



High-electrical-resistivity thermally-conductive phase change materials prepared by adding nanographitic fillers into paraffin



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ABSTRACT

We added exfoliated graphite nanoplatelets (xGNPs) as a filler into paraffin phase change materials (PCMs) to significantly improve the poor thermal conductivity of paraffin and enhance the heat transfer rate of the PCM. However, the electrical resistivity of paraffin was reduced from above $10^{12} \Omega\text{-cm}$ to below $10 \Omega\text{-cm}$ due to the high electrical conductivity of the added carbon fillers. For applications in electrical devices, such a low resistivity is undesirable since it might cause a short-circuit problem. In order to solve this problem, the xGNPs were thus modified by an acid treatment, which strongly oxidized the surface. The electrical resistivity of the PCM filled with the modified xGNPs (M-xGNPs) was significantly raised by up to 11 orders of magnitude while the thermal conductivity was not severely compromised. This method can improve the safety and usefulness of nanographite-filled PCMs when applied to electrical systems.

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1. Introduction

Phase change materials (PCMs), which can effectively store and release high latent heat during the phase transition, are often used in passive heat management. The temperature of the system can be limited at the phase-transition temperature of the PCM and thus the system is protected against over-heating [1,2]. Applications of organic and inorganic PCMs in latent heat thermal energy storage (LHTES) have been investigated [3–6].

Organic PCMs can be classified into two major groups: paraffinic PCMs and non-paraffinic PCMs [7]. Paraffin-based PCMs are widely used in thermal energy storage applications for the low-to-medium temperature range due to their advantages of high latent heat, low cost, good chemical stability, non-toxicity, and non-corrosiveness. The non-paraffin organic comprises a wide variety of materials such as fatty acids and other non-paraffinic chemicals with highly varied properties useful in different applications. For example, bio-based non-paraffinic PCMs made from natural resources such as soy beans and palm oils are significantly less flammable compared to paraffin [7]. However, organic PCMs usually have poor thermal conductivity, which will obstruct the uniform spreading and fast dissipation of excessive heat.

Graphitic materials are the most commonly used thermally conductive filler in composite PCMs: carbon nanofibers (CNFs) [8–12], carbon nanotubes (CNTs) [12–21], expanded graphite (EG) [22–27], exfoliated graphite nanoplatelets (xGNPs) [28–34] have long been studied in many works. Recently, several researchers have introduced various graphene materials [12,35–37] as a new type of fillers and reported many of their excellent properties. For example, Shi et al. compared the performance of graphene and xGNP fillers in paraffin PCMs [35]. Mehrali and coworkers prepared and characterized palmitic acid PCMs filled with N-doped graphene [36], and undoped graphene with different specific surface areas [37]. Additionally, it has been pointed out that adding graphene and xGNP fillers can effectively raise the dropping temperature of the PCMs, thereby achieving shape stabilization above the melting point [35–37].

In this study, xGNPs were chosen as the graphitic filler for paraffin PCMs. Usually xGNPs are produced by ultrasonically breaking accordion-like EG, which is obtained by rapid thermal expansion of acid-intercalated graphite. Due to the unique planar geometry of xGNPs, they have shown better enhancement effects on the thermal conductivity of PCMs than one-dimensional carbon materials such as CNTs and CNFs [38]. This was explained by the reduced phonon scattering at the interfaces between GNPs and the matrix due to their planar geometry [12]. Researchers have also pointed out that the lateral size of graphene and xGNPs would significantly affect the thermal conductivity in composite PCMs: smaller flakes

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would result in a larger number of thermal contacts, thereby reducing the thermal conductivity [35,39]. However, increasing the loading of xGnP fillers not only improved the thermal conductivity but also reduced the electrical resistance [34,35]. Such a reduction in the resistivity of PCMs is undesirable for applications in electrical devices or systems since this can lead to safety problems.

To solve this problem, we modified xGnPs by an acid treatment to strongly oxidize its surface, thereby blocking the conduction paths of electrons. The thermal and electrical properties for paraffin PCMs filled with pristine and modified xGnPs (M-xGnPs) were compared. Such PCMs with a combination of high thermal conductivity and good electrical insulation will find numerous applications in all kinds of electrical systems such as the thermal management for the battery packs in electric vehicles, portable electronic devices, wearable electronics, aviation electronics, and military electronic systems [34].

2. Experimental

2.1. Materials

The paraffin (n-nonacosane) with a melting temperature of 61.6 °C was obtained from Nippon Seiro. For the preparation of xGnPs and M-xGnPs, commercial acid-intercalated graphite obtained from International Carbide Technology was used as the starting material.

2.2. Preparation of xGnPs and M-xGnPs

To prepare the xGnPs, acid-intercalated graphite was first rapidly expanded in a furnace at 1100 °C for 1 h in argon to produce EG. Pulverization with an ultrasonic processor was employed to break down the worm-like structure into xGnPs. Finally, M-xGnPs were produced by an acid treatment with a mixture of sulfuric acid and nitric acid (3:1; 400 ml in total), which was heated to a temperature of 60, 80, 100 or 120 °C and stirred for 0.3, 0.5, 1, 3, or 5 h at 300 rpm after adding 2 g of xGnPs. After the reaction the solution was repeatedly rinsed with deionized water and filtered until the pH became neutral.

2.3. Preparation of nano-graphite/paraffin composite PCMs

The fabrication process of the PCMs is shown in Fig. 1. Paraffin was first dissolved in toluene at around 80 °C, and then xGnPs or M-xGnPs were added into the solution. Sonication was then applied for 30 min at 100 W to break down the nanographite material aggregates and to ensure a uniform dispersion of the nano-fillers. The resultant mixture was poured into a steel pan and heated to 130 °C in a fume hood to evaporate the solvent. The PCM was then poured into a mold to form a disk-shape sample (see Fig. 2) for the subsequent thermal conductivity and electrical resistivity measurements. Finally, the PCM samples were further dried in a vacuum oven over night at 150 °C.

2.4. Characterization

The morphology of the xGnPs and M-xGnPs was observed with a scanning electron microscope (SEM, JEOL, JSM-6500). Also, the D and G bands of these xGnP and M-xGnP samples in Raman spectroscopy were characterized with a Renishaw in via Raman Microscope. Finally, to investigate the chemical composition of the samples, X-ray photoelectron spectra (XPS) were measured using a high-resolution ULVAC-PHI XPS spectrometer (Model: PHI Quantera SXM/Auger: AES 650).

2.5. Thermal conductivity measurements

All thermal conductivities were measured by a transient plane heat source (hot disc) method (TechMax Technical Group, Hot Disk Analyzer, TPS 1500). The equipment was a basic model suitable for testing homogeneous and isotropic materials. The measurements were carried out according to the ISO 22007-2:2008 test method for the determination of thermal conductivity and thermal diffusivity. The sensor/heater used in this test method consists of a thin nickel foil in a double spiral pattern, which is embedded between two thin layers of Kapton polyimide protective films. This sensor/heater was sandwiched by two identical PCM disks with a diameter of 50 mm and a thickness of 10 mm. A constant power (3 W) was applied for 30 s and the generated heat dissipated within the double spiral was conducted into the surrounding samples, causing a rise in the temperature of the sensor and the samples.

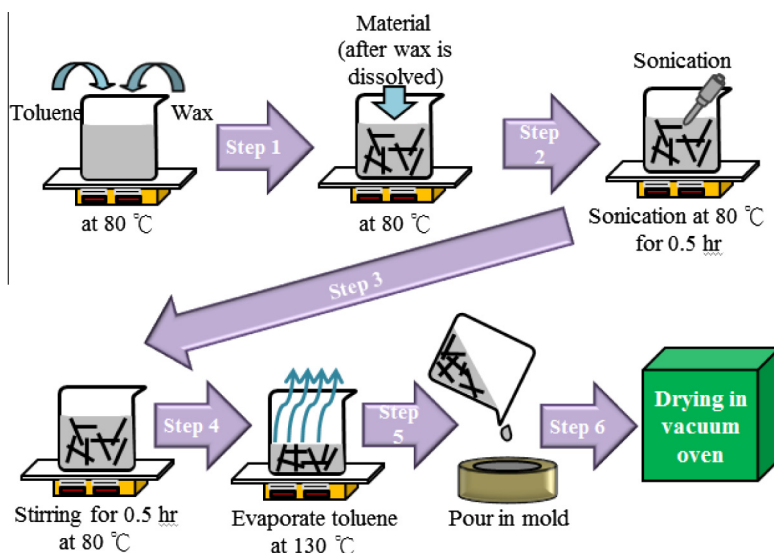


Fig. 1. Fabrication process of the composite PCMs.

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