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Evaluation and comparison of erythritol-based composites with addition of expanded graphite and carbon nanotubes

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HIGHLIGHTS

- Preparing erythritol-based composites with EG and CNTs respectively.
- Testing and comparing composites based on thermophysics parameters.
- Evaluating composites to seek optimal additive ratio.

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ABSTRACT

To seek an appropriate additive for the preparation of erythritol-based composites, the evaluation and comparison of composites with the addition of EG (expanded graphite) and CNTs (carbon nanotubes) have been conducted in this paper. Composites with additive mass ratios of 1 wt%, 3 wt%, 5 wt% and 7 wt% were prepared by melting dispersion. The thermophysical performances of the composites were discussed in terms of melting point, latent heat and thermal conductivity, which were characterized using Fourier transformed infrared (FT-IR) spectroscopy, differential scanning calorimetry (DSC) and the transient hot wire (THW) method. The cost effectiveness of the composites was also considered from the point of view of two indexes, i.e., thermal conductivity per unit cost k_c and heat capacity per unit cost Q_c. The results revealed that the melting point of composites with EG continuously decreased with increasing mass ratio of additive due to the surface energy variation, while for the CNTs composites, it remained nearly constant. The latent heat of both composites gradually decreased as a function of mass ratio because of the replacement of erythritol by additives. The thermal conductivities of the composites also increased continuously with increasing addition of EG/CNTs. At the same mass ratio, EG appeared more effective than CNTs in enhancing the thermal conductivity, especially above 3 wt%. The optimal proportion of EG for the erythritol-based composite, with respect to not only the variation of thermal conductivity but also the heat capacity and cost effectiveness, was approximately 4 wt%.

1. Introduction

Mobilized thermal energy storage (M-TES) is a promising technique to supply heat to detached users [1–4]. Owing to the latent thermal energy storage (LTES) method, M-TES containers can be designed with great capacity but small size, making them transportable. In this way, heat can be delivered with an M-TES container by truck from heat source to users.

To further understand the characteristics of M-TES systems, lab-scale experiments were conducted [5–7]. Satisfactory performances, with the notable exception of a long charging time, were presented [1,2,4,6]. High thermal resistance, caused by low thermal conductivity of phase change materials (PCM), viz., erythritol, impaired the heat transfer performance of the containers; this is also regarded as a major disadvantage of other heat storage processes [8–11]. Thus, studies aimed at enhancing the thermal conductivity of erythritol have been conducted recently. Oya et al. [12,13] prepared erythritol-based composites with the addition of porous nickel, nickel particles and graphite and discussed the optimal proportions for such additives. Lee et al. [14] studied erythritol-expanded graphite (EG) composites and investigated

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Nomenclature

Abbreviation

CNTs	Carbon nanotubes
DSC	Differential scanning calorimetry
EG	Expanded graphite
FT-IR	Fourier transformed infrared
THW	Transient hot wire
LTES	Latent thermal energy storage
M-TES	Mobilized thermal energy storage
PCM	Phase change material
TES	Thermal energy storage

Symbols

c	cost of composite, €
$C_{P,\ Comp}$	special heat capacity of composite, kJ kg^{-1} K^{-1}
f_c	coverage factor
H	heat flow during the measurement of DSC, J
k	thermal conductivity of composite, W m^{-1} K^{-1}
k_c	thermal conductivity per unite cost of composite, W m^{-1} K^{-1} €$^{-1}$
m	mass of DSC sample, mg
m_{Comp}	mass of composite, kg
n	measurement times
Q	heat capacity of composite, kJ
Q_c	heat capacity per unit cost of composite, kJ €$^{-1}$
$s(\bar{x})$	mean value of standard uncertainty
t	time, s
T	temperature during the measurement of DSC, K
$T_{m,r}$	melting point for a particle size of radius
$u_c(y)$	combined standard uncertainty
u_f	Type B uncertainty
$u(x_i)$	standard uncertainty of single measurement
U	half-width of confidence interval
x_i	ith time measurement result
$\bar{x}$	arithmetic mean value of measurement results
y	measurand value
Δh_{Comp}	latent heat of composite, kJ kg^{-1}
ΔH	latent heat of bulk, kJ kg^{-1}
ΔT_{Comp}	temperature difference of composite, K
ρ_s	solid phase density, kg m^{-3}
σ_{sl}	solid-liquid interfacial energy, J m^{-2}

the effect of preparing EG with different interlayer distances. Karthik et al. [15] focused on erythritol-based composites with graphite foam and presented the prospect of using this material for thermal energy storage (TES) applications. Luo et al. [16] studied the mechanism of dispersion by preparing a stable erythritol suspension with nanotitania particles. Nomura et al. [17] prepared erythritol-carbon fiber composites using a novel hot press method.

In light of the above review, EG has been regarded as a promising additive candidate for the thermal conductivity enhancement of PCM. Carbon nanotubes (CNTs) and graphene are also potential candidates due to their excellent heat conduction performance. As the price of CNTs drops year by year, it will eventually be economically feasible to use CNTs as an additive. Fan et al. [18] tested the thermal conductivity of paraffin-CNTs composites with additive mass fractions of 0, 1%, 2%, 3%, 4% and 5%. The preparation of CNTs composites based on palmitic acid and polystyrene has also been investigated [19,20]. Conversely, in spite of its superior heat conduction performance, graphene is not suitable for the preparation of erythritol-based composites due to its extravagant price.

Composites based on the addition of EG and CNTs have been prepared and tested in many studies, but comprehensive evaluation and comparison of these additives in the context of erythritol-based composites have not yet been reported. Moreover, the optimal proportion of additive reported in the previous studies was determined mostly based on the thermal conductivity. However, the heat capacity and the cost of composites also vary due to the replacement of pure PCM with additives. The optimal proportion of additive, therefore, should instead be determined with regard to not only thermal conductivity enhancement but also heat capacity and economy.

In this paper, composites of erythritol-EG and erythritol-CNTs were prepared using a melting dispersion method. The properties of the composites were evaluated by discussing the variations of the melting point, latent heat and thermal conductivity. The optimal proportion of additive was determined based on the thermal conductivity, heat capacity and economy of the composites. This paper can thus provide insight into the preparation of high thermal conductivity material for M-TES applications.

2. Materials

2.1. Description

Erythritol ($C_4H_{10}O_4$, analytical reagent, purity: 99%) was bought from Shanghai Jinsui Biotechnology Ltd. EG was prepared by expanding graphite flakes (200 mesh) in a muffle furnace at 800 K for 20 s [21]. The images of the graphitic materials before and after this process are shown in Fig. 1. Multi-walled CNTs with hydroxyl groups have been reported as the optimal CNTs additive for thermal conductivity

(a) before expansion (b) after expansion

Fig. 1. Images of graphitic materials: (a) before expansion; (b) after expansion.