



Core-sheath structural carbon materials for integrated enhancement of thermal conductivity and capacity

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HIGHLIGHTS

- Core-sheath CNT@PC with 3D network structure was synthesized.
- The carbon sheath was conducive to the stability of network structure.
- The CNTs provide heat transfer paths for ss-PCMs.
- The hierarchical porous carbon sheath reduced interfacial thermal resistance.
- The ss-PCMs exhibited excellent thermal storage capability (up to 99.9%).

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ABSTRACT

Energy storage capacity and heat transfer ability are two important indexes for shape-stabilized phase change materials (ss-PCMs). In this paper, a core-sheath CNT@PC was prepared via carbonation of CNT@ZIF-8, simultaneously 3D structural supports were obtained due to the porous carbon (PC) sheath stabilized the CNT@PC network structure. Porous carbon (PC), derived from carbonized metal organic frameworks (MOFs), exhibited high porosity and large specific surface area. PCMs, absorbed by capillary force of porous structure, was stabilized in the pores of PC sheath. Further, the interaction between PCMs and CNTs reduced the interfacial thermal resistance greatly. Carbon nanotubes (CNTs), acting as heat transfer pathways, provided continuous channels for phonons transfer and realized rapid heat transformation between ss-PCMs and external environment. The obtained SA/CNT@PC ss-PCMs exhibited excellent thermal conductivity (1.023 W/mK), large phase change enthalpy (155.7 J g^{-1}) and high thermal storage capabilities (99.9%). The thermal conductivity of SA/CNT@PC was improved 222.6% and phase change enthalpy was increased 92.6% over SA/PC ss-PCM. SA/CNT@PC with large energy storage density, flexible designation, simple operation and near-constant temperature properties during phase change process shows great potential in waste heat utilization.

1. Introduction

Today, due to the ever increasing nature resources consumption and environmental pollutions, thermal energy storage materials including sensible and latent heat storage have been played a significant role in reducing the gap between supply and demand of energy [1–4]. However, sensible heat storage requires large volume and high temperature resulting in uncontrolled energy storage and release characteristics [5]. Conversely, thermal energy storage based on latent heat of phase change materials (PCMs) have attracted extensive attentions due to their capability in absorbing and releasing large amounts of latent heat

during phase transition. Thus PCMs were extensively applied in various thermal energy management systems including waste heat recovery, solar energy utilization, etc [6,7]. Especially, solid-liquid PCMs, such as paraffins and fatty acids, were widely exploited owing to their high enthalpy density, constant phase transition and excellent thermal stability [8,9]. However, leakage during the phase change process and low thermal conductivity of pure PCMs results in decreased energy storage efficiency and low charging/discharging rate, limiting their real application [10]. Recently, porous carbon [11–15], metal foams [16], polymer networks [17], nano-Si₃N₄ [18] and microcapsules [19] have been utilized to prevent the leakage of pure PCMs. Traditionally, high

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porosity of the supports ensures a large energy storage capacity. Capillary force can be optimized to stabilize PCMs by adjusting the structures of the supports. However, high porosity usually results in amorphous structures which may restrict the transport of quantized atomic lattice vibrations and block the heat conductive ability [20]. It is inadequate for meeting most energy charging/discharging targets. Thus, ideal supports with large energy storage capacity and high heat conductive ability are urgently to be developed in the real applications.

Reducing interfacial thermal resistance is a key issue to enhance thermal conductivity [21–23]. It is well known, high-thermal conductive fillers such as carbon nanotubes [24–26], carbon nanofibers (CNFs) [27–29] and graphene [30–32] can improve the thermal conductivity of pure PCMs. For instance, CNTs and CNFs can improve the thermal conductivity of the PCMs, but the enhancement of thermal conductivity is inadequate for real applications [23]. On the other hand, excessive fillers in PCMs lead to an obvious interfacial thermal resistance between the fillers and the PCMs because too much fillers would reduce the energy storage density [33]. Moreover, the aggregations of fillers due to the weak van der Waals bonding also block the transfer of phonons. Recently, surface modified fillers [34,35] facilitate the molecular interaction between heat transfer paths and PCMs, which can reduce the interfacial thermal resistance. Li et al. [34] prepared thermal conductive PCMs microcapsules with stearyl alcohol modified CNTs, the molecular interaction between CNTs and microcapsules reduced the interfacial thermal resistance obviously. Wang et al. [15] employed n-octadecylamine functionalized CNTs to improve the thermal conductivity of the PCMs composites, the thermal conductivity increased significantly with 1 wt.% of modified CNTs owing to the reduced interfacial thermal resistance. However, further increased the mass ratio of CNTs would result in the increased contact thermal resistance between the CNTs. So 3D network supports [36–40] were developed to improve the thermal conductivity. For instance, ultrathin-graphite foams and graphene aerogel stabilized PCMs can increase the thermal conductive of the composites obviously, which provide continuous heat transfer paths and increase mean free paths for phonons transmission. Further, the interfacial thermal resistance of ss-PCMs was reduced effectively. Interestingly, the phonons transmission was expanded from microscopic scale to macro scale PCMs in the 3D structural supports, which realized rapid energy conversion from ss-PCMs to the outside. However, the production of porous supports with complex procedures and rigorous reaction conditions limits their large-scale exploitation. It is still a challenge to explore special supports with uniform heat transfer pathways in the matrix and lower interfacial thermal resistance on the macroscale.

Herein, we present a facial way to prepare core-sheath CNT@PC support for PCMs by carbonizing CNT@ZIF-8 template. Simultaneously, 3D network structure was obtained because the converted PC sheath stabilized the network structure. High porosity and large specific surface area of PC facilitates the absorption of the PCMs. Further, PCMs were absorbed in the pores of PC by capillary force. The interaction between PCMs and CNTs reduced the interfacial thermal resistance greatly. On the other hand, CNTs, acting as 3D network pathways for heat transformation, increased the phonons transmission pathways and accelerated heat transmission between the ss-PCMs and the environment. Traditionally, highly porous support ensures a large energy storage capacity, but leads to a low thermal conductivity. The superiority of CNT@PC with 3D core-sheath structure is the integrated enhancement of energy storage capacity and heat conductive ability. SA/CNT@PC with large energy storage density, flexible designation, simple operation and near-constant temperature properties during phase change process shows great potential in waste heat utilization.

2. Experimental

2.1. Synthesis of CNT@PC

The CNT@ZIF-8 was synthesized according to the previous report [41], and the detailed preparation process was shown in the supporting information. The CNT@PC support was obtained via the carbonation of CNT@ZIF-8 under nitrogen at 1000 °C for 6 h. The thickness of the porous carbon was controlled via tuning the amount of ZIF-8 precursors on the CNT.

2.2. Synthesis of SA/CNT@PC ss-PCMs

The SA/CNT@PC composites were synthesized by a wet impregnation method [42]. Typically, 0.2 g of stearic acid (SA) was dissolved in the absolute ethanol, and 0.067 g of CNT@PC was added to the solution. The mixture was stirred vigorously at 90 °C for 1 h. After that, the mixture was dried overnight at 80 °C to remove the ethanol solvent. Finally, the ss-PCMs were collected and named SA/CNT@PC. Shape stabilized PCMs with different loading amounts of SA (60, 70, 75 and 80 wt.%) were obtained to investigate the thermophysical properties of the ss-PCMs.

2.3. Characterizations

The morphology of prepared samples were characterized on a scanning electron microscopy (SEM, ZEISS SUPRA55). The crystallization behaviors of the ss-PCMs were tested by Powder X-ray diffraction (XRD, M21X). Thermal stability of samples was conducted with an apparatus (TGA, NETZSCH, STA449F) at a heating rate of 5 °C min⁻¹ under N₂ atmosphere. Fourier-transform infrared (FTIR) spectra were acquired on a Nicolet 6700. N₂ adsorption-desorption isotherms and pore size distributions of supporting materials were derived from Micromeritics ASAP 2420 adsorption analyzer and Barrett-Joyner-Halenda (BJH) model. Thermal properties of prepared PCMs were measured by differential scanning calorimetry (DSC, NETZSCH, STA449F3) at a heating rate of 5 °C min⁻¹ and a cooling rate of 5 °C min⁻¹. Thermal conductivity of PCMs was tested with transient plane source technique, using a hot disk thermal constants analyzer (Hot Disk TPS 2500S, Hot Disk AB Company).

3. Results and discussion

3.1. Characterization of CNT@PC support

The morphology and microstructure of raw CNT, CNT@ZIF-8 and CNT@PC were shown in Fig. 1. The CNTs with a smooth surface were about 30–40 nm in diameter and several micrometers in length (Fig. 1a). The ultra-long CNTs were applied as the template for the in-situ synthesis of ZIF-8. The surface of the composite became rough and the average diameter were increased to about 100 nm (Fig. 1b) after the growth of ZIF-8 on the CNTs, which means the ZIF-8 sheaths were successfully coated on the CNTs surface. The TEM images showed that the ZIF-8 contacted with the CNTs directly. Further, the ultra-long CNTs constructed 3D network support and ZIF-8 sheath stabilized the 3D network structure. Finally, CNT@PC was obtained via carbonizing CNT@ZIF-8 under the N₂ atmosphere at 1000 °C for 6 h. The average diameter of CNT@PC was decreased to about 60–70 nm (Fig. 1c) because the Zn ions were reduced to Zn metals by carbon and vaporized under 1000 °C. The synthesis process of SA/CNT@PC ss-PCMs was illustrated in Scheme 1.

The formation of ZIF-8 on CNTs was the key step in preparing CNT@PC support. XRD and FT-IR results (Fig. S1, Supporting Information) confirmed the ZIF-8 crystals were grown on the CNTs (Fig. 1b). XRD diffraction peaks of the CNT@ZIF-8 were in agreement with that of pure ZIF-8 [43], which suggested the formation of ZIF-8

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