



Electro/photo to heat conversion system based on polyurethane embedded graphite foam [☆]



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HIGHLIGHTS

- Solid–solid phase change of polyurethane is realized within porous graphite foam.
- PU@GF exhibits high thermo/electro conductivity and light absorptivity.
- The high efficiency electro/photo-to-heat conversion system is reported.

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ABSTRACT

Organic phase change materials (PCMs) have exhibited many promising potentials for thermal energy conversion and storage, but they are still confronted with many technical bottlenecks for practical application, such as low conductivity, leakage during phase transition process, and lack of functionality. In this article, a highly-efficient electro/photo to heat conversion system of polyurethane@graphite foam (PU@GF) phase change composites was successfully fabricated through in situ polymerization of polyethylene glycol (PEG) in GF. The obtained PU presents solid–solid phase transition behavior that is different from the solid–liquid phase change of original PEG, which can prevent PU from leakage even if it was loaded in micrometer pores in the GF during application. On the other side, PU can improve the thermal stability and decrease the overcooling degree of the composites in effect. Excellent conductive network was provided by the GF, with which the light absorption and thermal conductivity of the composites were enhanced dramatically. Consequently, the solid–solid phase change composites can effectively store electricity or sunlight energy. The electro-heat storage efficiency of the composites can exceed 80% at 1.2 or 1.4 V, meanwhile, the photo-heat storage efficiency can close to 67% under simulated solar illumination. This result presents the highest efficiency for electro-to-heat conversion by PCMs techniques driven by a quite low voltage up to now, and will give rise to a new expectation of functional PCM application for energy conversion and storage.

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1. Introduction

Phase change materials (PCMs) can store and release a huge amount of thermal energy as the latent heat from environment, solar irradiation and electronic products, etc. This heat storage usually happens during their phase change course (e.g. melting and solidification) at a constant temperature [1–8]. As a kind of important PCMs, organic PCMs have an extensive potential for practical applications in solar energy storage, green house construction

and waste heat recovery and so on [9–16]. This is because of their promising phase change enthalpies, wide melting points and chemical stability. However, there are still some problems need to solve properly to use organic PCMs effectively in energy storage in practice. For example, (i) the low thermal conductivities could result in low energy conversion and storage efficiency; (ii) the fluidity after phase change may result in leakage during cyclic use; (iii) electrical insulator behavior prevents taking advantage of electricity as energy source, or non-absorb light property makes difficult to employ sun power. Therefore, PCMs have to enhance their thermal and electrical conductivity, light absorption and fabricate appropriate encapsulation so that they could play a significant role in future energy conversion and storage.

Thermal conductivities of PCMs have been effectively increased by using various materials with high thermal conductivities, such

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as calcium carbonate [17], expanded graphite [18], carbon nanotubes [19,20], nanographite [21], graphene [22] and carbon nanofillers [23]. On the other side, the efficiency of light-to-heat conversion of PCMs can be increased by introducing different organic dyes [24–26]. However, these samples are usually electrically insulating or still present too low electrical conductivities. Recently, more attention has been paid to porous and conductive skeleton to encapsulate PCMs [27–30], which can make an electrically conductive composite with enhanced thermal conductivity. For example, (i) a significant enhancement of thermal conductivity was achieved by applying porous metal foams, which were nearly 15 times larger than that of pure paraffin [27]; (ii) a high thermal conductivity of $2.635 \text{ W m}^{-1} \text{ K}^{-1}$ was obtained from graphene aerogel/octadecanoic acid composite, which was 14 times higher than that of octadecanoic acid [28]. In previous work of our group, carbon nanotube sponge and carbon nanotube array were successfully utilized as porous matrix to encapsulate paraffin wax or eicosane. The carbon nanotube sponge/paraffin wax composite realized both electro and photo to heat conversion and storage for the first time with a high thermal conductivity of $1.2 \text{ W m}^{-1} \text{ K}^{-1}$ [29]. The carbon nanotube array/eicosane composite with excellent electrical conductivity can store heat at modulated voltages (1–2 V) and obtain 74.7% electro-heat storage efficiency [30]. Therefore, porous and conductive skeletons are effective to improve thermal conductivity and even realize electro/photo to heat conversion and storage. But these porous materials are still difficult for widespread and practical application, because of the non-absorb light property of metal foams and the high price of graphene or carbon nanotubes.

Graphite foam (GF), composed of ordered graphitic ligaments, has been widely applied in heat-exchangers, evaporative cooling systems, electronic heat sinks, etc. [31–35]. It has raised great interests because of the unique properties, like high thermal and electrical conductivity, low bulk density, high chemical inertness, good light absorption and low cost. Hence, it has been considered as an ideal candidate to load PCMs for advanced heat storage. However, the leakage of PCMs in GF remains a substantial problem. The interconnected pores in the GF skeleton are too large to prevent the leakage of PCMs during the solid to liquid phase transition.

In this paper, in order to solve the leak problem and functionalize PCMs, we have prepared a smart storage system based on stable PCM@GF composite, which can not only increase the thermal/electrical conductivity, but also store heat energy driven by small voltages (e.g. 1.0 V) or light illumination with high electro-to-heat or photo-to-heat storage efficiencies. The smart storage system was synthesized by the reaction of diisocyanates and polyethylene glycol (PEG) directly inside the GF. The cross-linking copolymerization of the two organic compounds results in the formation of polyurethane (PU) with durable solid to solid phase transition. The obtained composite was denoted as PU-PEG, which made it free from leak problem after thermal energy storage. The thermal conductivity of the prepared PCM composite was enhanced to a high value of $3.5 \text{ W m}^{-1} \text{ K}^{-1}$, which was nearly 12 times higher than that of pure PEG. Besides, GF offered a good consecutively conductive network that can provide homogeneous heat conduction to the well distributed PU to achieve efficient electro/photo to heat conversion and storage.

2. Experimental

2.1. Materials

PEG with different molecular weight (PEG4000, PEG6000 and PEG8000) and 3-isocyanatomethyl-3,5, 5-trimethylcyclohexyl isocyanate (IPDI) were supplied by Aladdin Industrial Inc., China.

Graphite foam was purchased from Gaotai Wuchan Inc., Qingdao, China.

2.2. Preparation of PU@GF composite

PEG4000, PEG6000 and PEG8000 were degassed under high vacuum at $100 \text{ }^\circ\text{C}$ for 4 h, respectively. The degassed PEGs and IPDI in the 1:2 mol ratio were mixed under stirring at $80 \text{ }^\circ\text{C}$ for 10 min, then the mixtures were injected to GF in vacuum at $80 \text{ }^\circ\text{C}$ for 1 h. After the pores of GF were filled with polymers, the composites were kept at $100 \text{ }^\circ\text{C}$ in vacuum for additional 6 h.

2.3. Composite characterization

The thermogravimetric analysis (TGA) was carried out at a TASDT-Q600 instrument with a heating rate of $10 \text{ }^\circ\text{C}/\text{min}$ in N_2 flow. The differential scanning calorimetry (DSC) data were collected with a differential scanning calorimeter (Setaram DSC 131 evo) with a heating or cooling rate of $5 \text{ }^\circ\text{C}/\text{min}$ under Ar. All the sample weights were between 1–5 mg. The scanning electron microscope (SEM) characterization was performed on a field-emission microscope (Hitachi-s4800) operated at 10 kV. Thermal conductivity measurements were carried out with hot-wire thermal conductivity instrument (Xiotech TC3010). The thermal conductivity of each sample was calculated by the average value from five measurements.

2.4. Electro to heat conversion

Plate-like samples were connected to an electrochemical workstation (Zahner ennium/IM6) by two copper wire electrodes on each side. A certain bias (1.0–1.4 V) was applied to the sample by the workstation for about 600s, while the current flowing through the sample was recorded simultaneously. Thermal resistance (Pt100) was inserted into the target material to measure the inside temperatures during heating and cooling. Accuracy of the Pt100 thermal resistance is $\pm 0.15 \text{ }^\circ\text{C}$. Temperature evolution of the sample was recorded by a data acquisition system connected to the thermal resistance.

2.5. Photo to heat conversion

PU@GF composites were illuminated by simulated sunlight using a solar simulator (Newport Thermo Oriel 91195A-1000) at a controlled intensity ($90 \text{ mW}/\text{cm}^2$). The sample temperature was measured by a Pt100 thermal resistance and recorded by a data acquisition system using the same configuration. The light intensity was calibrated by a standard Si solar cell (91,150 V). Each sample was exposed to light for 15 min and then light was turned off to let the sample cool down. During the tests, samples were placed in a plastic box to minimize the environmental influence of air convection.

3. Results and discussion

3.1. Morphology and microstructure

Unloaded GF has large interconnected pores as shown in Fig. 1a and b. However, larger pores can easily promote liquid PCMs not only penetrating them, but leaking out of them. When impregnation and polymerization of the mixture of PEG8000 and IPDI completed in the skeleton of the GF, PU was in situ formed as a solid–solid PCM. This makes a good contact between the PU and the GF, moreover, the pores of the GF were almost fully filled with PU as illustrated in Fig. 1c. According to the result of TGA, 20 wt%

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