



Preparation and thermal properties of porous heterogeneous composite phase change materials based on molten salts/expanded graphite

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Abstract

Three kinds of porous heterogeneous composite phase change materials were synthesized from expanded graphite (EG) and binary molten salts ($\text{LiNO}_3\text{--KCl}$, $\text{LiNO}_3\text{--NaNO}_3$ and $\text{LiNO}_3\text{--NaCl}$) through solution impregnation method. Binary molten salt content in the composite phase change materials was calculated to be between 77.8% and 81.5% and high encapsulation efficiency was calculated to be between 72.8% and 78.8%. The thermal conductivity of binary molten salts was enhanced by 4.9–6.9 times after impregnation with EG. SEM photographs showed that the prepared composites were more homogeneous in comparison to other salt/EG composites prepared by infiltration or compression. Phase change properties of the porous heterogeneous composite phase change materials showed great thermal stability, which was maintained after 100 cycles.

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1. Introduction

Thermal energy storage (TES), as an important branch of energy science and technology, has been applied to solar energy systems, industrial waste heat recovery and thermal control in buildings (Zalba et al., 2003). However, collecting energy discontinuously as well as using energy already collected are difficult in practical engineering. For example, solar energy is available only during the day. Therefore the utilization of solar energy requires TES so that the excess heat can be collected during daytime and used overnight in order to secure continuity in energy output. Similarly,

industrial waste heat availability and utilization are not synchronized. So TES techniques are needed to recover the recyclable energy, and then release it when needed.

Latent heat storage, which uses phase change materials (PCM) as energy storage media through phase change, is highly attractive among several TES methods because it provides better energy storage density while temperature fluctuation is small (Farid et al., 2004). Solid–liquid PCMs are most widely used as ideal materials for collecting and releasing energy because of their high energy storage capacity. They can be classified according to their usage temperatures. Organic phase change materials, comprised of paraffin waxes, fatty acids and polyalcohols, together with many inorganic phase change materials like hydrated salts, can be used in a low temperature environment or at room temperature depending on their melting temperatures. On

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the other hand, molten salts and molten salt compositions, such as nitrates, chlorides, carbonates, and metal alloys, can be chosen as high temperature phase change materials in the range of 120–1000 °C (Kenisarin, 2010; Nomura et al., 2010).

We are interested in utilizing molten salts as PCM due to their potential use in high temperature TES systems. In general, several issues such as low thermal conductivity (around $1 \text{ W m}^{-1} \text{ K}^{-1}$) and liquid leakage of molten salts are limiting factors in the application of molten salts for heat storage.

In order to avoid the leakage of liquid PCMs, form-stable composite phase change materials have been offered by dispersing phase change materials into porous matrix which is stable in structure at phase change temperature and chemically inert. Many porous matrix are successfully applied in phase change materials, such as metal foams (Shiina and Inagaki, 2005; Wu and Zhao, 2011), porous carbon materials like expanded graphite (Bauer et al., 2006; Sari and Karaipekli, 2007; do Couto Aktay et al., 2008; Pincemin et al., 2008; Acem et al., 2010; Lopez et al., 2010; Zhao and Wu, 2011; Li et al., 2013; Wang et al., 2013; Xiao et al., 2013; Zhang et al., 2013), carbon nanotubes (Cui et al., 2010) and graphite foams (Lafdi et al., 2008), and porous ceramics including expanded perlite (Zhang et al., 2007), vermiculite (Karaipekli and Sari, 2009), montmorillonite (Fang et al., 2008), diatomite (Karaman et al., 2011) and halloysite (Mei et al., 2011).

Out of many potential PCM supports, expanded graphite (EG) is one of the most suitable porous matrixes used as support for PCMs because of its high thermal conductivity, low density and strong resistance to corrosion. Form-stable composite phase change materials choosing EG as the matrix to solve the problem of liquid leakage and low thermal conductivity of molten salts have been reported (Bauer et al., 2006; do Couto Aktay et al., 2008; Pincemin et al., 2008; Acem et al., 2010; Lopez et al., 2010; Xiao et al., 2013).

Recently, cascaded latent heat storages (CLHS) have been introduced as an effective TES alternative (Gong and Mujumdar, 1997; Michels and Pitz-Paal, 2007; Tabrizi et al., 2010; Shabgard et al., 2012; Tian and Zhao, 2013;), which ensures the optimal utilization of the storage material. In CLHS models, the PCMs with cascaded phase change temperatures were arranged according to their melting temperatures, from lowest to highest, and this design was calculated to be more effective than using only one kind of PCM (Gong and Mujumdar, 1997; Michels and Pitz-Paal, 2007; Shabgard et al., 2012; Tian and Zhao, 2013). However, only pure molten salts without support have been studied in the CLHS models. We expect that our composite phase change materials can offer good phase change storage and high thermal conductivity, which solves the low conductivity problem of pure PCM existing in the current CLHS models. In the present work, three kinds of composites made of expanded graphite and binary molten salts were prepared by solution impregnation method as a kind of porous heterogeneous

composite phase change materials. EG was used as the porous matrix to improve the conductivity and prevent the liquid leakage of molten salts. The prepared composites were characterized by scanning electron microscope (SEM) and X-ray diffraction (XRD), and their phase change properties were investigated by differential scanning calorimetry (DSC). Thermal conductivity of the composites was also measured by TPS method. Results showed that the prepared porous heterogeneous composite phase change materials possessed excellent phase change properties. Furthermore, thermal conductivity of the binary molten salts was increased by adding expanded graphite. To the best of our knowledge, three composite phase change materials, $\text{LiNO}_3(58.1)\text{--}41.9\text{KCl/EG}$, $\text{LiNO}_3(49)\text{--}51\text{NaNO}_3/\text{EG}$ and $\text{LiNO}_3(87)\text{--}13\text{NaCl/EG}$ have not yet been reported. The differences between the cascaded melting temperatures of these synthesized PCM materials are about 30 °C.

2. Experimental

2.1. Materials

Three kinds of binary eutectics of molten salts were chosen as phase change materials with cascaded melting temperatures of about 30 °C temperature (Table 1). Suitable salts that phase change occurs at under 300 °C were the targets for the use in TES system in this study. Lithium salts, such as lithium nitrate was chosen because of its high heat of fusion (373 J/g), which is significantly higher than sodium nitrate (177 J/g) and potassium nitrate (88 J/g) (Kenisarin, 2010). As a result, binary eutectics employing lithium nitrate were chosen. Lithium nitrate with purity $\geq 99.0\%$ was supplied by Tianjin Fu Chen Chemical Reagents Factory, China. Potassium chloride, sodium nitrate and sodium chloride with purity $\geq 99.0\%$ were obtained from Beijing Chemical Reagent Company, China. Expandable graphite with an expandable rate of 100–200 mL/g was purchased from Shanghai Crown Road Plastic Hardware Co., Ltd. Expanded graphite (EG) was produced by heating expandable graphite in a muffle furnace at 850–950 °C for one minute. SEM photographs are presented in Fig. 1. The resulting EG here presents worm-like structure (Inagaki and Suwa, 2001) with a diameter of about 200–500 μm , with a rich honeycomb network constructed from graphite flakes, which provides abundant pores for absorption. Chlorides salts were used in the study because of their high heat of fusion and thermal stability. Chlorides could cause the corrosion of components of the TES system, and construction of a barrier between salts

Table 1
Melting Temperature of binary eutectics chosen as PCMs.

Binary eutectics of molten salts (mass ratio)	Melting temperature (°C)
$\text{LiNO}_3(58.1)\text{--}41.9\text{KCl}$	166
$\text{LiNO}_3(49)\text{--}51\text{NaNO}_3$	194
$\text{LiNO}_3(87)\text{--}13\text{NaCl}$	208

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