



Graphene Aerogels Enhanced Phase Change Materials prepared by one-pot method with high thermal conductivity and large latent energy storage

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ARTICLE INFO

Keywords:

Phase change material (PCM)

Graphene

Ag

One-pot hydrothermal method

Thermal conductivity

Thermal energy storage

ABSTRACT

Phase change Materials (PCMs) have a promising future in the energy fields of latent heat storage, solar energy conversion and building thermal management. In this work, two types of reduced graphene oxide (rGO) with different diameters were prepared and combined with paraffin, octadecanol and stearic acid PCMs, respectively. The one-pot hydrothermal method was employed for the synthesis of three-dimensional graphene network encapsulating organic PCMs. There is some research investigate that rGO addition has different degrees of impact on the latent heat and thermal conductivity of different kinds of organic PCMs. The rGO/PCM composite has a significantly enhanced thermal conductivity of $3.21 \text{ W m}^{-1} \text{ K}^{-1}$ with an rGO loading of 10 wt%. Furthermore, a fraction of graphene was substituted by Ag nanoparticles. The thermal conductivity of rGO/PCM/Ag could be as high as $5.89 \text{ W m}^{-1} \text{ K}^{-1}$. The size effect of rGO has a significant influence on the thermal contact resistance. The synergistic effect of rGO network and Ag nanoparticles also benefits the thermal conductivity of nano-enhanced phase change materials (NePCMs).

1. Introduction

Phase change Materials (PCMs) have been widely applied in the energy fields of latent heat storage, solar energy conversion, building thermal management, etc. [1–8] Organic PCMs, like paraffin wax, have prominent advantages of the high latent heat of fusion and low melting temperature, which are suitable for thermal storage applications. However, the thermal conductivity restricts, which leads to the low energy charge and discharge efficiency during melting and solidification cycles and, therefore, limits the overall storage capacity of the PCM. A lot of efforts have been taken to enhance the thermal conductivity of PCMs [9–20]. One practical approach is to use porous structure, high-conductivity materials impregnated with PCM. Studies have reported a variety of materials were embedded into PCMs to compensate the conductivity, such as metal particles [9,10], metal foam [11,12], expanded graphite [13] and carbon nanotubes [14–17].

Graphene is the one-atom-thick carbon sheet with high thermal conductivity ($3100\text{--}5300 \text{ W m}^{-1} \text{ K}^{-1}$) [21,22], high specific surface area (SSA, $\sim 2630 \text{ m}^2 \text{ g}^{-1}$) [23] and excellent mechanical property [24,25]. Superior to other nano-enhanced phase change materials (NePCMs), graphene-based PCMs are available to achieve higher thermal conductivity with an extremely low graphene loading, mitigate some potential leakage by encapsulation and improve the shape stability of NePCMs [26–32]. Yu et al. [15] investigated the effect of adding

various carbon nanomaterials on the thermal conductivity of paraffin PCMs. The study proved graphene nanoplatelets (GNPs) could reduce thermal interface resistance associated with their two-dimensional planar structure to increase the thermal conductivity of composite, which was as high as $0.29 \text{ W m}^{-1} \text{ K}^{-1}$ at the graphene addition of 4 wt %. Due to the efficient conductivity network and uniform distribution, graphene aerogel (GA) has attracted increasing attention on NePCMs. Yang et al. [29] prepared by vacuum-assisted impregnating of polyethylene glycol (PEG) into the cellulose/GNP aerogels, which exhibit high thermal conductivity, good shape stability and high latent heat of fusion. With only 5.3 wt% of GNP, the composite exhibits a high thermal conductivity of $1.35 \text{ W m}^{-1} \text{ K}^{-1}$. Furthermore, the 3D graphene aerogel/ octadecanoic acid PCM was produced by Zhong et al. [32]. The composite had a thermal conductivity about $2.635 \text{ W m}^{-1} \text{ K}^{-1}$ at a graphene loading fraction of $\sim 20 \text{ vol}\%$, which was about 14 times that of the pure octadecanoic acid ($0.184 \text{ W m}^{-1} \text{ K}^{-1}$).

Although there are many excellent types of research have been reported, the effect of graphene sheets on thermal behaviors of NePCMs still has potential to be investigated. In this work, two types of reduced graphene oxide with different diameters were prepared and combined with paraffin (PA), octadecanol (OA) and stearic acid (SA) PCMs, respectively. Preparation techniques for the NePCMs are generally achieved by impregnation method, melted blending and pickering emulsion method. There are some difficulties in dealing with the

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Nomenclature

PCM	phase change material
SSA	specific surface area
NePCM	nano-enhanced phase change material
GNP	graphene nanoplatelet
GA	graphene aerogel
PEG	polyethylene glycol
SDBS	sodium dodecylbenzene sulfonate
PA	paraffin
OA	octadecanol
SA	stearic acid
GO	graphene oxide
rGO	reduced graphene oxide
GON	graphene oxide nanoplatelet
EG	expanded graphite
APS	average pore size

κ	thermal conductivity
C_p	specific heat capacity
α	thermal diffusivity
ρ	density
ω	porosity
δ	conductivity
T_m	melting point
ΔH_{fus}	latent heat of fusion
φ	mass fraction of rGO
κ_{eff}	effective thermal conductivity
R_{κ}	thermal contact resistance
$\kappa_{p,11}, \kappa_{p,33}$	thermal conductivity in-plane and along-thickness direction
κ_0	thermal conductivity of matrix
P	aspect ratio of 2D nanoparticles
D	average diameter of nanoparticles

dispersion of filler and the enhancement of thermal conductivity. Unlike the previous method, the one-pot hydrothermal method was employed for the synthesis of three-dimensional GA network encapsulating organic PCMs. It has advantages in easy operation and good dispersion.

2. Experimental

2.1. Materials

The 325 mesh and 8000 mesh of graphite powder were obtained from Aladdin Industrial Corporation (China). $KMnO_4$, H_2SO_4 , sodium dodecylbenzene sulfonate (SDBS), PA, OA and SA were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai China). All these chemicals were analytical grade and used as received without further purification. Distilled water was used throughout the experiments.

2.2. Preparation of graphene oxide

Graphene oxide (GO) was synthesized by a modified Hummer's method [30]. Firstly, 5 g of graphite powder with concentrated H_2SO_4 (200 mL) in a round-bottom flask was stirred under an ice bath. Then, 15 g of $KMnO_4$ was gradually added with intensive stirring, and the reaction was maintained below 5 °C for 2 h. Further, the solution was stirred for 6 h at a constant temperature of 35 °C to achieve a dark brown color paste. After this, 225 mL of distilled water was poured into the mixture, and the temperatures were raised to 90 °C and kept for 1 h. Subsequently, 150 mL of H_2O_2 (3.5%) solution was added. The final

product was subjected to centrifugation and thoroughly washed with 5% HCl and water. The obtained graphite oxide paste was diluted into 3 mg mL⁻¹ and sonicated for 5 h to be a homogeneous GO aqueous dispersion. The GO-1 and GO-2 samples were obtained by the 325 and 8000 mesh graphite, respectively.

2.3. Preparation of rGO/PCMs

Self-assembled rGO-1 and rGO-2 gels were directly reduced from GO-1 and GO-2 via the hydrothermal method reported by Shi et al. [31]. Traditionally, as-prepared rGO aerogel was put into the melted organic PCM via vacuum impregnation. Whereas, in this work, the preparation process of rGO/PCMs is shown in Fig. 1. PCM was heated to the temperature above the melting point. SDBS, acting as the emulsifier, was added into the melted PCM and stirred. After complete emulsification, a certain proportion of GO aqueous was poured into the mixture and kept heating and stirring. Then, the composite solution was transferred into a Teflon-lined autoclave and maintained at 180 °C for 4 h. The gel sample was naturally cooled to room temperature and dried for 24 h at 40 °C. The samples prepared by common impregnation method were also carried as the controlled experiments.

2.4. Preparation of rGO/PCMs/Ag

The Ag nanoparticles were prepared via a solution-phase reduction method. The diameter of Ag nanoparticle is about 1.2–1.8 μm, which is shown in Fig. 4f. The Ag suspension was rapidly added into the compound after GO in synthesis process of rGO/PCMs/Ag.

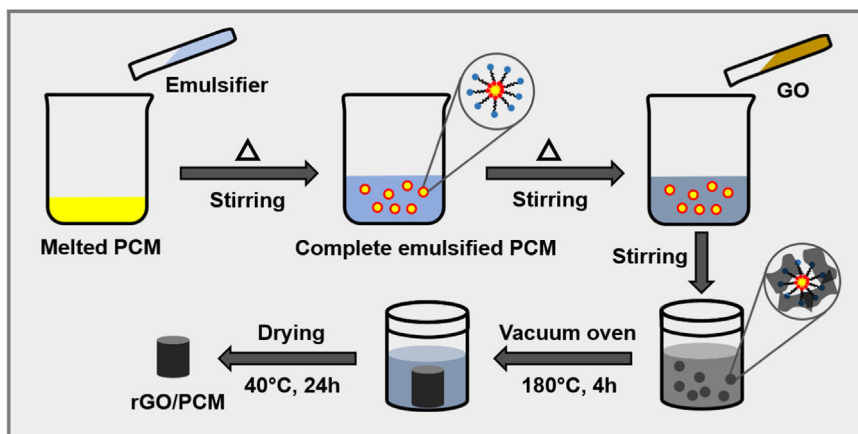


Fig. 1. Schematic of the preparation of rGO/PCMs by hydrothermal treatment.

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