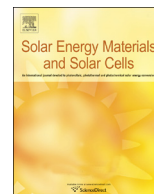




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Experimental and numerical study on the effective thermal conductivity of paraffin/expanded graphite composite



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ABSTRACT

Expanded graphite (EG) powder as a typical porous material has high thermal conductivity and large surface area that are often used to enhance the thermal conductivity of phase change materials (PCMs). Ten composites of paraffin/EG powder with various weight fractions were elaborated by high pressure compression. Their microstructure configuration and thermal properties were characterized with Scanning Electron Microscopy (SEM), Thermal Constant Analyzer. The saturation sorption capacity of EG under compression was experimentally determined. The thermal conductivity of the paraffin/EG composite is greatly enhanced by 41 times in maximum as compared to pure paraffin. Based on the experimental quantification, we propose a novel two-level scale model from micro-scale to macro-scale to compute the thermal conductivity of the composite. The numerical results agree well with the experimental data that validate the precision and reliability of the proposed model. Furthermore, the results reveal that both keeping porous EG structure at original morphology and increasing the saturated sorption capacity of paraffin in EG could eventually improve the heat transfer capacity of the composites. In addition, the proposed model would require more experimental validation based on the composites with different materials in future work.

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1. Introduction

Thermal energy storage (TES) using phase change materials (PCMs) has received a considerable attention over the last decades [1–5]. The latent heat provides a high heat storage density, and significantly reduces the storage material volume and the temperature difference between PCM and heat transfer fluid (HTF) [6]. Organic PCMs are typical thermal storage materials with the advantages of low cost, stability, high storage density, non-toxicity and non-corrodibility [7]. However, the extant organic PCMs have some unresolved problems such as poor thermal conductivity, typically no more than 0.2 W/(m K) [9], which restricts their wide applications by reducing the rate of heat charging and discharging during the melting and solidification period [8]. To date, various techniques have been developed to overcome the limitations mentioned above to increase the heat transfer rate as summarized in the literature [10], such as embedding PCMs into the porous matrices [11–13], adding carbon fibers or carbon fiber brushes [14], and dispersing highly thermal conductivity particles within the PCMs [8,15].

Expanded graphite (EG) materials as a porous matrix has been inserted into PCMs due to their high thermal conductivity (about 200 W/(m K)), chemical stability and bargain price. Being around 90% volume of EG connected honeycomb holes, this material has a higher porosity for increasing absorbability and a larger specific surface area than that of graphite. EG with these desirable properties hence has attracted researchers paying more attentions in developing high heat performance of TES [12,16–21].

EG matrix connected with struts and pores has a high effective thermal conductivity. Py et al. [12] studied the heat transfer behavior of the composites paraffin/EG matrix, in which the paraffin was impregnated into the matrix by capillary action. The thermal conductivities of these composites were found as high as 70 W/(m K), depending on the effective graphite density and orientation. Pincemin et al. [18,19] tested the thermal conductivity for integrating graphite with inorganic PCMs with high melting points at both laboratory and industrial scales within a high melting temperature range. The results from the work [18] show the thermal conductivity relies on not only the amount of the graphite but the particle size, they hence proposed for a greater mass percentage of graphite in the composite a larger particle size should be employed. They also found the composite has a nearly constant melting point, which is an advantage for practical application in a nearly isothermal operation. The effects of three

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Nomenclature

C	given total volume fraction (%)
T	temperature ($^{\circ}\text{C}$)
W	weight fraction (%)
V	volume fraction (%)
δ	thickness of sheets (μm)
λ	thermal conductivity ($\text{W}/(\text{m K})$)

Subscripts

c	composite
EG	expanded graphite
exp	experimental
num	numerical
p	particle in microscale
$para$	paraffin
T	total in the composite

elaboration routines for graphite integrated with PCMs were tested by Pincemin et al. [19]. They found that the cold compressed composites presented highly anisotropic properties and strong intensification in thermal conductivity (a factor of 31 at 200°C for 20 wt% graphite in composite), compared to the compound composites made by dispersion of graphite within molten salts showed isotropic properties and medium thermal conductivity (a factor of 10 for 7 wt% graphite in composite). However, the cold compressed composites suffered from important salt leakage resulted from the impurities and remaining mechanical stress. Meanwhile, the infiltration route was demonstrated to be inefficient in filling the PCM into the EG. The permeability of EG matrix was experimentally studied by Wang et al. [22,23]. They found the permeability decreases while the density of EG matrix increases as higher pressure imposed on the EG matrix sample. The reason is that the reduced mass transfer inside the EG matrix with larger density has a higher resistance for the mass transfer process, hence the permeability is lowered.

EG powder has a lower effective thermal conductivity than that of EG matrix because of its unconnected graphite structure, while has a higher porosity and larger specific surface area. It is more important that the elaboration way of mixing dispersive EG powder to the molten PCMs above the melting temperature is much easier than that of cold compression or evacuation infiltration for interconnected graphite matrix. Thus, EG powder might have a higher potential value than EG matrix for enhancing heat transfer of PCMs in real applications.

Experimental methods are capable of testing the positive effects of EG materials, but are limited to provide a clear insight into the heat transfer mechanisms inside the composites of PCMs/EG powder. Efforts have been spent on developing numerical methods based on rational physical models and computational approaches. The characteristics of composites such as complicated porous structure, irregular dispersion and discontinuous graphite flake, however make numerical methods hard and complex to predict the effective thermal conductivity of the composites with EG in a wide-spread weight fraction. Some empirical models or simplified assumptions have been adopted to study the heat transfer behaviors in porous media with connective structure [24–28]. Calmidi and Mahajan proposed a model for predicting the effective thermal conductivity in their earlier work [29], which not only involves the porosity but the geometric structures of the porous material. This model gave a nearly perfect agreement with the experimental results [30]. Nevertheless, as far as we know, very few open references have ever carried out the numerical analysis on the thermal conductivity of the composite with non-connective structure, such as EG powder dispersed into paraffin.

Our current works have two intentions: the first is to experimentally explore a simple and practical technique to prepare the composites of paraffin/EG, then the effect of microstructures of the composites on the thermal behavior is quantified; in light of the experimental quantification on the material properties, a novel numerical analysis approach is proposed to predict the effective

conductivity of the composites. By comparing the calculation results to experimental data, the concept of the computation method is verified and validated.

2. Material and methods

2.1. Materials preparation and PCM/EG elaboration

EG was prepared from expandable graphite powder (Fig. 1). The expandable graphite powder is made by 50 mesh large flake graphite ore and higher than 99% carbon content. After the drying expandable graphite powder was placed in muffle furnace at 1000°C for 30 s, we got the expanded graphite (EG) with worm-like structure (Fig. 2a). The structure magnified by 50 times was captured with scanning electron microscopy (SEM) (Fig. 2b), in which flakes evenly intercalate on the surface to produce massive micropores with the dimensions down to several microns (Fig. 2c). These micropores thus create a high specific surface area and a larger volume.

We currently used paraffin (Sinopharm Chemical Reagent Co., Ltd.) with melting temperature at $58\text{--}60^{\circ}\text{C}$ as the PCM, due to it is safe, predictable, less expensive and non-corrosive. The massive paraffin was firstly cut into tiny pieces. Then these pieces were mixed with EG powder at room temperature in a mixer under low rotation speed (200 rpm) to avoid destroying the fine structure of EG. When the paraffin was evenly distributed in EG powder, the mixing work was continued on a heating plate at 80°C till the micropores of EG was full of the melting paraffin. Fig. 3 shows the dispersive solid composite of paraffin and EG powder, in which the paraffin was well infiltrated into EG and the composite keeps the same worm-like structure as that of EG powder.

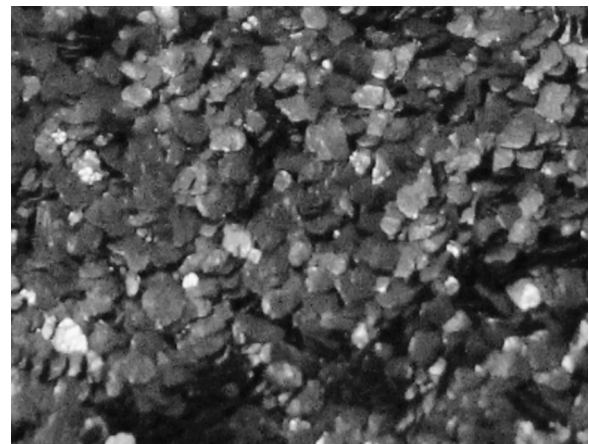


Fig. 1. Expandable graphite powder.

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