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High thermal conductivity phase change composite with a metal-stabilized carbon-fiber network

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HIGHLIGHTS

- A phase change composite with carbon fiber and indium as fillers was developed.
- The composites were prepared by a hot pressing method.
- Thermal conductivity of the composites improved with increasing indium volume fraction.
- Cyclic durability of the composites improved with increasing indium volume fraction.
- The carbon-fiber network was welded and stabilized by the indium.

ARTICLE INFO

ABSTRACT

To enhance the thermal conductivity of phase change materials (PCM) such as sugar alcohol and molten salts, the preparation of a phase change composite (PCC) with a PCM and a filler with high thermal conductivity has been widely investigated. Although many reported PCCs have high thermal conductivity, the stability during thermal cycling endurance is often too low for practical use. This paper describes the development of a PCC with both high thermal conductivity and high cyclic durability. The PCCs were prepared by a hot-pressing method. Erythritol (melting point: 118 °C, thermal conductivity: $0.73 \, \mathrm{W \, m^{-1} \, K^{-1}}$) was used as a PCM, and carbon fiber (thermal conductivity: $900 \, \mathrm{W \, m^{-1} \, K^{-1}}$ in the fiber direction) and indium particles (thermal conductivity: $82.8 \, \mathrm{W \, m^{-1} \, K^{-1}}$) were used as the high thermal conductivity fillers. The effective thermal conductivity of the PCC was measured using the laser flash method and the network structures were analyzed using energy dispersive spectroscopy and scanning electron microscopy. Thermal cycling tests through the melting and solidification phases of the erythritol were performed to investigate the cyclic durability of the PCCs. We found that the indium particles melted during hot pressing, welding together the carbon fiber to produce a stable percolating network, which significantly enhanced the thermal conductivity and cyclic endurance of the PCCs.

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1. Introduction

To effectively exploit solar energy and achieve reduced fossil fuel consumption, advanced thermal energy systems are required. Such systems should have high-heat capacity, a high heat exchange rate, and long-term durability. Conventional sensible heat storage (SHS) technology has been widely applied in various fields and in fact, is the foundation of the current industry. However, SHS technology is not suitable for future thermal energy storage (TES), which require high heat capacity materials.

Latent heat storage (LHS) using phase change materials (PCM) is a promising technology which can satisfy the high-performance requirements of future TES applications. LHS is typically based on the storage or release of latent heat during a solid-to-liquid phase transition of a PCM. The use of latent heat allows LHS systems to achieve higher heat capacities than SHS, the ability to convert intermittent heat sources to constant ones, and excellent long-term durability. These advantages have allowed LHS systems to be applied in many fields [1], such as building materials [2], battery thermal management [3], solar heat utilization (including concentrated solar power) [4], and industrial exhaust heat recovery [5–7].

A low heat transfer rate, which is caused by low thermal conductivity of common PCM materials such as sugar alcohol

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and molten salts, is a major drawback for LHS systems [8]. Generally, heat conduction is dominant for typical indirect-contact LHS heat exchangers, such as shell-tube and packed-beds of encapsulated PCM. However, common PCM candidates such as paraffin [9,10], salt-hydrates [11], sugar alcohol [12,13], and molten salts [14,15] have thermal conductivities that are too low for practical application in TES systems. Generally, organic PCMs have conductivity values less than 0.4 W m^{-1} K^{-1} and those of inorganic PCMs are less than 1.0 W m^{-1} K^{-1} [8]. Although using high thermal conductivity metals as a PCM is attractive [16–20], there are few candidate materials in the temperature range below 300 °C. Therefore, identifying and investigating PCMs with high thermal conductivities is an important area of research.

Several studies have focused on developing phase change composites (PCC) with high thermal conductivities. Recently, Zhang et al. [8] reviewed state-of-the-art PCC technologies from over 200 studies presented in the literature. They broadly classified the fabrication methods for high thermal conductivity PCCs into three groups: simple mixing, impregnation, and compression. The most important step for enhancing the thermal conductivity of any PCC is forming a percolating network of a high thermal conductivity material [12].

With respect to the heat transfer network of a PCC, impregnating a porous material (such as a metal or carbon foam) with a liquid PCM has many advantages [8], as the foam has a high quality percolating network. It has been shown that the effective thermal conductivity of PCMs can be enhanced by 1–2 orders of magnitude using metal or carbon foams [17]. Unfortunately, foam substrates such as porous metals (usually Cu [21,22], Al [23,24], and Ni [21,25]) and carbon [26–29] are difficult to process, and are usually too expensive to use as PCM supports.

The compression method [12,30,31] is also promising for enhancing the thermal conductivity of PCCs by forming efficient heat transfer networks. In this method, solid PCM particles and fillers such as carbon fiber are mixed and then compressed with or without heating. Our previous study revealed that the fillers occupy the voids between the PCM particles during compression treatment, and this phenomenon accelerates the formation of a percolating network of filler [12]. In addition, fillers with anisotropy thermal conductivity, such as carbon fiber, become vertically oriented with respect to the direction of the press in the final PCC. This orientation enhances the drastic increase of the effective thermal conductivity of the PCC [12,30,32–34]. In addition, this method has economic advantages related to facile processing and the use of inexpensive carbon materials, such as expanded graphite [30,32–34].

However, the PCC prepared by the compression method would have a fatal problem that fillers in the PCC just contacts to each other. This condition of filler network in the PCC caused two challenges for practical use of the PCC. The first is that the stability under thermal cycling of PCCs prepared by compression is often too low for practical use. The second is that thermal conductivity of the PCC is still low with respect to the quite high thermal conductivity of filler materials; e.g. carbon fiber: 900 W m^{-1} K^{-1} [12]. This was because the condition that filler just contacts to each other would cause the increase of thermal contact resistance.

Therefore, this study attempts to solve the two challenges simultaneously and to develop PCCs with both high thermal conductivity and high cyclic durability, using a hot-press method. Erythritol was used as the PCM, and carbon fiber and indium particles were used as the high thermal conductivity fillers. Here, indium particles would be expected as a welding material for carbon fibers. The effective thermal conductivity of the PCC was measured using the laser flash method and the network structures were analyzed using energy dispersive X-ray spectroscopy (EDS) and scanning electron microscopy (SEM). In addition, cyclic

melting and solidification tests were performed to investigate the cyclic durability of the PCCs.

2. Materials and methods

2.1. Materials

Table 1 shows the thermophysical properties and specifications of the PCM and filler materials used in this study. Erythritol ($C_4H_8O_4$) was selected as a PCM; it has a melting point of 118 °C, a latent heat of 340 J g^{-1}, and a thermal conductivity of 0.73 W m^{-1} K^{-1} [35]. Particles of erythritol were used as a raw material for the preparation of PCCs. Fig. 1 shows the particle distribution of the PCM. The average particle size and packing ratio was 614 μm and 0.66, respectively. Note that the particle distribution is controlled as relatively broad to get high packing ratio of PCM.

We selected carbon fiber (CF, XN-100, Nippon Graphite Fiber Co., Ltd.) and micro-particles of indium (In, Kojundo Chemical Laboratory Co., Ltd.) as the high thermal conductivity filler The CFs had an average diameter of 10 μm, average length of 64 μm, and density of 2220 kg m^{-3}. The anisotropic thermal conductivity in the fiber direction was 900 W m^{-1} K^{-1} and 10 W m^{-1} K^{-1} in the direction normal to the fiber. The indium had an average particle size of 45 μm, a melting point of 156 °C, a latent heat of 28.7 J g^{-1}, and a thermal conductivity of 82.8 W m^{-1} K^{-1}. Note that both the erythritol and the carbon fiber were the same materials as used in our previous study [12].

2.2. Preparation of the PCCs

We prepared PCCs by hot-pressing (HP), as described in detail in our previous study [12]. First, the PCM particles, CF, and indium particles were wet-mixed in 99.9% acetone at 1500 rpm with a stirrer, for 30 min at room temperature. Then, the well-mixed wet samples were dried in air at room temperature. Next, about 3 g of the dry sample was packed into a carbon mold with a diameter of 12.7 mm and then placed in a compression machine with an electric heater. The samples were pressed at 10 kN with heating at a rate of 10 °C min^{-1} from room temperature to 160 °C; the pellets were kept at this temperature for 1 h. The hot-pressed samples with diameters of 12.7 mm and thicknesses around 10 mm were cooled in the furnace, and then removed from the mold. Finally, samples with a size of 10 × 10 × 1 mm were cut from the horizontal direction to the press-direction and carefully polished using #2000 emery paper.

2.3. Thermal cycling tests

Thermal cycling tests were undertaken on the as-prepared PCCs in a furnace with forced air cooling to evaluate their cyclic durability during the melting and solidification. One cycle consisted of heating the furnace to 125 °C, where it was kept for 15 min, and then cooling to room temperature. Tests were undertaken for up to 10 cycles.

2.4. Characterization

The density of the PCC samples was measured using a pycnometer (Quantachrome Instruments, Ultrapycnometer 1000). The accuracy of the instrument was ±0.02%. The measurement was repeated five times for each specimen and the average calculated; the standard deviation was within ±0.01%. The average value of the measurement was applied. The real volume fraction of CF; $V_{f,CF}$ and that of indium; $V_{f,In}$ in the final PCC was calculated based on