



Thermal conductivity enhancement of polyethylene glycol/expanded vermiculite shape-stabilized composite phase change materials with silver nanowire for thermal energy storage

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HIGHLIGHTS

- EVM benefited the encapsulation of PEG and decreased its supercooling extent.
- A theoretical calculation method was applicable to predict the thermal conductivity enhancement ability of Ag NW.
- Latent heats and thermal conductivity of PEG–Ag/EVM ss-CPCMs maintained simultaneously reasonable.

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ABSTRACT

A series of novel polyethylene glycol–silver nanowire/expanded vermiculite shape-stabilized composite phase change materials (PEG–Ag/EVM ss-CPCMs) were prepared by physical blending and impregnation method to overcome liquid leakage during phase transition and enhance the thermal conductivity of PEG. In these PEG–Ag/EVM ss-CPCMs, PEG served as the phase change material for thermal energy storage; Ag NW served as thermal conductivity enhancement filler; EVM acted as the supporting material to provide structural strength and prevent the leakage of melted PEG. SEM analysis results indicated that Ag NW wrapped with PEG was well dispersed and enwrapped inside the pores and surfaces of EVM due to capillary force and surface tension. It was found that the maximum encapsulation capacity of PEG in all PEG–Ag/EVM ss-CPCMs with good shape stability was 66.1 wt.%. The thermal conductivity of PEG–Ag/EVM ss-CPCMs could be greatly enhanced by the prepared Ag NW with a length of 5–20 μm and a diameter of 50–100 nm. A theoretical calculation method was developed to predict and evaluate the thermal conductivity enhancement ability of Ag NW. The predictions were consistent with experimental results. The thermal conductivity of PEG–Ag/EVM ss-CPCMs19.3 reached 0.68 W/m K, which was 11.3 times higher than that of pure PEG, and corresponding phase change latent heat was 96.4 J/g. The supercooling extent of PEG in PEG–Ag/EVM ss-CPCMs decreased approximate 7 °C because the EVM could act as a heterogeneous nucleation center to promote the crystallization of PEG. FT-IR and TGA results showed that the PEG–Ag/EVM ss-CPCMs exhibited excellent chemical compatibility and thermal stability.

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1. Introduction

Latent heat storage technology with phase change materials (PCMs) is considered as one of the most efficient technologies to store and release large amounts of thermal energy. Compared to sensible heat storage materials, the PCMs provides the higher heat storage densities and nearly isothermal behaviors [1,2]. Nowadays,

PCMs have been applied in many fields, such as building energy conservation, solar energy utilization, waste heat recovery, and other thermal energy storage systems [3,4].

According to the material properties, the PCMs can be classified into two major categories: organic and inorganic compounds. As promising solid–liquid organic PCM, polyethylene glycol (PEG) has adjustable and suitable phase change temperature, high phase change latent heat, excellent chemical and thermal stability, low vapor pressure, and low cost and shows no toxicity, corrosiveness, or phase separation [5–7]. However, the PEG has low thermal

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conductivity and liquid leakage during phase transition [5,8]. Low thermal conductivity of PCM strongly suppresses the energy charging/discharging rates, thus severely limiting its application scope. Liquid leakage of melted PCM is harmful to the surroundings of energy storage systems and may lead to potential dangers.

Thermal conductivity of PCMs has been enhanced in two ways: impregnating PCMs into porous supporting materials with high thermal conductivity, such as expanded graphite and mesoporous silica sphere [9–11], and dispersing thermal conductivity enhancement filler into PCMs, such as carbon nanotubes [12], graphene nanomaterials [5,13], activated carbon [14], carbon fiber and metallic/oxide nanoparticles [15–17]. Several reports indicated that the latter was an effective and promising way to enhance the thermal conductivity of PCM. Qi et al. [5] reported that the thermal conductivity of PEG/graphene oxide (GO)/graphene nanoplatelets (GNP) composite PCM with 2 wt.% GO and 4 wt.% GNP achieved 1.72 W/m K. Qian et al. [17] discovered that the thermal conductivity of PEG/diatomite PCM containing 7.2 wt.% Ag nanoparticles reached 0.82 W/m K, which was 127% higher than that of the PEG/diatomite composite. However, an available theoretical calculation method should be developed to predict and evaluate the thermal conductivity enhancement ability of filler. In addition, one-dimensional metal nanowire could enhance the thermal conductivity of fluids because of high thermal conductivity and rapid heat transport along the relatively long distance [18]. Hence, metal nanowire was also applied for the purpose of thermal conductivity enhancement of PCM. Among all the metal nanowires, silver nanowire (Ag NW) exhibits the highest thermal conductivity (429 W/m K). Moreover, the large specific surface area of Ag NW is also beneficial to the adsorption of PCM. Therefore, Ag NW is considered as the most promising and effective thermal conductivity enhancement filler. The effects of Ag NW on thermal performance of PCM were seldom reported.

Shape-stabilized composite phase change material (ss-CPCM) composed of working substance and porous supporting materials was prepared to prevent liquid leakage of PCM during phase transition [19,20]. Various ss-CPCMs have been prepared by impregnating PCM into porous supporting materials, such as expanded vermiculite (EVM) [21–23], expanded perlite [23,24], diatomite [25,26], graphene oxide [27], and expanded graphite [9,10]. Among all the supporting materials, EVM is a hydrous phyllosilicate mineral and its volume can be expanded by 8–30 times depending on thermal conditions [28,29]. Hence, EVM is highly porous and lightweight due to expanded microstructures, which allows high heat storage capacities of PCM. Chung et al. [23] reported that the maximum load of n-octadecane in EVM reached 80 wt.% without any liquid leakage during phase transition. Guan et al. [22] prepared paraffin/EVM ss-CPCM, the paraffin adsorption with good shape-stability reached 64.6 wt.%. EVM is considered as one of the most suitable supporting materials of ss-CPCM for thermal energy storage due to its high heat storage capacities, chemical inertness, ecological friendliness, and low cost. In our study, it is expected that the EVM has large encapsulation capacity of PEG.

This paper aims to overcome liquid leakage during phase transition and enhance thermal conductivity of PEG by impregnating Ag NW wrapped by PEG into the pores of EVM. In this study, the prepared Ag NW was employed to enhance the low thermal conductivity of PEG. The PEG achieved satisfactory thermal conductivity while maintaining reasonable latent heat by adjusting the ratio of PEG and Ag NW. Moreover, a theoretical calculation method was developed to predict and evaluate the thermal conductivity enhancement ability of Ag NW. The microstructure, chemical compatibility, crystalline phase, thermal energy storage properties, thermal conductivity, and thermal stability of the prepared PEG–Ag/EVM ss-CPCMs were also evaluated in detail.

2. Experimental

2.1. Materials

Chemically pure polyethylene glycol (PEG) with an average molecular weight of 4000 was purchased from Xilong Chemical Reagent Beijing Co., Ltd. China. Ethylene glycol (EG, A.R.), silver nitrate (AgNO_3 , >99.8%, A.R.), polyvinylpyrrolidone (PVP, K-90, Molecule weight = 630,000) and sodium chloride (NaCl , >99.5%, A.R.) were purchased from Beijing Chemical Reagent Co., Ltd. EVM (Table 1) was obtained from Hebei Lingshou. The specific surface area and average pore diameter of EVM were measured to be 4.153 m^2/g and 138.70 nm, respectively.

2.2. Preparation of PEG–Ag/EVM ss-CPCMs

The preparation procedure of PEG–Ag/EVM ss-CPCMs included two steps: the synthesis of Ag NW and the preparation of PEG–Ag/EVM ss-CPCMs (Fig. 1). In Step 1, Ag NW was synthesized through a simple solvothermal method by reducing AgNO_3 with EG as both solvent and reducing agent in the presence of PVP as an adsorption agent. The detailed synthesis process was drawn as follows: (1) 20 mL of EG solution of 0.3 M AgNO_3 was vigorously stirred after adding 20 mL of EG solution containing 2.76 mM NaCl with an injector. The obtained AgCl could serve as seeds for heterogeneous nucleation and growth of silver particles. (2) 20 mL of EG solution containing 0.6 M PVP was added to the above mixed solution with magnetically stirring. (3) The mixture was transferred to a 150-mL Teflon-lined stainless steel autoclave and maintained at 160 °C for 5 h. (4) The product was washed, centrifuged, and dispersed in ethanol. In Step 2, the PEG–Ag/EVM ss-CPCMs were prepared by a physical blending and impregnation method. The detailed preparation process was drawn as follows: (1) The Ag NW dispersed in ethanol was dispersed in the melted PEG while stirring at 70 °C for 12 h. (2) The EVM was added into the above mixture and the temperature was maintained above the melting point of PEG at 65 °C for 10 h. The PEG would be forced to impregnate into the pore spaces of EVM due to capillary force and surface tension. (3) The impregnated sample was transferred onto filter paper and put into a drying oven at 70 °C to remove the leaky liquid PEG from EVM. Filter paper was replaced continuously until no liquid leakage trace was observed. Four types of PEG–Ag/EVM ss-CPCMs were obtained (Table 2). The maximum adsorption mass fractions of PEG retained in the pores of EVM reached 66.1 wt.%, 64.7 wt.%, 62.1 wt.%, and 58.8 wt.% and the corresponding ratio of Ag NW was 0, 7.1 wt.%, 13.7 wt.%, and 19.3 wt.%.

2.3. Characterization

X-ray diffraction (XRD) patterns were acquired with an X-ray diffractometer (DMAX 2400, Rigaku, Japan) under Copper $K\alpha$ radiation ($\lambda = 0.1541$ nm, 40 kV and 100 mA) at a scanning rate of 8°/min in the 2θ range of 0–85°. The crystal structures and crystallization properties of PEG in the pores of EVM was investigated. Infrared spectra of the PEG and ss-CPCMs were obtained with Fourier transform infrared spectroscopy (FT-IR, FTIR8400, SHIMADZU, Japan) to investigate chemical compatibility between PEG and EVM. Samples were ground sufficiently and mixed with KBr. The range of wavenumber was 4000–500 cm^{-1} . The morphology of samples was observed by using scanning electronic

Table 1
Chemical contents of the expanded vermiculite (wt.%).

Constituents	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	K_2O	H_2O
Ratio (wt.%)	43.2	12.68	4.56	24.2	0.96	5.95	7.6

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