Energy Conversion and Management 82 (2014) 229–237

Contents lists available at ScienceDirect

Energy Conversion and Management

journal homepage: www.elsevier.com/locate/enconman

Thermophysical properties estimation of paraffin/graphite composite phase change material using an inverse method

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ARTICLE INFO

Article history:
Received 21 November 2013
Accepted 8 March 2014

Keywords:
Phase change material (PCM)
Graphite
Periodic temperature method
Thermal conductivity
Thermal diffusivity
Inverse technique

ABSTRACT

In this paper, two types of graphite were combined with paraffin in an attempt to improve thermal conductivity of paraffin phase change material (PCM): Synthetic graphite (Timrex SFG75) and graphite waste obtained from damaged Tubular graphite Heat Exchangers. These paraffin/graphite phase change material (PCM) composites are prepared by the cold uniaxial compression technique and the thermophysical properties were estimated using a periodic temperature method and an inverse technique. Results showed that the thermal conductivity and thermal diffusivity are greatly influenced by the graphite addition.

1. Introduction

Thermal energy storage plays an important role in an effective use of thermal energy and it has applications in various areas, such as building heating/cooling systems, solar energy collectors and industrial waste heat recovery [1]. Thermal energy can be stored as a change in internal energy of a material as thermo-chemical reaction, sensible heat or latent heat [2].

In this work, we are interested in the latent heat storage method through phase change materials (PCM). Recent studies have been focused on the solid–liquid phase change because of its high storage capacity and nearly isothermal heat storage/retrieval process [3–6]. Phase change materials for latent heat storage can be classified into three major categories as organic material, inorganic material and eutectic PCMs [7,8]. Paraffin is taken as the most promising phase change material because it has a large latent heat, low cost, little or no super cooling, low vapor pressure, good thermal and chemical stability, nontoxic and noncorrosive [1,9,10]. However, paraffin suffers from a low thermal conductivity (0.21–0.24 $W\,m^{-1}\,K^{-1}$) [11]. These drawbacks reduce the rate of heat storage and extraction during the melting and solidification cycles and restrict their wide applications, respectively. Consequently, more working effort has been focus to improve the thermal conductivity of PCMs, by dispersing of high conducting particles within the PCM [12–15], impregnation of PCM into high thermal conductivity material with porous structures [16–18].

The use of graphite particles has advantages such as high thermal conductivity, low density in contrast to metals and high resistance to corrosion. The scope of this study is to make an experimental investigation on the effect of graphite on thermal conductivity, diffusivity and specific heat of paraffin/graphite composites. Two kinds of graphite were used to enhance thermal conductivity of the paraffin: Synthetic graphite (Timrex SFG75) and graphite waste obtained from damaged Tubular graphite Heat Exchangers. Paraffin/graphite phase change composites with the masse fraction of 5%, 10%, 15% and 20% were prepared by cold uni-axial compression method. A periodic measurement method was used to determine simultaneously the experimental thermophysical properties of paraffin/graphite composites at room temperature. Composite sample is fixed between two metallic plates, the front side of the first metallic plate is heated periodically using a sum of five sinusoidal signals and the temperature at the front and rear sides of both plates is measured and the experimental transfer function is calculated. The theoretical thermal heat transfer function is calculated by the quadrupole method. Then, thermal conductivity and diffusivity are simultaneously identified by comparison of experimental and theoretical heat transfer function.

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http://dx.doi.org/10.1016/j.enconman.2014.03.021
0196-8904/

Nomenclature

e	thickness (m)
D	conductivity ratio between the two phases
m	measured mass (kg)
t	time (s)
f	frequency (Hz)
I	thermal conductivity intensification
a	thermal diffusivity ($m^2\ s^{-1}$)
C_p	specific heat capacity ($J\ kg^{-1}\ K^{-1}$)
V	volume of composite (m^3)
b	thermal effusivity ($W\ s^{1/2}\ m^{-2}\ K^{-1}$)
H	heat transfer function
T	temperature (K)
$\tilde{T}$	Fourier transform of the temperature

Greek symbols

λ	thermal conductivity ($W\ m^{-1}\ K^{-1}$)
ϕ	flux, filler fraction
ρ	density ($kg\ m^{-3}$)
ω	pulsation

Subscripts

eff	effective (composite)
m	matrix, mass
f	filler
c	composite, contact
v	volume fraction
exp	experimental
th	theoretical

2. Experimental investigation

2.1. Investigated materials

2.1.1. PCM and graphite selection

The PCM tested in the present work is: paraffin with melting temperature of 56–58 °C and with specific density of 900 kg m^{-3}. The thermal conductivity enhancement is obtained by addition of conductive graphite particles. Two different kinds of graphite were used in this study:

- One type of graphite is the Timrex (SFG75) powder supplied by Timcal Graphite & Carbon at a bulk density of 2240 kg m^{-3}. It is a synthetic graphite with spherical shape and an average size of 75 μm, it is characterized by a well-aligned crystal structure and by a high thermal conductivity in plane [3].
- The second kind that has been tested is an industrial graphite "graphite waste". It was obtained from damaged Tubular graphite Heat Exchangers. It is a form of carbon with crystalline structure; it has good thermal and mass transfer characteristics that have led to its use for thermal conductivity enhancement. The measured bulk density is 1936 kg m^{-3} with an average size of 85 μm. Moreover, graphite has strong resistance to corrosion and chemical attacks which makes it compatible with most PCM. The recycling of graphite has a lot of benefits, it can preserve natural resources of graphite for future generations i.e. recycling graphite reduces the need for raw materials; it also uses less energy, and it have economic benefits.

2.1.2. Paraffin/graphite material elaboration

The elaboration method developed in the present study is based on the cold uni-axial compression, in this method paraffin powders and graphite particles are mixed together, then the obtained mixture (paraffin + graphite) is poured into a stainless steel mould followed by a uni-axial compression (80 bar) at ambient temperature (Fig. 1).

This technique leads to an anisotropic composite structure whose porosity is partially occupied by paraffin grains. Cylindrical paraffin/graphite samples were prepared under the same manufacturing conditions by the cold uni-axial compression method with mass fraction of 5%, 10%, 15%, and 20%. The thickness and the diameter of all this specimens were 5 mm and 60 mm respectively (Fig. 2).

Then, neatly cut of the cylindrical samples (Fig. 2b) to obtain a parallelepiped-shape specimens (Fig. 2c), with dimensions (42 mm·42 mm·5 mm) for thermophysical property measurements.

2.2. Thermophysical property measurements

2.2.1. Experimental set up

A periodical method was used to estimate simultaneously thermal conductivity, diffusivity and specific heat of paraffin/graphite composite materials at room temperature (Fig. 3). This method is based on the use of a small temperature modulation in a parallelepiped-shape sample ($42 \times 42 \times 5 mm^3$). The advantage of this method is that allows estimating simultaneously the effective thermal conductivity and diffusivity with their corresponding statistical confidence bounds [19]. It is well suited for polymers and composite materials with a thickness between 1 and 10 mm.

In the configuration used (which is presented in Fig. 3), the sample is sandwiched between two metallic plates. A thermal grease of high conductivity is applied on the contact surfaces between the sample and the metallic plates to ensure good thermal exchange between the various elements.

The front side of the first metallic plate (brass) is also fixed to heating device (thermoelectric cooler) and heated periodically using a sum of five sinusoidal signals. The whole device is placed in a vacuum chamber connected to a pumping system. The rear side of the second metallic plate (copper) is in contact with air at ambient temperature and high vacuum. The temperature is measured with thermocouples placed inside both front and rear metallic plates.

2.2.2. Theoretical model

The system under study is composed of several layers with different thermophysical properties. A heat transfer function is defined at each frequency as the ratio between the Fourier transforms of temperature at the x_r and x_f point of the x-axis (Fig. 4). We assume that the temperature of the front side of the metallic plate is modulated. The heat transfer exchanges on the rear face are taken into account trough a global exchange coefficient, which is considered constant during the experiment and the thermal properties of the sample are supposed constant.

By assuming a one-dimensional heat transfer in the x-direction, the conservation of energy equation in each layer can be written as:

$$\frac{\partial^2 T}{\partial x^2} = \frac{1}{a} \cdot \frac{\partial T}{\partial t} \quad \forall (t > 0) \tag{1}$$

In the case in periodic heating, the heat transfer equation is defined by [20]:

$$\frac{\partial^2 \tilde{T}}{\partial x^2} = \frac{j \cdot w}{a} * \tilde{T} = \alpha^2 \cdot \tilde{T} \tag{2}$$