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# Energy efficient thermal storage montmorillonite with phase change material containing exfoliated graphite nanoplatelets



Solar Energy Material

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### ABSTRACT

In this experiment, we used a vacuum impregnation method to prepare shape stabilized PCM that contained sodium montmorillonite (Na-MMT) and Exfoliated graphite nanoplatelets (xGnP), to improve the thermal conductivity of PCMs, and prevent leakage of the liquid state of PCMs. Na-MMT has low cost and natural abundance, high adsorption and absorption capacities, and fire retardant heating rate. In the used materials, xGnP, usually produced from graphite intercalated compounds, are particles consisting of several layers of graphene sheets. As a result, we found that the FTIR adsorption spectra of paraffinic PCMs did not change, and there was no chemical interaction between paraffinic PCMs and xGnP/Na-MMT mixture. From the DSC analysis, xGnP made an impact on the thermal properties of the paraffinic PCMs/Na-MMT composites. The oxidation rate of paraffinic PCMs based composite with xGnP was greater than that of the composite without xGnP. FTIR, DSC, TGA and TCi were used to determine the characteristics of the paraffinic PCMs/Na-MMT composites.

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#### 1. Introduction

Recently, thermal energy storage (TES) systems have been used to reduce energy use in buildings, to contribute to more efficient environmental energy use, and to supply thermal comport for occupants. The main advantage of using a thermal storage system is that it can contribute to matching supply and demand, when they do not coincide in time [1]. The best known method of TES in buildings involves sensible heat storage, by changing the temperature of a storage material. This can be used for the storage and release of thermal energy in a passive way, but in comparison with latent heat storage, by changing the phase of a storage material, a much larger volume of material is required to store the same amount of energy. Therefore, an effective way to reduce the energy consumption of buildings for heating and cooling is by incorporating phase change materials (PCMs) in passive latent heat thermal energy storage (LHTES) systems of building walls, windows, ceilings, or floors [2]. Latent heat storage technology has been widely used in building. The storage and application of heat are achieved through phase change materials. This has the advantages of high heat storage density, and keeping the temperature stable during the heat storage/release process. The application of PCM in building can not only save energy, but also decrease the temperature fluctuation. The application of PCM in building has been one of the hot topics in latent heat storage technology [3-6]. Unlike conventional sensible storage materials, PCMs absorb and release heat at a nearly constant temperature. They store 5-14 times more heat per unit volume than sensible storage materials, such as water, masonry, or rock. A large number of PCMs are known to melt with a heat of fusion in any required range. The PCM to be used in the design of thermal storage systems should possess desirable thermophysical, kinetic, and chemical properties, which are as follows [7]. They also play an important role in solving energy imbalance, by improving thermal efficiency, and protecting the environment [8,9]. PCMs can be categorized into two major groups: inorganic compounds and organic compounds. Inorganic PCMs, such as salt hydrates, salts, metals, and alloys, generally have high volumetric latent heat storage capacity [7], which is almost twice as much as that of organic PCMs. But their utility is often limited by incongruent melting and supercoiling effects. Also, organic PCMs are classified into paraffinic PCMs, and non-paraffinic PCMs, such as fatty acids [10]. Paraffinic PCMs are considered as one of the most promising candidates, due to their large latent heat, low vapor pressure in the melt, good chemical stability, self-nucleating behavior and safety [11-13]. However, paraffinic PCMs have flowability during the phase change process, as a solid-liquid PCM. Therefore, it is necessary to prepare formstable paraffinic PCM with a micro-capsulation methods or incorporation methods or shape stabilized process [14-17]. Also, paraffinic PCMs suffer from low thermal conductivity and liquid leakage, when they undergo the solid-liquid phase change. These drawbacks reduce the rate of heat storage and extraction during

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the melting and solidification cycles, and restrict their wide application. To overcome the problem related to the low thermal conductivity of paraffinic PCMs, a great many efforts have been made, which include dispersing metallic or nonmetallic particles with high thermal conductivity into PCMs [18,19], using finned tubes with different configurations in a storage unit [20], and impregnating PCM into high thermal conductivity material with porous structure [21-24], such as carbon materials and metal foams. Compared with carbon materials, metal foams, additives or fins not only significantly increase the weight and cost of the storage systems, but for some of them are also incompatible with PCMs. In previous work, we studied the effect of exfoliated graphite nanoplatelets addition on the thermal properties of the paraffin wax/xGnP composite prepared as a form-stable PCM, and reported that the thermal conductivity of PCM increased with increasing graphite mass fraction [25]. Therefore, we prepared PCM/montmorillonite composite with xGnP, to improve the thermal conductivity for energy saving. This study also aims to investigate the effect of the xGnP addition on the dispersibility, thermal conductivity, and latent heat capacity of form stable PCM [26,27]. Also, to solve the leakage problem of PCMs, some investigators have studied the possibility of a container that can prevent the leakage of liquid PCMs, by using shape-stabilized PCM (SSPCM), microencapsulated PCM (MPCM), and incorporated PCM techniques [28-31]. Actually, paraffinic PCMs should be incorporated into porous materials, such as gypsum wallboard, plaster, concrete, clay minerals, and others [32]. Clay minerals have been used for years in many applications, such as nanocomposites [33], catalysts [34], adsorbents for removal of hazardous compounds [35], and supports for highly fluorescent probes [36]. The demand for clays in diverse scientific and technological areas lies in their low cost and natural abundance, high adsorption and absorption capacities, and fire retardancy, among other properties. Among clay minerals, the most common smectite clay mineral is montmorillonite (MMT). MMT evolves from volcanic ashes by weathering or hydrothermal effects, like other aluminum-rich minerals, and composes the highest part of the volcanic ash clay termed bentonite. The terminology of the word 'bentonite' can be summarized as follows: the rock term bentonite, which is commonly used for the smectite group minerals (sodium montmorillonite (Na), calcium montmorillonite, saponite (Mg), nontronite (Fe), and hectorite (Li)), is a clay material, altered from a glassy igneous material, usually volcanic ash. Therefore, this paper uses sodium montmorillonite (Na-MMT) as a container of PCM [37]. We prepared thermal enhanced paraffinic PCM/Na-MMT composite by compounding xGnP, to improve the thermal conductivity of the PCMs. They have a very high aspect ratio, comparable to that of carbon nanotubes. Drzal et al. successfully developed a microwave exfoliation and ultrasonic grinding process, to prepare exfoliated graphite nanoplatelets of different sizes and surface areas. These particles have been incorporated into different thermoplastic and thermoset materials, and PCMs, to improve the electrical, thermal and mechanical properties of nanocomposites [38,39]. In this study, we prepared thermal enhanced paraffinic PCM/Na-MMT, by using a vacuum impregnation process with xGnP. The vacuum impregnation method guarantees the high heat storage of paraffinic PCMs, due to capillary forces and surface tension forces during the incorporation process. The Na-MMT is needed for applying grouting materials of ground heat exchange system because the Na-MMT is used to grouting materials, originally. So we developed the ground heat energy storage composite through mixture of Na-MMT and paraffinic PCMs. And we enhanced thermal conductivity of Na-MMT/paraffinic PCM composites by mixing xGnP. And the thermal conductivity enhanced composite brought preventing super-cooling phenomenon of paraffinic PCMs. In this paper, we analyzed the microstructure, chemical bonding, heat capacity, thermal resistance and thermal conductivity of thermal enhanced paraffinic PCM/Na-MMT composites, from the results of Fourier transform infrared spectroscopy (FTIR), differential scanning calorimetry (DSC), thermogravimetric analysis (TGA) and TCi thermal conductivity analyses.

#### 2. Experimental

#### 2.1. Materials

This study used two types of liquid paraffinic PCMs, with different melting points. In the experiment, we used n-hexadecane and n-octadecane as PCM, which have 254.7 J/g and 247.6 J/g of latent heat capacity, and melting points of 20.84 °C and 30.4 °C, respectively. The n-hexadecane and n-octadecane are made of the alkane series, and belong to the paraffinic PCMs. The paraffinic PCMs were obtained from Celsius Korea, South Korea. xGnP is a graphitic carbon-based material. The graphite was obtained from Asbury Graphite Mills, Inc., NJ, USA, by applying a cost- and timeeffective exfoliation process initially proposed by Drzal's group [10]. xGnP, which combines the layered structure and low price of nanoclays with the superior mechanical, electrical and thermal properties of carbon nanotubes, is very cost effective, and can simultaneously provide a multitude of physical and chemical property enhancements [11-13]. Table 1 shows the physical properties of xGnP. The used Na-MMT has 8-12% free moisture, 8-14cc/2 g swelling, 0.75–0.85 g/cm<sup>3</sup> loose bulk density and more than 80% particle size (200 mesh pass). This Na-MMT was obtained from Ilsung chemical Co., Ltd. South Korea.

#### 2.2. Preparation

The paraffinic PCMs/Na-MMT with xGnP or without xGnP composites were prepared by an impregnation method in a vacuum, following the manufacturing process. The preparation of paraffinic PCMs based composites with xGnP and without xGnP is almost the same. So we only describe the preparation process of paraffinic PCMs based composites with xGnP. The detailed preparation process is as follows: 5 wt% of xGnP according to weight percentage of Na-MMT was mixed in Na-MMT, before the vacuum process. The xGnP/Na-MMT mixture was placed inside a filtering flask, which was connected to a water tromp apparatus, to evacuate air from its porous surface. Then, the valve between the flask and the container of 200 g of liquid paraffinic PCMs was turned open, to allow it to flow into the flask, to cover the xGnP/Na-MMT mixture. After the vacuum process was continued for 90 min, air was allowed to enter the flask again, to force the liquid paraffinic PCMs to penetrate into the pore space of the xGnP/Na-MMT mixture. To find the maximum PCM amount in the xGnP/Na-MMT mixture, we impregnated 200 g of paraffinic PCMs into xGnP/Na-MMT mixture. In this case, no impregnated excess of paraffinic PCMs remained in the flask. Therefore, we needed to remove the excess of paraffinic PCMs in the flask through filtering. The colloid state of SSPCM was filtered by 1 µm filter paper, until a granule type of sample appeared on the filter paper. Then the granule type of thermal enhanced SSPCM was dried in a vacuum drier, at 80 °C for 48 h.

| Table 1   Physical properties of xGnP. |              |
|--|--------------|
| Surface area (m <sup>2</sup> /g)       | 20.41        |
| Bulk density (g/m <sup>3</sup> )       | 0.0053-0.010 |
| Pore volume (cm <sup>3</sup> /g)       | 0.081        |
| Thermal conductivity (W/m K)           | 2-300        |
| Specific heat capacity (I/kg K)        | 710          |

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