

A functional form-stable phase change composite with high efficiency electro-to-thermal energy conversion



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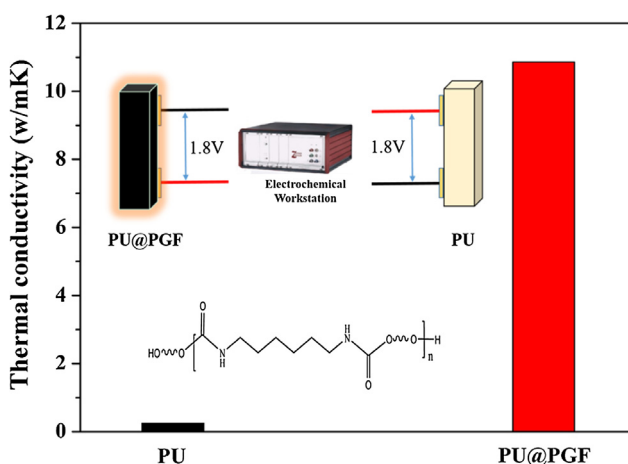
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HIGHLIGHTS

- The composite exhibits an in-situ solid-solid phase change behavior.
- The enthalpy of polyurethane is enhanced within the matrix.
- The thermal conductivity of the composite is 43 times as much as that of the polyurethane.
- Supercooling of polyurethane is greatly reduced.
- The composite is applied to cold protection as a wear layer.

GRAPHICAL ABSTRACT

The thermal conductivity of PU was enhanced to 43 times of the pristine value by encapsulation in a PGF, PU@PGF can be used for highly efficient electro-to-heat energy conversion and storage with the highest energy storage efficiency up to 85%.



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ABSTRACT

A novel solid-to-solid phase change composite brick was prepared by combination of polyurethane (PU) and pitch-based graphite foam (PGF). The carbonaceous support, which can be used for mass production, not only greatly improves the thermal conductivity but promote electro-to-heat conversion efficiency of organic phase change materials (PCMs). Our composite retained the enthalpy of PCM and exhibited a greatly reduced supercooling temperature. The novel composite was investigated by differential scanning calorimetry (DSC), thermogravimetric analysis (TGA) and scanning electron microscope (SEM). The enthalpy of polyurethane has increased about 8.6% after infiltrating into graphite foam. The composite was very stable during thermal cycle test, and the electro-to-heat conversion efficiency achieves to 85% at lower voltages (1.5–1.8 V), which can vastly reduce energy consumption. The as-prepared composite was used in a wear layer to test its performance comparing with normal fabric.

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1. Introduction

With the accelerated process of human industrialization, global energy consumption is increasing dramatically. According to one survey on the global energy consumption structure conducted in the year 2012, oil accounted for 33.1% of the global primary energy consumption, while coal and natural gas contributed 29.9 and 23.9%, respectively [1]. As hydrocarbon resources, these three resource types made up 86.9% of the total global primary energy consumption. Both the global demand for energy and the fossil-fuels-based pattern of energy consumption continue to worsen the shortage of energy resources, environmental pollution, climate change, etc. [2]. In order to solve the above problems, solar, wind, and nuclear energy are gradually being explored by humans as new energy forms to replace traditional energy sources like fossil fuels. In Europe, for example, renewable energy is expected to account for about 70% of the total power generation by 2040 (about 32% in 2015). For America, the share of renewable energy is expected to increase from 14% in 2015 to 44% by 2040, while the percentage of natural gas and coal-fired power is expected to reduce from 33 to 31%, indicating that the proportion of renewable energy is expected to be greater than that provided by natural gas and coal [3]. Meanwhile, energy storage technologies have received widespread industrial and governmental attention due to their ability to tune the mismatch between supply and demand [4–8].

Among the various energy storage methods, phase change materials (PCMs) can be viewed as a valid strategy for thermal energy storage due to their high energy storage capacity over a small temperature range [9–11]. For PCMs, a large amount of thermal energy can be absorbed or released directly during phase transition in a physical process without any chemical reaction involved, namely through latent heat storage [12–15]. PCMs encapsulated by porous materials have been the focus of research in industry and academics. Fang combined SiO_2 gel with lauric acid and achieved a maximum loading capacity of 64.8% [16]. By changing the scaffold to expanded dolomite, the loading percentage could be increased up to 90% [17]. Nevertheless, due to the limitation of the supporting materials, electro-to-thermal conversion cannot be applied for these composites. This challenge may be tackled by infiltrating PCMs into carbon materials, such as carbon nanotube (CNT) sponges or CNT arrays, which were shown to yield electro-to-thermal conversion efficiencies of 50 and 74%, respectively [18,19]. However, the efficiency of electro-to-thermal conversion should be further increased for more practical applications.

Furthermore, PCM-based thermal energy storage and conversion techniques have additional advantages, such as a smaller volume change during the phase change process, non-toxicity, no corrosion of the container, and long-term cycle life [20]. Therefore, PCMs may be strong candidate materials for solar energy storage, building energy efficiency, waste heat recovery, etc. [21–24]. PCMs can be classified into three main categories based on their phase states, including solid-to-liquid, solid-to-solid and liquid-to-gas PCMs, among which solid-to-liquid PCMs are the most popular since they can provide high latent heat and multiple types. However, solid-to-liquid PCMs suffer from limitations including leakage in their liquid state and their obvious volume change. In comparison, solid-to-solid PCMs can stay intact while storing energy and in some cases be easily obtained by simple functionalization of solid-to-liquid PCMs. Currently, academic efforts are focusing on developing a variety of solid-to-solid PCMs [25,26].

Nonetheless, despite the superior energy storage ability of latent heat systems, there are still many hurdles hindering the wide application of PCMs, among which the low thermal conductivity of pure organic PCMs preventing their further application.

The limited thermal transport speed will lower the system's efficiency. Over the past decade, researchers have introduced different supports or additional agents into the scaffolds of PCMs to boost the thermal conductivity, such as expanded graphite [27–31], CNTs [32,33], graphite [34], graphene sheets [35,36], sponges [18] and reduced graphene oxide (rGO) aerogel [37], which could remarkably enhance thermal conductivity. However, these additives usually either prevent large-scale industrial production, or exhibit poor coupling with PCMs on the molecular level, stopping their industrial application.

Graphite foam (GF) consists of a highly ordered porous carbon structure with some intriguing features, such as a high thermal and electrical conductivity, low density ($0.2\text{--}0.8\text{ g/cm}^3$), high temperature tolerance, and high corrosion resistance [38], which make it ideal for many functional applications based on their very high porosity. Moreover, a GF and polydimethylsiloxane (PDMS) composite was shown to have a very high electrical conductivity of *ca.* 10 S cm^{-1} , much higher than that of the graphene base composite and that of CNTs, at an ultralow graphene loading of $\sim 0.5\text{ wt.}\%$ ($\sim 0.22\text{ vol.}\%$) [39]. Due to its high thermal and electric conductivity and low bulk density, it was regarded as a new ideal loading material for PCMs. For encapsulation, PCMs could be injected into the matrix network and absorbed by the loading material. However, considering the pore size of GF, it is difficult to keep solid-to-liquid PCMs from leaking [4,26]. Since solid-to-solid PCMs can remain in a solid state when the temperature rises above the phase change temperature, it can avoid the leakage problem. With a special skeleton and crosslinking structure, the free movement of polymer molecular chains can be restrained at high temperatures [23]. The combination of GF and solid-to-solid PCMs may therefore produce an excellent phase change composite.

In our previous study, we introduced a solid-to-solid PCM into a graphitic matrix to solve the leakage of PCMs [23]. Herein, we improve this method by in-situ synthesis of polyurethane (PU) prepared by polyethylene glycol 6000 (PEG-6000) and hexamethylene diisocyanate (HDI). The solid-to-solid PU can suppress supercooling intervals. Macroporous pitch-based GF (PGF) with a high thermal conductivity was produced by high-temperature graphitization of meso-phase pitch. The resulting product showed enhanced thermal conductivity, supercooling suppression, and stable latent heat.

2. Material and methods

2.1. Materials

Polyethylene glycol 6000 (PEG-6000, analytical grade) was purchased from J&K Scientific Ltd and used without further purification.

2.2. Preparation, characterization of PGF and PU@PGF composite

The synthesis process of PGF started with a commercial sponge, which was cut into the desired shape and washed to remove dirt. The clean foam was used as a template, immersed into molten pitch, and sealed in an autoclave. The autoclave was filled with nitrogen gas of 5 MPa as protective atmosphere. It was heated up to $500\text{ }^\circ\text{C}$ and kept for 5 h to obtain a raw foam, which was taken out of the autoclave and subsequently carbonized at $1000\text{ }^\circ\text{C}$ in a furnace under N_2 protection. The carbon foam was further graphitized at a temperature as high as $3000\text{ }^\circ\text{C}$, resulting in the final product. PEG-6000 was degassed at $100\text{ }^\circ\text{C}$ under vacuum for 24 h. A mixture of degassed PEG-6000 and HDI at a stoichiometric ratio of 1:2 was stirred rapidly at $70\text{ }^\circ\text{C}$ for 3 min, followed by injection into PGF in a vacuum furnace at $70\text{ }^\circ\text{C}$ for 2 h. The composites

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