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Single-walled carbon nanotube for shape stabilization and enhanced phase change heat transfer of polyethylene glycol phase change material

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ABSTRACT

In order to simultaneously stabilize the shape and enhance the heat transfer efficiency of a typical polyethylene glycol (PEG) phase change material (PCM), a family of novel nanocomposites consisting of single-walled carbon nanotubes (SWCNs) and PEG was tailor-made via a facile impregnation method. The effects of SWCNs loadings ranging from 2% to 10% on the chemical structure, thermal performance and heat transfer enhancement of PEG were investigated experimentally. The fabricated nanocomposite exhibits high adsorption of PEG as high as 98% and can completely preserve its original shape without any PEG leakage even when subjected to a 400 melt-freeze cycle. The maximum adsorption of PEG is the highest value in literature up to now. The melting point of PEG/SWCNs nanocomposite shifts to a lower temperature while the freezing point shifts to a higher temperature while compared to the pure PEG, resulting in a substantial reduction of the supercooling degree. Above all, the thermal conductivity was found to increase with SWCN loadings whether in solid or liquid state. In detail, only with a small SWCN loading of 4%, a dramatically high, 375% enhancement is obtained in the solid state and a relatively high enhancement of 121% is achieved in the liquid state.

1. Introduction

Today, the consumption of resources is increasing, especially that of fossil fuels. The increasing energy shortage and grave environmental crisis have motivated the discovery of renewable and sustainable energy sources [1]. Energy storage technology (EST) can not only boost the performance and long-term reliability of the energy system, but also provide a bridge between energy supply and demand, which will in turn reduce the impact on the environment. Now it has become the research hotspot of energy utilization [2]. Among multifarious EST methods, thermal energy storage technique is one such technology which can substantially reduce the total energy consumption and conserve the indigenous fossil fuels resource [3]. There are particularly three methods for energy storage: latent heat storage, sensible heat storage and chemical reaction heat storage [4]. Among these methods, latent heat storage technology (LHST) using phase change material (PCM) as promising medium can be designed to have higher heat capacity and smaller temperature fluctuation, which is particularly attractive and has been proved to be the most efficient way to improve the energy utilization efficiency [5].

Although traditional PCMs like polyethylene glycol (PEG) exhibit desirable properties, however, they often suffer from leakage during the solid-liquid phase transition and exhibit low thermal conductivity. This property reduces the thermal response of the thermal protection system and thereby may cause system overheating [6,7]. To address the above issues, considerable efforts have been devoted to introducing appropriate supporting materials to develop form-stable composite PCM. Form-stable composite PCM is similar to a certain kind of solid–solid PCM, which can maintain its original shape and prevent leakage throughout the phase change process [8]. Previous study once indicated that dry powders with extremely large specific surface areas could be used to package the PCM, preventing PCM leakage in the melt state. Various supporting materials, including polymers, clay minerals (diatomite, vermiculite, perlite, kaolin, etc.), metal foam, expanded graphite, and silica materials have been widely reported [9,10].

In our previous work, a series of PEG/diatomite form-stable composite PCMs have been successfully fabricated. In these composites, PEG was tightly impregnated into the diatomite pores with

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excellent long-term durability. Ag nanoparticles and expanded graphite were subsequently added to accelerate the heat transfer rate [11,12]. We call it a "two-step method": the first step is to guarantee the shape stability; the second step is to improve the thermal conductivity by embedding high thermal-conductive materials [13,14]. It's our understanding that most studies applied this "two-step method" to solve the problem of leakage and low thermal conductivity. Little information on applying "one-step method" is available.

In order to hold back the leakage of PCM and improve its thermal conductivity all at once, high thermal-conductive and form-stable composite PCMs (HTFSCPs) were widely researched. As known, carbon family materials such as carbon fiber, carbon nanotubes, and expanded graphite are considered as the most promising fillers for enhancing thermal conductivity since only a small mass fraction is needed due to their high thermal conductivity and low density [15,16]. In addition, the great specific surface areas of carbon nanomaterials enable it to coat the PCM and prevent the PCM leakage.

Carbon nanomaterial, single-walled carbon nanotube (SWCN) in particular, has attracted great interests and was widely studied as an attractive candidate for LHST applications, due to its fascinating quasi-one-dimensional nanostructure, remarkably high intrinsic thermal conductivity, high surface area-to-volume ratio, and superior mechanical properties, which will contribute to form the uniform network structure in PCM and increase the heat transfer rate in applications with weight and volume constraints [17,18]. However, up to now, related studies and experimental results of the effect of carbon nanomaterials on PEG PCMs are not plenty and lake of details, not to mention the systematic study of the potential effect of SWCNs on the thermophysical properties of PEG PCM. In addition, the homogeneous dispersion of nanoparticles here I mean SWCNs may not be stable after the long-term use. This is because appreciable thermal interface resistance between PEG and SWCNs may weaken its effectiveness [19]. Of the utmost importance is to form a percolating network of the conductive SWCNs in order to obtain a PCM nanocomposite with a high thermal conductivity, less SWCNs content and excellent durability.

In this paper, we have introduced a novel series of high thermal-conductive and form-stable composite PCMs (HTFSCPs) using SWCNs as supporting material by the conventional melting dispersion method. The thermal conductivity enhancement varies significantly among different SWCNs loadings and states of the PEG during the solid–liquid phase change process. The inherent thermal properties of the prepared HTFSCPs including the thermal storage performance, heat enthalpies, thermal conductivities, stability and reliability were investigated experimentally, making the PEG nanocomposite stands distinct with reference to the previous literatures. The research results provide valuable reference for the future practical application of energy storage.

2. Experimental

2.1. Materials

In our work, PEG (Mw = 6000, ρ = 1.27 g/mL, λ = 0.24 W/m K, c = 2.53 kJ/(kg °C)) was used as PCM and purchased from Beijing Chemical Reagent Ltd., China. Single-walled carbon nanotubes (SWCNs, ρ = 1.42 g/mL, λ = 4000 W/m K) were used as the supporting material to improve its thermal conductivity and shape stability. It was supplied by Xilong Chemical Reagent Beijing Co., Ltd. China. Toluene solvent was supplied by Beijing Chemical Reagent Ltd., China. The above reagents were all of analytical grade.

2.2. Preparation of PEG/SWCNs HTFSCPs and the leakage test

A series of PCM nanocomposite were tailored by a facile melt-mixing method and the corresponding schematic illustration is presented in Fig. 1(a). The whole fabrication procedure was started by dissolving 5 g of PEG into 100 ml toluene at 80 °C. After that, SWCNs powders with the elevated mass fractions from 2% up to 10% were added with an increment of 2%. The suspension was treated under the strong shear mixing using a magnetic stirrer for 20 min, followed by an intensive ultrasonication for 30 min at power of 180 W to reduce the SWCNs aggregates and obtain a homogeneous dispersion of the nano particles. Then, the mixture was left inside a fume hood at 120 °C to evaporate the toluene. Next, the PCM nanocomposite samples were dried at 80 °C inside a vacuum oven overnight to eliminate the liquid PEG which had not been adhered to SWCNs. Finally, the energy storage nanocomposites made of PEG and SWCNs were obtained. The nanocomposites with 2–10% SWCNs were named S1–S5, respectively.

The leakage test was conducted as follows in Fig. 1(b). All the specimens (S1–S5) were first put on the different filter papers. After that, each specimen along with the filter paper was placed in a drying oven with the temperature controlled above the melting point of PEG (80–120 °C) for 1 h, to make sure that PEG inside was melted. After taking out and cooling to the room temperature, a careful examination on the filter papers was carried out to find out the trace of the melted PEG. Besides, the mass loss was recorded to further estimate the leakage of PEG. As seen in Fig. 1 (b), at the testing temperature of 80 °C, no liquid PEG was observed on the surface of the PEG/SWCNs nanocomposite and filter paper was visually dry. Besides, the mass loss during the testing process can be neglected. Here, similar results have been obtained at other testing temepratures. Thus, PEG/SWCNs nanocomposite fabricated in this study is well shape-stablized. The maximum impregnation ratio of PEG in the nanocomposite could be determined as the absorption amount when no liquid PEG trace was observed even when heated above its melting temperature. In our study, the form-stable PEG/SWCNs nanocomposite PCM was tailored at the designed PEG impregnation percentage of 98%, which is the highest value in the literature till now.

2.3. Analysis methods

2.3.1. Structure observations of the prepared PEG/SWCNs HTFSCPs

The specific surface area and pore volume of SWCNs were determined by a N_2 adsorption analyzer (Quantachrome Instruments, US). X-ray photoelectron spectroscopy (XPS) (ESCALAB 250Xi) was carried out to further analyze the chemical composition of SWCNs. Raman spectrums were obtained by using a dispersive Raman spectrometer (Nicolet, ALMEGA) with the excitation wavelength of 532 nm.

The microstructures of SWCNs and PCM nanocomposite were characterized by scanning electron microscopy (SEM, S-4800, Hitachi, Japan) and transmission electron microscopy (TEM, JEOL JEM-2010, Japan, accelerating voltage 200 keV).

The chemical compatibility of the prepared heat storage nanocomposites was obtained via Fourier transform infrared spectroscopy (FT-IR, Model Frontier). The FT-IR analysis was recorded in transmittance mode at room temperature from 400 to 4000 cm^{-1} using KBr pellets.

2.3.2. Thermal analysis

Thermal properties in terms of latent heat capacity and phase transition temperature for pure PEG and the prepared PCM nanocomposites were tested using differential scanning calorimeter (DSC, Q2000). In detail, the phase transition temperatures and