowers the relative mass fraction of the active hexadecanol component within the composite. Specifically, PCM0 displays melting and solidification temperatures of 49.23 °C and 46.15 °C, with measured enthalpies of 202.38 J/g and 195.72 J/g, respectively. PCM1 exhibits a melting point of 49.72 °C and a solidification point of 45.45 °C, yielding a melting enthalpy of 199.16 J/g and a solidification enthalpy of 189.63 J/g. For PCM2, with a carbon nanotube mass fraction of 10 wt.%, the melting and solidification temperatures are 48.84 °C and 45.04 °C, respectively; the corresponding enthalpies are 193.18 J/g and 158.44 J/g. Finally, PCM3 exhibits a melting temperature of 49.72 °C and a solidification temperature of 44.08 °C, with further reduced melting and solidification enthalpies of 167.51 J/g and 135.19 J/g, respectively.

3.4 Thermal stability

Put pure hexadecanol and the carbon nanotube (CNT)/graphene composite phase change materials on a heating platform maintained at 70 °C, to evaluate shape stability of those materials. Figure 6 presents the visual comparison of the samples before and after heating. It can be observed

that pure hexadecanol, without the addition of carbon nanotubes and graphene as reinforcing materials, exhibited the most severe leakage, melting completely after being heated at 70°C for 15 minutes. According to the illustration, the introduction of graphene and carbon nanotubes into the composite phase change materials effectively resolved the leakage issue. After the complete leakage of pure hexadecanol, no significant leakage was observed in the composite phase change materials. Increasing the doping amount of carbon nanotubes significantly enhanced the anti-leakage performance of the materials. The introduction of carbon nanotubes can strengthen the morphological stability of hexadecanol. The excellent shape stability of the sample originating from hydrogen bonding between GO and HD, interfacial tension and capillary interaction, making it highly suitable for practical applications.

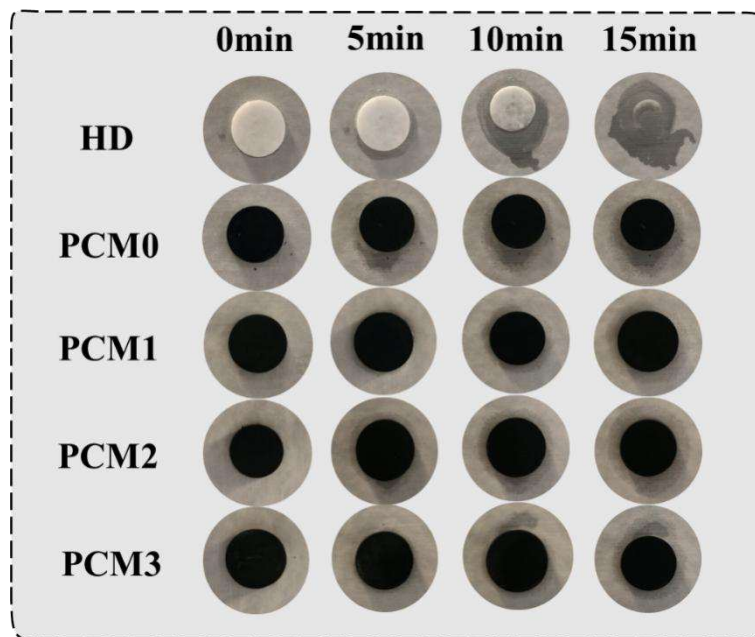


Fig.6 Leakage experiment diagram of carbon nanotube/graphene composite shaped phase change material

The thermal gravimetric analyzer (TGA) was employed to investigate the thermal stability of pure hexadecanol, PCM0, PCM1, PCM2, and PCM3. As shown in Figure 6, the analysis revealed that hexadecanol initiated mass loss at 151°C and completed its mass loss at 240°C. PCM0 initiated mass loss at 141°C and completed its mass loss at around 250°C. PCM1 started mass loss at 142°C and ended at approximately 249°C. PCM2 initiated mass loss at 72°C and finished at around 247°C. PCM3 started to mass loss at 130°C and concluded at approximately 234°C. The mass loss rates were 98.87% for hexadecanol, 98.86% for PCM0, 98.54% for PCM1, 98.10% for PCM2, and 97.45% for PCM3. These results indicate that the three-dimensional framework of carbon nanotubes and graphene in the composite phase change materials slowed down the weight loss, thereby enhancing the thermal stability of hexadecanol.

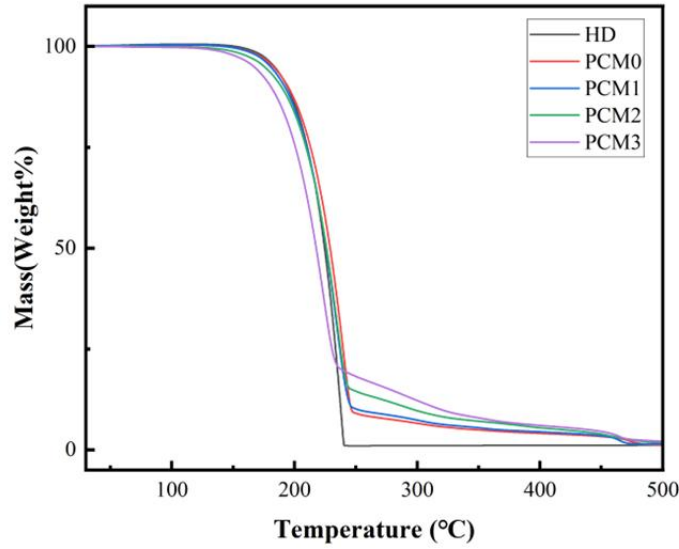


Fig.7 Thermogravimetric analysis of hexadecanol and carbon nanotube/graphene composite shaped phase change materials;

3.5 Thermal conductivity

Figure 7 compares the thermal conductivity of pure hexadecanol with that of the CNT/graphene composite PCMs incorporating varying CNT fractions. The measured thermal conductivities for CNT loadings of 0 wt.%, 5 wt.%, 10 wt.%, and 20 wt.% were 0.434, 0.446, 0.455, and 0.464 $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, respectively. Relative to pristine hexadecanol, these values correspond to enhancements of 17.30 % (PCM0), 20.54 % (PCM1), 22.97 % (PCM2), and 25.40 % (PCM3). These data confirm that the inclusion of CNTs consistently elevates the thermal transport properties of the composites, and the observed conductivity increases monotonically with rising CNT content

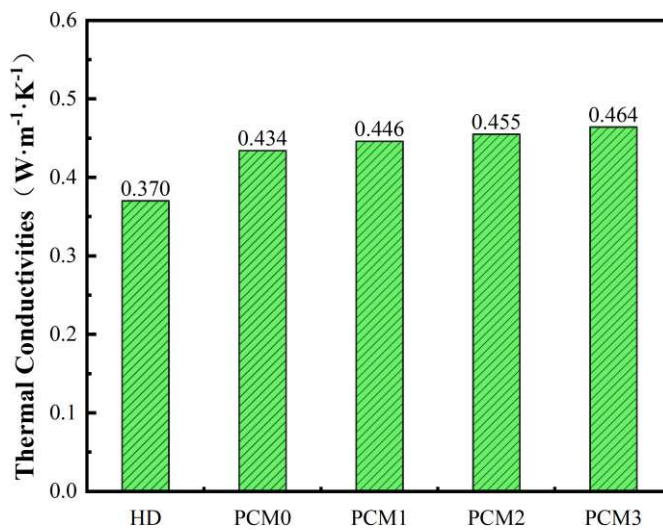


Fig.8 Thermal conductivity of carbon nanotube/graphene composite shaped phase change materials;

3.6 Photothermal conversion performance

To assess the photothermal conversion capability of the composite PCMs, a dedicated photothermal measurement setup was constructed. Under simulated solar irradiation, photons are captured by the surface of the carbonaceous carrier material and directly converted into thermal energy, which is subsequently transferred to the hexadecanol via thermal conduction within the composite structure. Consequently, the composite phase change material (PCM) effectively store the collected solar radiation as thermal energy. Throughout the process, infrared thermography was employed to record the time-temperature profiles of pure hexadecanol and the composite samples (PCM0, PCM1, PCM2, and PCM3) to assess their photothermal conversion efficiency.

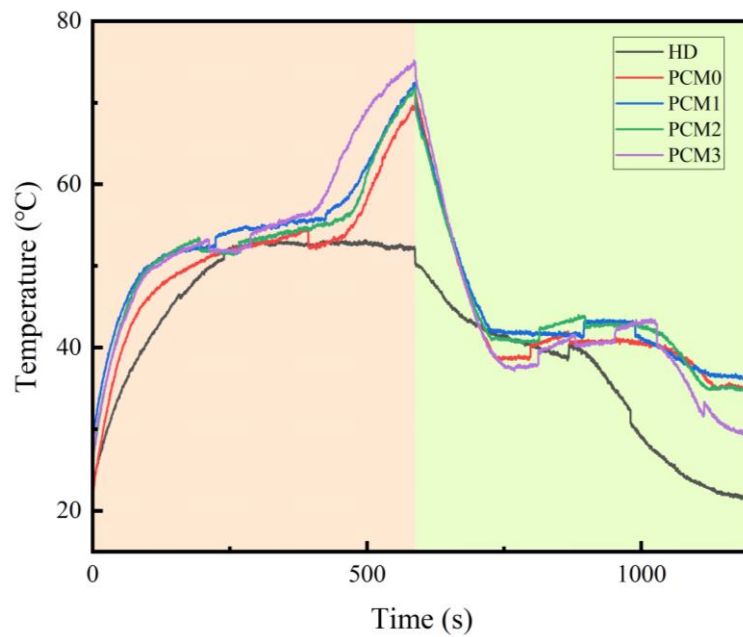


Fig.9 Infrared thermal imaging of carbon nanotube/graphene composite phase change materials

The time-temperature curves obtained from the photothermal experiments are presented in Figure 8. The heating rate decreased when the composites' temperature approaches the phase transition point. The curves exhibit a distinct phase transition platform between 49 °C and 57 °C. And the surface temperatures of PCM1, PCM2, and PCM3 are consistently higher than that of pure hexadecanol in given heating duration. Compared with pure hexadecanol, the composite materials exhibit a superior heating rate, which can be attributed to the addition of carbon nanotubes and graphene has significantly enhanced the photothermal conversion efficiency. Furthermore, the maximum equilibrium temperature and the heating rate of the composites exhibit a positive correlation with the carbon nanotube content. Notably, PCM3 displays the most rapid temperature rise, reaching 75 °C within 600 s, whereas PCM0 only reaches 70 °C. This suggests that a higher concentration of photon-capturing carbon fillers can enhances the ability of materials to harvest

photons, thereby accelerating the thermal response rate. During the cooling phase, the slopes of the curves indicate that the composite PCMs exhibit a greater cooling rate compared with pure hexadecanol. Among these samples, PCM3 decreasing from 73 °C to 40 °C within 110 s which exhibit a most rapidly cooling rates, while the cooling rates of the other composites are slightly lower than that of PCM3.

4 Conclusions

Using graphene as the three-dimensional skeleton and carboxylated multi-walled carbon nanotubes (MWCNTs-COOH) as the thermal conductive filler. This study successfully developed a carbon nanotube/graphene composite shaped phase change material (PCM) via one-pot synthesis based on Pickering emulsion. The Pickering emulsion method provides a simple and scalable approach for constructing three-dimensional graphene-based PCMs. Graphene replaces conventional emulsifiers, simplifying the preparation process and raw materials, while ensuring uniform dispersion of graphene and carbon nanotubes within the matrix and strong interfacial interaction.

The composite exhibits significantly improved comprehensive performance. Its thermal conductivity increases by up to 25.40 % compared to pure hexadecanol (HD) for the sample containing 20 wt.% MWCNT-COOH, with the interconnected thermal conduction network contributed by graphene and carbon nanotubes playing a key role. The three-dimensional graphene skeleton and carbon nanotubes together provide excellent shape stability and good thermal stability, with almost no leakage even after prolonged heating at 70 °C. The latent heat remains at an effective level, ensuring energy storage capacity. Furthermore, the presence of graphene and carbon nanotubes enhances photothermal conversion efficiency, enabling rapid absorption and conversion of solar energy into thermal energy.

This carbon nanotube/graphene composite PCM exhibits high thermal conductivity, excellent leak-proof performance, and efficient photothermal conversion. Therefore, it shows promise for management applications and thermal energy storage, such as thermal management of electronic equipment, solar energy storage and building energy conservation.

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Author Contributions

Conceptualization, Y. S. and Z. L.; methodology, Y. S. and H. J. Y., investigation, H. J. Y.; writing—original draft preparation, H. J. Y. and Y. S.; writing—review and editing, Y. S., and Z. L.; project administration, M. Q. C.; All authors have read and agreed to the published version of the manuscript.

Letter Symbols

PCM	Phase change materials
CNT	Carbon nanotube
RGO	Reduced graphene oxide
GO	Graphene oxide
HD	Hexadecanol
MWCNTs-COOH	Carboxylated multi-walled carbon nanotubes
VC	Ascorbic acid
DSC	Differential scanning calorimetry
TGA	Thermogravimetric analysis
FT-IR	Fourier transform infrared spectroscopy
SEM	Scanning electron microscopy
ΔH_m	Melting enthalpy (J/g)
ΔH_f	Solidification (freezing) enthalpy (J/g)
T_m	Melting temperature (°C)
T_f	Solidification temperature (°C)

Data availability

Data will be made available on request.

References

- [1] Y. Wang, W. Zhang, Y. Yin, et al., 2026. High mechanical strength flexible phase change gel: enhanced thermal buffering materials for human skin temperature control. *Thermochimica Acta*, 756, p.180202-180202.
- [2] Mokhtarpour, M., Rostami, A., Shekaari, H. et al., 2023. Novel protic ionic liquids-based phase change materials for high performance thermal energy storage systems. *Science Report*, 13, p.18936.
- [3] B. Eleonora, B. Michele, C. Silvia, 2023. Addition of granular phase change materials (PCMs) and graphene to a cement-based mortar to improve its thermal performances. *Applied Thermal Engineering*, 229.
- [4] W. Huang, Z. Wang, H. Zhou, et al., 2024. Highly thermal conductive Graphene/Paraffin composite for efficient thermal management of electronics. *Applied Thermal Engineering*, 246, p.122958.
- [5] Ren, Z., Wang, Z., Liu, Z. et al., 2025. Enhancing energy efficiency of walls in summer and winter with novel phase change storage concrete design. *Science Report*, 15, p.40655.
- [6] Hussien S. A., Ali, A. B. M., Alkhatib O. J. et al., 2026. Enhancing battery thermal management system using the emulsion of encapsulated phase change material and water in a mini helical tube. *Science Report*, 16, 573.
- [7] Golovchak R., Plummer J., Kovalskiy A. et al., 2023 Phase-change materials based on amorphous equichalcogenides. *Science Report*, 13, p.2881.
- [8] Wang, H. Zhang, B. Dou, et al., 2022. Effect of copper metal foam proportion on heat transfer enhancement in the melting process of phase change materials. *Applied Thermal Engineering*, 201.
- [9] F. He, W. Hong, Z. Liu, et al., 2024. Phase change materials encapsulated in graphene hybrid aerogels with high thermal conductivity for efficient solar-thermal energy conversion and thermal management of solar PV panels. *Thermochimica Acta*, 740, p.179853-179853.
- [10] X. Yan, H. Zhao, Y. Feng, et al., 2022. Excellent heat transfer and phase transformation performance of erythritol/graphene composite phase change materials. *Composites Part B: Engineering*, 228.
- [11] M. Li, Q. Ma, 2024. Performance improvement of phase change materials encapsulated with graphene oxide for thermal storage. *Journal of Energy Storage*, 80, p.110315.
- [12] L. Jiang, G. Chen, L. Zhao, et al., 2023. Preparation and low temperature heat storage properties of 1-hexadecylamine/3D graphene aerogel composite phase change materials. *Solar Energy Materials and Solar Cells*, 251.
- [13] J. Wu, L. Shi, J. Liu, et al., 2024. In-situ copper-loaded hollow porous carbon nanospheres derived from phenolic resin for thermal energy storage. *Thermochimica Acta*, 736, p.179746-179746.
- [14] M. Shaker, Q. Qin, D. Zhaxi, et al., 2023. Improving the Cold Thermal Energy Storage Performance of Paraffin Phase Change Material by Compositing with Graphite, Expanded Graphite, and Graphene. *Journal of Materials Engineering and Performance*, 32 (22), p.10275-10284.
- [15] R. Chen, D. Li, N. Sheng, et al., 2024. Direct synthesis of porous aluminum nitride foams for enhancing heat transfer and anti-leakage performance of phase change materials. *Thermochimica Acta*, 734, p.179706-179706.

-
- [16] L. An, Y. Yu, Q. Cai, et al., 2023. Hexagonal boron nitride nanosheets: Preparation, heat transport property and application as thermally conductive fillers. *Progress in Materials Science*, 138.
- [17] W. Chen, L. Jiang, X. Hu, 2025. A shape-stabilized and thermally enhanced composite phase change material based on Ag@rGO-CMF three-dimensional porous skeleton. *Journal of Energy Storage*, 114, p.115751-115751.
- [18] Y. Yang, Q. Yin, F. Xu, et al., 2022. Fabricated Polyethylene glycol/hydroxylated carbon nanotubes shape-stabilized phase change materials with improving thermal conductivity. *Thermochimica Acta*, 718.
- [19] Y. Chen, Q. Ma, L. Chen, et al., 2023. Enhanced light-to-thermal conversion performance of self-assembly carbon nanotube/graphene-interconnected phase change materials for thermal-electric device. *Journal of Energy Storage*, 72.
- [20] R. K. Rajamony, J. K. S. Paw, J. Pasupuleti, et al., 2024. Experimental investigation on the performance of binary carbon-based nano-enhanced inorganic phase change materials for thermal energy storage applications. *Journal of Energy Storage*, 86, p.111373.
- [21] F. He, W. Hong, Z. Liu, et al., 2024. Shape-stabilized phase change materials based on polyvinyl alcohol/carboxylated carbon nanotubes/graphene hybrid aerogels for efficient solar-thermal conversion and thermal camouflage. *Solar Energy Materials and Solar Cells*, 274, p.112973.
- [22] X. Kong, J. Lu, M. Xu, et al., 2025. Facile preparation of functional decorated of phase change materials composites based on paraffin @ sodium alginate. *Thermochimica Acta*, 744, p.179922-179922.
- [23] H. N. R. Jung, V. G. Parale, H. Choi, et al., 2024. Investigation of compound state of SiO₂-TiO₂ aerogel synthesized through controlled sol-gel reaction. *Journal of Alloys and Compounds*, 980, p.173561.
- [24] Q. He, H. Fei, J. Zhou, et al., 2023. Utilization of carbonized water hyacinth for effective encapsulation and thermal conductivity enhancement of phase change energy storage materials. *Construction and Building Materials*, 372.
- [25] J. Jiang, H. Zhang, L. Liu, et al., 2023. One-step preparation of double network phase change composite with GO for enhanced thermal management performance. *Composites Science and Technology*, 243.
- [26] J. Singh, S. Parvate, V. J. Reddy, et al., 2022. Facile method to prepare 1-dodecanol@poly(melamine-paraformaldehyde) phase change energy storage microcapsules via surfactant-free method. *Journal of Energy Storage*, 49.
- [27] W. Li, D. Mei, J. Wang, et al., 2023. Preparation of Microencapsulated Phase Change Materials from Sulfonated Graphene Stabilized Pickering Emulsion. *Polymers*, 15 (11).
- [28] Q. Zhao, S. Y. Wong, Y. Zhang, et al., 2025. Supercooling suppression of microencapsulated capric acid-stearic acid eutectic via lignin-zein stabilized Pickering emulsion for highly efficient thermal energy storage. *International Journal of Biological Macromolecules*, 307, p.141989.
- [29] W. Liu, Q. Lin, S. Chen, et al., 2023. Microencapsulated phase change material through cellulose nanofibrils stabilized Pickering emulsion templating. *Advanced Composites and Hybrid Materials*, 6 (4).

-
- [30] X. Shen, B. Wang, N. Li, et al., 2025. Self-emulsifying polyethylene glycol-based nanocapsule phase change material with enhanced thermal conductivity. *Journal of Energy Storage*, 108, p.115061-115061.
- [31] L. Cao, Z. Li, Y. Zhang, et al., 2024. Silica-based aerogels encapsulate organic/inorganic composite phase change materials for building thermal management. *Journal of Energy Storage*, 97, p.112858-112858.
- [32] F. Shao, L. Wang, R. Luo, et al., 2023. Shape-Stable Hybrid Emulsion Gel with Sodium Acetate Trihydrate and Paraffin Wax for Efficient Solar Energy Storage and Building Thermal Management. *ACS Applied Materials & Interfaces*, 15.