



# Thermal performance enhancement of palmitic-stearic acid by adding graphene nanoplatelets and expanded graphite for thermal energy storage: A comparative study



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## ABSTRACT

The effects of adding GnPs (graphene nanoplatelets) (nanoscale) and EG (expanded graphite) (micro-scale) were investigated to improve the thermal performance of PA-SA (palmitic-stearic acid) eutectic mixture as a PCM (phase change material). Carbon materials were dispersed into PA-SA with loadings of 1wt%, 2wt%, 4wt%, and 8wt%. The thermal properties measurement results show that the phase change latent heats of composite PCMs decreased with increasing loadings. The thermal conductivities of the composite PCMs were measured by the transient plane heat source method. Both GnPs and EG can effectively improve the thermal conductivity of PA-SA, but EG is more effective due to its worm-like structure. For the highest loading (8wt%), the thermal conductivity of the composite PCMs is 2.7 times higher with GnPs and 15.8 times higher with EG than that of PA-SA at 25 °C. The thermal energy storage and release rates of the composite PCMs also increased due to the high thermal conductivity of carbon materials. The density of composite PCMs were found to increase with the addition of GnPs but to decrease with EG. All composite PCMs show good thermal reliability. This work shows that EG has the greater potential to enhance the thermal energy storage performance of PCMs.

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## 1. Introduction

In recent years, the energy crisis has become increasingly serious. Thermal energy storage is a crucial technology to solve this crisis. Storing the energy from renewable energy resources such as solar and wind, especially the energy generated during off-peak periods, is crucial for the widespread application of renewable energy technologies. Using PCM (phase change materials) in LHTEs (latent heat thermal energy storage) systems is one of the most attractive methods for energy storage because PCMs can store or release thermal energy during the phase transition process. Based on the type of phase transition, PCMs are classified as solid–solid, solid–liquid, solid–gas, and liquid–gas PCMs. Among these, the solid–liquid PCMs are the most suitable for thermal energy storage. When the temperature increases, PCMs change phase from solid to liquid, and they absorb heat. As the temperature decreases, the phase change back from liquid to solid, and they release heat. Using PCMs is an advantageous way to increase the energy efficiency of

LHTEs systems because of the advantages of high latent heat storage capacity and small temperature variation during the phase transition process [1–3]. PCMs have been widely applied in many energy conservation fields, such as waste heat recovery [4], building energy efficiency [5–7], solar energy storage [8,9], etc. [10].

Inorganic compounds such as salt hydrates, metals, and organic compounds such as paraffin waxes, fatty alcohols, fatty acids and their mixtures have been applied as solid–liquid PCMs [11,12]. Among the organic PCMs, PA (palmitic acid) and SA (stearic acid) have attracted a great deal of attention due to their high heat capacity, low supercooling, self-nucleating behaviour, non-toxicity, abundance, low cost, and good thermal and chemical stability. The phase change temperature of the PA-SA (palmitic-stearic acid) eutectic mixture is more suitable for some thermal energy storage systems than those of PA and SA. However, regardless whether PCMs are inorganic or organic, their application is restricted by their low thermal conductivity, which affects the thermal energy charge and discharge rates. This has prompted efforts to enhance the heat transfer of PCMs in latent heat thermal energy storage systems [13], which can be realized by introducing materials with high thermal conductivity into PCMs to form a composite. Early

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research focused on inserting metal powders, fins, foams or rings into PCMs [14–16], which provide the disadvantages of increasing the weight and cost of the system while damaging the fluidity of PCMs during the melting process.

There is a recent tendency to introduce various thermal enhancement additives such as carbon materials into PCMs to improve the thermal conductivity of PCMs. Carbon materials are non-toxic, safe, and environmentally friendly, and they have high thermal conductivity and relatively low density. Thus, they can significantly enhance the thermal conductivity of PCMs with only a slight loss in the heat storage capacity [17–20]. Carbon nanomaterials with excellent properties are favoured for improving the thermal conductivity of PCMs [21–23]. Tao et al. [24] mixed four kinds of carbon nanomaterials with different microstructures into binary carbonate eutectic salts to enhance the performance of salt PCMs. The results show that the nanomaterial microstructure has significant effects on the thermal properties of composite PCMs. Sheet-like graphene is the best additive to enhance the specific heat. SWCNTs (single-walled carbon nanotubes), which have a columnar structure, showed the best results, improving the thermal conductivity up to 56.98%. Shi et al. [25] studied improvements in the thermal conductivity and the shape-stability of paraffin by adding xGnP (exfoliated graphite nanoplatelets) and graphene. Kim et al. [26] mixed xGnP into liquid paraffin, and the obtained paraffin/xGnP composite PCMs manifested high electrical and thermal conductivities. Cui et al. [27] studied the improvements in thermal conductivity due to CNF (carbon nanofiber) and CNT filled soy wax and paraffin wax. Fan et al. [28] investigated the effects of short and long MWCNTs, CNFs and GnPs (graphene nanoplatelets) on the thermal conductivity and energy storage properties of paraffin-based nanocomposites PCMs for thermal energy storage.

Meanwhile, EG (expanded graphite), which has a worm-like network pore structure at the micrometre scale, is a common additive adopted to enhance the thermal conductivity of PCMs [29–32]. Xia et al. [33] prepared EG/paraffin composite PCMs with varying mass fractions of EG from 0 to 10 wt%, and the results show that adding 10 wt% EG can yield a thermal conductivity 10 times higher than that of pure paraffin. Mills et al. [34] loaded paraffin into graphite blocks made by compaction EG, and the results showed that the thermal conductivity of the composite is 20–130 times higher than that of pure PCMs. Lee et al. [35] combined EG with various interlayer distances with molten erythritol to prepare erythritol/EG composites by a simple blending and impregnating method. The composite containing the EG with the largest interlayer distance had a thermal conductivity of 3.56 W/mK and a latent heat value of 90 mass% of pure erythritol. Zeng et al. [36] improved the thermal conductivity of tetradecanol by adding EG, and the thermal conductivity of the composite form-stable PCM containing 7 wt% EG was 2.76 W/(m·K).

Among the various light carbon materials with high thermal conductivity, micrometre-scale EG and nanometre-scale GnPs are considered to be excellent heat transfer promoters. Although the effects of both EG and GnPs on the thermal performance of PCMs have been investigated in the literature, thus far there has been no study comparing their thermal enhancement effects on PCMs. Therefore, this paper aims to compare the different effects and mechanisms of GnPs and EG on the thermal performance of PA-SA (palmitic-stearic acid) eutectic mixture. Moreover, to study the effect of the amount of additive on the thermal performance of PCMs, four different mass fractions of GnPs and EG were added into PA-SA. The various PA-SA/GnPs and PA-SA/EG composite PCMs were prepared by the blending and ultrasonic processing method. The effects of EG and GnPs on the thermal

properties, thermal conductivity, and thermal reliability were studied experimentally.

## 2. Experimental

### 2.1. Materials

Palmitic acid (PA, AR) and stearic acid (SA, 98% pure) were chosen to prepare the fatty acid eutectic mixture, and then PA-SA was used as the matrix PCM. Expandable graphite (80 meshes, expansion coefficient: 200 ml/g, carbon content: 99%) was used to prepare the EG via microwave treatment. GnPs (>99.5 wt%, thickness: 4–20 nm, layers: <20, size: 5–10  $\mu\text{m}$ ) were purchased from Chengdu Organic Chemical Co. Ltd. China and used as a thermal conductivity enhancement additive. PVP (polyvinyl pyrrolidone) (AR) was used to stabilise the EG and GnP dispersions in the PA-SA.

### 2.2. Preparation of the PA-SA composite PCMs

Based on our previous work [37,38], PA-SA was prepared with a mass ratio of 62 wt% PA and 38 wt% SA. The phase diagram of the PA-SA is shown in Fig. 1. To obtain the PA-SA, a certain amount of PA and SA was heated in a beaker at 80 °C and then stirred for 30 min to ensure a homogenous mixture. The EG was obtained by treating the as-received expandable graphite in a microwave oven an irradiation power of 700 W for 30 s.

The PA-SA/GnPs composite PCMs were prepared by the following procedure: the PA-SA was melted in a thermostatic water bath at 70 °C; GnPs and PVP were added in the molten PA-SA to achieve equivalent mass fractions of 1 wt%, 2 wt%, 4 wt%, and 8 wt%, and the mixtures were stirred vigorously by a magnetic stirrer for 1 h and then ultrasonicated for 1 h again to disperse the GnPs and PVP in the PA-SA. Next, the prepared samples were labelled as PA-SA/GnPs-1, PA-SA/GnPs-2, PA-SA/GnPs-4, and PA-SA/GnPs-8. The PA-SA/EG composite PCMs were prepared following the same procedure, substituting the EG for the GnPs, and the samples were listed as PA-SA/EG-1, PA-SA/EG-2, PA-SA/EG-4 and PA-SA/EG-8.

### 2.3. Characterization

The microstructures of the carbon materials and the composite PCMs were observed using a SEM (scanning electronic microscope) (Fei Inspect FEI, the Netherlands). The structural analysis of samples was carried out using a FT-IR (Fourier transform infrared spectrometer) (Nicolet 6700, USA). The specific surface areas of the GnPs and EG were measured by the BET (Brunauer–Emmett–Teller) method using a specific surface area analyser (Beishide Instrument 3H-2000PS2). The FT-IR spectra were recorded on a KBr pellet in the frequency range of 4000  $\text{cm}^{-1}$  to 400  $\text{cm}^{-1}$ . The phase change temperatures and latent heats of the samples were measured using a DSC (differential scanning calorimeter) (TA Q20 USA) at 5 °C/min under a constant stream of argon at a flow rate of 50 ml/min. The DSC instrument was calibrated with indium as a standard reference material, and the accuracy of the enthalpy measurement was  $\pm 4\%$ . The DSC measurements were carried out on three 5–10 mg specimens of each sample to ensure data reproducibility. Accelerated thermal cycling tests were carried out in a metal bath (CHB-T2-E, BIOER, China). The prepared composite PCMs were placed in a 2 ml centrifuge tube and heated in the metal bath at 70 °C for 1 min and then cooled at 30 °C for 1 min. The PCMs undergo a phase change between these temperatures. The heating and cooling rates were 10 °C/min to save time and ensure

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