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Thermal characteristics of expanded perlite/paraffin composite phase change material with enhanced thermal conductivity using carbon nanotubes

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A B S T R A C T

Paraffins constitute a class of solid-liquid organic phase change materials (PCMs). However, low thermal conductivity limits their feasibility in thermal energy storage (TES) applications. Carbon nano tubes (CNTs) are one of the best materials to increase the thermal conductivity of paraffins. In this regard, the present study is focus on the preparation, characterization, and improvement of thermal conductivity using CNTs as well as determination of TES properties of expanded perlite (ExP)/*n*-eicosane (C20) composite as a novel type of form-stable composite PCM (F-SCPCM). It was found that the ExP could retain C20 at weight fraction of 60% without leakage. The SEM and FTIR analyses were carried out to characterize the microstructure and chemical properties of the composite PCM. The TES properties of the prepared F-SCPCM were determined using DSC and TG analyses. The analysis results showed that the components of the composite are in good compatibleness and C20 used as PCM are well-infiltrated into the structure of ExP/CNTs matrix. The DSC analysis indicated that the ExP/C20/CNTs (1 wt%) composite has a melting point of 36.12 °C and latent heat of 157.43 J/g. The TG analysis indicated that the F-SCPCM has better thermal durability compared with pure C20 and also it has good long term-TES reliability. In addition, the effects of CNTs on the thermal conductivity of the composite PCM were investigated. Compared to ExP/C20 composite, the use of CNTs has apparent improving effect for the thermal conductivity without considerably affecting the compatibility of components, TES properties, and thermal stability.

1. Introduction

Rapid depletion of carbon based fossil energy sources and the continuous enlargement of the gap between energy supply and demand are forcing the researchers to develop new sources of energy or to store the available energy. Thermal energy storage (TES) using phase change materials (PCM) is an efficient method of storing excess energy, a clean method, and has received significant attention of the researchers and energy engineers [1]. Known as latent heat storage materials, PCMs are promising materials for storing and releasing large amount of energy. Therefore, latent heat thermal energy storage (LHTES) system can store a considerable amount of heat with a very little temperature drift. Organic PCMs including fatty acids and paraffins are considered to be the best materials suitable for latent heat storage due to their thermal and chemical durability, non-toxicity, and little/no subcooling properties [2–4]. However, these PCMs possess extremely low thermal conductivity (0.1–0.3 W/m K) which limits their use in many industrial and domestic applications due to the low heat storage/release rate. Insertion of metal matrix such foam or dispersion of micro/nano particles of metals and their oxides have been a proven way to increase the conductivity of organic PCMs [5]. Dispersion of micro size particles causes sedimentation and this problem can be resolved by dispersing nano size of particles in the PCM [6]. Nano sized particles have the ability to enhance the thermal conductivity of base PCM significantly [7–9]. Apart from a number of advantages using nano particles as thermal conductivity enhancer, they have some disadvantages, too. Addition of the nanoparti-

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cles increases the weight and volume of TES system considerably. Such composites also show the poor thermal and chemical stability due to the difference in volume of nanoparticles and base organic PCM [10]. Moreover, thermal conductivity enhancer, carbon-based composite materials such as carbon nanofibers (CNFs) [11–17], and graphene/graphene nano platelets [18–22] have been preferred as alternative for nanoparticle of metal or metal oxides. In those studies, significant increase was reported in the thermal conductivity of PCMs after adding carbon-based materials.

On the other hand, carbon nanotubes (CNTs) are effective materials for the enhancement of heat storage/release performance of PCMs due to their high thermal conductivity in the range of 2000–6000 W/m K [23]. Also, some superior properties of CNTs, such as good dispersion ability due to low density, large surface, and high stability make them great promising matrices to increase the thermal conductivity of PCMs [24,25]. In this regard, many studies have been conducted to investigate the effect of CNTs on the TES properties of different PCMs. Li et al. [26] worked for nano-composite of stearic acid (SA) and multi-walled carbon nanotube (MWCNT). They reported that, compared with the pure SA, the melting temperature of SA/MWCNT nano-composite shifted to a lower value during the storing phase, and freezing temperature shifted to higher value during the releasing phase. Wang et al. [27] prepared a composite of paraffin wax and different weight fraction of MWCNT, and investigated their thermal properties. Their results showed that the melting point of composite reduces and thermal conductivity increases with increasing amount of MWCNT in paraffin wax. The effect of CNTs on the LHTES properties of PA-SA eutectic mixture for energy storage was studied experimentally [28]. Thermal conductivity of polyethylene glycol as organic PCM was enhanced using functionalized CNTs [29]. In another study, the effect on the thermal conductivity of palmitic acid using MWCNTs treated with different methods was investigated. The results were showed that the type of treatment processes significantly affected the interfacial thermal resistance between the MWCNTs and the palmitic acid. Also, the MWCNT surface status was reported to have significant role on the thermal conductivity increments of the composites [30].

On the other hand, other problem regarding with paraffin is the additional need for packing container due to leakage problem of liquid phase over their melting temperatures. Thus, the cost of LHTES system including paraffins as PCMs increases considerably in case of their usages together with plastic or metal containers. In this regard, porous and lightweight construction materials are very suitable for the encapsulation of paraffins especially for solar passive TES purposes in buildings. Such a matrix type allows successful impregnation of paraffins in specific amounts without observing any leakage during the cycling heating treatments. In order to obtain this kind of energy storage composite PCMs, several paraffins have been encapsulated in form-stable mass fractions into different porous matrices such as diatomite, expanded perlite, and vermiculite as host matrix [31–36]. Among these, perlite is an amorphous volcanic glass possessing high porosity and very low density. It also shows high thermal stability and it is relatively cheaper [37,38]. Perlite can expand 7–16 times over its original volume if heated between 760 °C and 1100 °C [39], and it is thermal and acoustical insulator. The low density and light weight characteristics of expanded perlite (ExP) makes it one of the best retainer materials suitable for preparing form-stable composite PCMs (F-SCPCMs) for TES purposes [40,41].

This study is focused on solving the mentioned above-disadvantages of *n*-eicosane (C20) as a paraffin type PCM by its impregnation into ExP and increasing its thermal conductivity using CNTs. To our knowledge, this is the first study on the preparation, characterization and improvement of thermal conductivity of form-stable ExP/C20 composite PCM including CNTs. The structural and morphological characterizations of the fabricated F-SCPCMs were carried out using SEM and FTIR analysis techniques. The LHTES properties of the F-SCPCMs were measured using DSC technique. The effect of the mass fraction of CNTs (0.3, 0.5 and 1 wt%) on the LHTES, thermal conductivity and thermal storage/release rate of the composite PCMs were analyzed. The LHTES reliability was also investigated by conducting accelerated thermal cycling test. The thermal resistance of the produced F-SCPCMs against to temperature was determined by TG analysis. Especially the fabricated ExP/C20/CNTs(1%) composite can be considered as promising F-SCPCM for passive solar TES applications with purposes of heating, ventilating and air conditioning (HVAC) in building envelopes, greenhouses, and heat recovery systems.

2. Materials and methods

2.1. Materials

Eicosane (C20) used as organic PCM in this study was obtained from Sigma-Aldrich Company. ExP was obtained from Izper Company (Izmir/Turkey). The physical properties and chemical composition of the ExP was reported in literature [31]. Before use, it was dried at 105 °C for 24 h to remove the humidity and then sieved from 100 mesh. The CNTs used as thermal conductivity enhancer was supplied from Sigma-Aldrich Company (O.D. × L 6-9 nm × 5 μm, assay: 95% (carbon) and density:2.1 g/mL at 25 °C).

2.2. Preparation of ExP/C20/CNTs composite PCM

The preparation of ExP/C20/CNTs composites was carried out at two steps. In the first step, the ExP/C20 composites were prepared using vacuum impregnation method. For this process, dry ExP sample at a specified amount was placed into a flask integrated with vacuum system. The vacuum procedure was kept for 90 min under the pressure of 65 kPa. After that, C20 in liquid state was added gradually to the surface of ExP by using a funnel, the vacuum was stopped and air was allowed for 30 min to enter inside enabling the infiltration of C20. The fabricated composite was cooled for 24 h to solidify C20 completely into the pores of ExP. All of these operations were applied for the fabrication of all ExP/C20 combinations by changing the mass fraction of C20 from 30 to 70%. The maximum holding ratio of C20 into ExP was determined by applying leakage test to each composite. In the test process, the prepared composite sample was heated on a heater platform at 50 °C for 60 min to check seepage behavior of paraffin in melted state. The sample showed no seepage behavior was described as form-stable composite PCM (F-SCPCMs). The maximum mass fraction of C20 into the prepared F-SCPCMs corresponds to 60 wt%.

In the second step, ExP/C20/CNTs composites were prepared. For this process, the specified amount of CNTs was dispersed in acetone under intensive ultrasonic treatment. Then, previously prepared form-stable ExP/C20 composite was added to CNTs/acetone suspension. To guarantee the dispersion of CNTs uniformly into ExP/C20 composite, the suspension was continually stirred for 3 h with a magnetic stirrer. In order to remove the acetone thoroughly, the obtained mixture was maintained in an oven at 60 °C for 6 h. Three kind of composite PCM were prepared ExP/C20(60%)/CNTs (0.3%), ExP/C20(60%)/CNTs(0.5%) and ExP/C20(60%)/CNTs(1%) by arranging the amount of the added CNTs. Fig. 1 shows the leakage test results obtained for the final composite samples with impregnation ratio of 55, 60 and 65 wt%. As clearly seen from the photographs, the composite PCM did not show any leakage behavior as long as the impregnation ratio of C20 was equal to or lower as the form-stable combination ratio, 60 wt%.