Contents lists available at ScienceDirect



International Journal of Heat and Mass Transfer

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

## High performance form-stable expanded graphite/stearic acid composite phase change material for modular thermal energy storage



HEAT and M

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#### ARTICLE INFO

Article history: Received 18 January 2016 Received in revised form 13 April 2016 Accepted 20 June 2016

Keywords: Composite PCM Expanded graphite Stearic acid Form-stable Thermophysical property Thermal storage unit

#### ABSTRACT

A new synthesis method is developed for preparing form-stable expanded graphite (EG)/stearic acid (SA) composite phase change material (CPCM) by using "impregnation of liquid PCM into EG matrix and then compressing into stable-shape block". The effects of EG content and packed density on the thermal-physical property and thermal stability of form-stable EG/SA CPCM are firstly analyzed and its optimum parameters are investigated, and then an advanced form-stable modular thermal energy storage unit is manufactured and its thermal performance is evaluated. The SEM analysis shows that SA is well impregnated into graphite flakes and a regular laminar structure gradually forms with increasing packed density. The DSC analysis reveals that the CPCM nearly has no supercooling problem and the addition of EG and compressing operation have negligible effect on the phase change temperature and latent heat of SA. The form-stable CPCM has obvious anisotropic thermal conductivity and the largest difference between axial and radial thermal conductivity reaches up to 4 times. Both axial and radial thermal conductivities can be enhanced significantly by using the additive of EG, and the radial thermal conductivity is as high as 23.27 W/mK. The TGA analysis indicates that the form-stable CPCM has good thermal stability within a wide range of temperature and those samples with low porosity are susceptible to liquid leakage. Experimental results show the form-stable EG/SA CPCM exhibits excellent overall thermal performance by employing the optimum parameters of 25 wt.% EG content and packed density of 900 kg/m<sup>3</sup>. An advanced design method for modular energy storage unit is proposed by integrating copper tubes into the form-stable CPCM, and it can be easily used to achieve different energy storage capacities by assembling different numbers of modular thermal energy storage units. It appears that the proposed new method is very effective to synthesis high-performance form-stable graphite-based CPCM in comparison with the conventional methods, and the thermal conductivity can be enhanced about 130 times higher than that of pure PCM.

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#### 1. Introduction

Energy storage is a key technology to meet the global energy challenge, and it plays an important role in improving the efficiency of energy utilization by adjusting the discrepancy of energy supply and energy demand. The common methods for thermal energy storage mainly include sensible heat storage, latent heat storage, and thermochemical energy storage [1]. Among these approaches, latent thermal energy storage (LTES) is widely applied into various fields such as the solar energy utilization, industrial waste heat recovery, building energy saving, electronic heat dissipation, aerospace and so on, due to its advantages of high energy storage density, stable output temperature and remarkable energy

http://dx.doi.org/10.1016/j.ijheatmasstransfer.2016.06.066 0017-9310/© 2016 Elsevier Ltd. All rights reserved. saving. Currently, LTES has become a very popular research spot in the aspects of energy science and material science [2–5].

LTES technology realizes thermal energy storage/release by absorbing/releasing latent heat in the process of phase transformation, which is the most potential and important heat storage way [6]. However, almost all solid–liquid phase change materials suffer low thermal conductivity and liquid leakage problem when they undergo the solid–liquid phase change, which reduce the rate of heat storage and extraction during the melting and solidification cycle and restrict their wide applications [7]. To overcome these drawbacks, many efforts have been attempted, including utilization of metal fin or cellular construction to expand the heat exchange area [8], dispersing metallic or nonmetallic particles with high thermal conductivity into PCMs [9,10], and impregnating PCM into high thermal conductivity additives with porous structure, such as carbon materials and metal foams [11–13]. Among all

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| Nomenciature          |  |                        |  |
|-----------------------|--|------------------------|--|
| SA<br>EG<br>PCM       | stearic acid<br>expanded graphite<br>phase change material   | $T^m_{e} T^f_{e}$      | extrapolated end temperature during melting phase [°C]<br>extrapolated end temperature during freezing phase<br>[°C] |
| CPCM                  | composite phase change material                              | $T_{in}$               | inlet water temperature [°C]   |
| SEM                   | scanning electron microscopy                                 | Tout                   | outlet water temperature [°C]  |
| DSC                   | differential scanning calorimetry                            | $T_s$                  | surface temperature of CPCM [°C]   |
| TGA                   | thermal gravity analysis                                     | t                      | time [min]   |
| LTES                  | latent thermal energy storage                                |                        |  |
| CENG                  | compressed expanded natural graphite                         | Greek symbols          |  |
|                       |  | λα                     | axial thermal conductivity [W/(mK)]  |
| Dimensional variables |  | $\lambda_{\mathbf{r}}$ | radial thermal conductivity [W/(mK)]   |
| т                     | mass [kg]  | $\lambda_{PCM}$        | thermal conductivity of PCM [W/(mK)]   |
| $V_{\rm c}$           | volume of CPCM [m <sup>3</sup> ]                             | $\lambda_{CPCM}$       | thermal conductivity of CPCM [W/(mK)]  |
| $V_{\text{pore}}$     | volume of pore [m <sup>3</sup> ]                             | $ ho_{ m c}$           | density of CPCM [kg/m <sup>3</sup> ]   |
| Н                     | latent heat [kJ/kg]  | $ ho_{	extsf{g}}$      | density of graphite [kg/m <sup>3</sup> ]   |
| $T_o^m$               | extrapolated onset temperature during melting phase          | ω                      | mass fraction  |
| £                     | [°C]   | 3                      | porosity   |
| $T_o^J$               | extrapolated onset temperature during freezing phase<br>[°C] | $\phi$                 | volume fraction  |
|                       |  |                        |  |

additives and fins, carbon materials not only significantly decrease the weight and cost of the storage system, but also are well compatible with PCMs. Recently, new CPCMs made of PCM and inorganic matrix with high thermal conductivity have drawn wide attention. Carbon additives are widely employed as inorganic matrix to enhance the heat transfer of PCMs and prevent the leakage of liquid PCM due to its desirable properties including the porosity, high thermal conductivity, high stability, good compatibility with organic PCMs and lower density as compared with metal additives or fins [14–17].

According to the experimental research conducted by Energy Storage Research Group from Shanghai Jiao Tong University [18,19], EG is the most effective additive for enhancing the heat transfer of SA among three kinds of carbon additives (multiwalled carbon nanotube, graphene, EG), and the addition of 10 wt.% EG results in a more than 10-fold increase in the thermal conductivity when compared with that of the pure paraffin. Zhong et al. [16] reported the thermal conductivity of compressed expanded natural graphite (CENG)/paraffin composite can be improved by 28-180 times in comparison with that of the pure paraffin wax. Zhang et al. [17] prepared the EG/paraffin CPCM by absorbing liquid paraffin into EG, and found that paraffin is uniformly dispersed in pores of EG, the melting temperature of the CPCM is close to that of paraffin, and its latent heat is equivalent to the calculated value based on the mass fraction of paraffin. Fang et al. [20] prepared EG/SA CPCMs with different mass fractions of EG by using the same method, and found that the thermal diffusivity of the CPCM is more than 40 times higher than that of the SA when the mass percentage of EG is up to 50%. Py et al. [21] prepared EG/paraffin CPCMs by compressing loose EG powders into EG matrix block and then adsorbing the liquid paraffin into EG block, and they found that the addition of EG with mass fraction of 5–35% could improve the thermal conductivity of pure paraffin by 17-291 times.

The CPCMs based on EG matrix have the anisotropic characteristics of thermal conductivity due to the anisotropy of graphite. Mill et al. [22] reported that the anisotropic phenomenon of thermal conductivity of graphite-based CPCM becomes obvious with increasing bulk density, and the thermal conductivity in that direction perpendicular to the compressing direction is greater than that in the direction parallel to the compressing direction. The thermal conductivity of the composite matrix is 20–130 times greater than that of pure PCM.

From the works done by previous researchers, it can be concluded that methods involving the preparation of form-stable EG/PCM composite mainly include two kinds: one is directly to impregnate the liquid PCM into EG matrix in natural or vacuum condition, and the other one is to compress the EG into porous cubic matrix firstly and then make it soak the liquid PCM. The former concerns the impact of EG amount on the thermal conductivity of CPCM instead of the density, which can well ensure the PCM distribution inside of EG is uniform, but the craft will become complicated if the vacuum condition is created, and it will not be suitable for preparing CPCM with high EG content. The latter focuses on the influence of bulk density of porous EG matrix on the thermal conductivity of CPCM, but it is very difficult to assure the PCM is homogeneous inside CPCM due to the capillary resistance, especially for the graphite matrix with high bulk density. The impregnation kinetics will become too low and the impregnation requires more sophisticated process like vacuum condition [21]. In fact, the thermal performance of form-stable EG/PCM composite is relevant to the bulk density and EG content, however, it can be concluded that the abovementioned methods cannot be used to consider the bulk density and EG content as independent variables when preparing the form-stable EG/PCM composite.

In this paper, a new synthesis method is developed to prepare form-stable graphite-based CPCM by using "impregnation of liquid PCM into EG matrix and then compressing into stable-shape block". The proposed method has the distinct advantage that packed density and EG content can be as independent variables according to the different demands in the preparation of formstable CPCM. Fourteen kinds of CPCM samples (density: 700, 800, 900, 1000 kg/m<sup>3</sup>; EG content: 15 wt.%, 20 wt.%, 25 wt.%, 30 wt.%) are prepared by introducing the new approach, and the comprehensive effects of different EG contents and packed densities on the thermal conductivity, thermal-physical property and thermal stability of form-stable EG/SA CPCMs are investigated and analyzed. Moreover, an advanced modular thermal energy storage device using the form-stable CPCM is manufactured and its thermal performance is evaluated during the charging and discharging phases.

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