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Shape-stabilized Phase Change Materials Based on Carbon Matrix for Thermal Performance Enhancement: a Review

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With the growingly serious energy crisis and the dramatically increasing greenhouse gas emission worldwide, thermal energy storage has been attracting extensive attention for its application in re-regeneration energy, which provides a perfect solution to coordinate the mismatch between energy supply and demand. Organic phase change materials are the most widely used materials for thermal energy storage, but the inherent disadvantages drastically impede their further application, such as low thermal conductivity and leakage. Previous research shows that carbon-based scaffold can effectively prevent the leakage during phase transition attributing to the capillary force and the surface tension between the phase change material and the micro-channel surface. The porous structure provides abundant heat transfer areas, and the scaffold itself is perfect heat transfer media. It makes the heat transfer greatly enhanced by combining with carbon-based matrix. This composite shows huge potential for promoting the efficiency of thermal management and energy conversion. This paper gives a state-to-art review about the development of shape-stabilized composite phase change material with carbon-based matrix as the supporting material for organic phase change material including the fabrication and thermal property of the composite phase change material, and the newest application of the composite.

1. Introduction

With the global depletion of conventional fossil fuel and the dramatically rising emission of greenhouse gas, the demand for exploring a perfect alternative is growing more and more urgent. Renewable energy offers a good solution to this issue and has been attracting extensive attention during the past decades, including solar (Munkhchuluun et al., 2020), biomass (Mohebbi et al., 2020), wind (Li et al., 2020), tidal energy (Gaurier et al., 2020), geothermal power (Gil et al., 2020), water (Comino et al., 2020), nuclear energy (Batchelder, 2020), and hybrid power systems (Li et al., 2020). However, renewable energy cannot be collected at any time and any region needed, which is highly dependent on the climate condition (Li et al., 2019). Developing thermal energy storage (TES) materials is essential to coordinate the mismatch between energy supply and demand in time and space.

TES are classified into three categories: sensible heat storage, latent heat storage (LHS) and chemical heat storage (Pinto et al., 2020). Among them, LHS is most widely used due to its relatively high heat capacity and narrow temperature variation range during phase transition (Wu et al., 2019). For LHS, the low thermal conductivity and the leakage during the phase change process are the two main drawbacks that have blocked the application of phase change materials (PCMs).

In recent years, composite PCM (CPCM) combined with carbon-based three-dimensional (3D) framework as supporting material has exhibited desirable thermal performances and less or no leakage without a large decrease in heat capacity (Tong et al., 2019). Several graphene-based supporting materials have been extensively reviewed in the previous literature (Wu et al., 2019). However, previous review work rarely focused on the preparation and application of carbon-based matrix utilized as supporting material for PCM. To give a state-to-art review for 3D carbon-based supporting materials applied for TES is very meaningful.

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This paper reviews the latest development of CPCMs based on 3D porous carbon materials for organic PCMs, including the three main kinds of scaffolds: expanded graphite, graphene aerogel, and carbon foam. The applications of this kind of CPCMs are discussed. There are 4 sections in this paper. In Section 1 the objective of this paper is illustrated. In Section 2 the main carbon-based scaffolds are described, including expanded graphite, graphene aerogel and carbon foam. In Section 3 the applications of carbon-based CPCMs are listed, including thermal management and thermal conversion. In Section 4 a conclusion is given, and the future works on this topic are recommended.

2. Carbon-based scaffold

The liquid leakage and shape change during phase transition have long bothered researchers and engineers since the beginning of PCMs (Deng et al., 2019). Various methods have been proposed and developed to optimize the properties of PCMs, including encapsulation (Zhao et al., 2020), nanoparticles (Jebasingh et al., 2020), porous structure (Huang et al., 2019), solid-solid PCM (Shi et al., 2019). Amongst, impregnating PCM into carbon-based scaffold is a desirable way to enhance thermal conductivity attributed to the abundant heat transfer channels offered by the nano- or micro- pores. The leakage of PCM during melting procedure is also improved as the PCM is restricted in the pores of scaffold. As to shape stability, the CPCM keeps solid state during the melting/freezing cycles (Gao et al., 2018). That is to say, the CPCM can hold its solid shape even when the temperature is above the melting point of PCM (Umair et al., 2019).

The list of CPCMs are shown in Table 1. According to the previous study, PCMs are impregnated into supporting materials mainly by two ways: melting impregnation method, vacuum impregnation method. On the one hand, the thermal conductivity is enhanced by adding supporting materials. But on the other hand, the latent heat is decreased as partial PCM is replaced by supporting materials. Previous research shows that the minimum mass ratio of supporting material to adsorb all the PCM by melting impregnation method is always higher than that by vacuum impregnation method, so the loss of latent heat is lower by vacuum method. In a word, vacuum method is more desirable.

Table 1: List of CPCMs

Supporting material	PCM	Melting point (℃)	Thermal conductivity (W/m·K)	Reference
Carbon foam	Polyethylene glycol	36.39	Null	Maleki et al., 2020
Carbon foam	Paraffin	56.83	Null	Maleki et al., 2020
Carbon foam	Palmitic acid	64.08	Null	Maleki et al., 2020
Carbon foam	Paraffin	57.69	0.6	Ola et al., 2019
Carbon foam	Paraffin	59.7	5.23	Wilson et al., 2019
Carbon foam	Erythritol	~ 118	< 0.12	Kim et al., 2019
Textile-structured carbon scaffolds	Paraffin	Null	0.63	Sheng et al., 2020
Expanded graphite	paraffin	45 ~ 50	4.331	Zhang et al., 2020
Expanded graphite	Dodecane	- 9.67	2.2745	Song et al., 2019
Expanded graphite	Paraffin	59.74	12.46	Wang et al., 2020
Graphene aerogel	Paraffin	Null	0.22 ~ 14	Feng et al., 2020
Graphene aerogel	Paraffin	49.95	1.432	Tang et al., 2019
Phosphorylated polyvinyl alcohol and graphene aerogels	Polyethylene glycol	60.9	0.610	Shen et al., 2019
Reduced graphene oxide aerogel beads	1-tetradecanol paraffin	45.64	~ 0.4	Zhao et al., 2020
Nanofibrillated cellulose and carbon nanotubes	N-octacosane	Null	0.583	Du et al., 2020
Polystyrene-carbon nanotubes	Paraffin	56.88	0.39	Maleki et al., 2019
Graphene oxide Pickering	Paraffin	43.65	0.645	Zhao et al., 2019
Melamine foam	Paraffin	56.8	0.096	Wu et al., 2020

2.1 Expanded graphite

Expanded graphite (EG) is a type of commonly used porous matrix for PCM with worm-like structure. The abundant pores with widely distributed sizes make it possible to adsorb PCMs into the micro-confined structure by capillary force and surface tension. Generally, expanded graphite is made from flake graphite through chemical or thermal treatment, or by microwave irradiation power, and exhibit excellent properties to enhance heat transfer of CPCM due to its large surfaces per mass, high thermal conductivity and good compatibility with organic PCM (Badenhorst, 2019). Expanded graphite is a wonderful supporting material to keep the shape of CPCM and prevent leakage with relatively high PCM adsorption ratio (Tong et al., 2019).

EG has attracted intensive attention for its application in improving liquid leakage, whilst the heat capacity decreases inevitably as EG occupies the space of PCM. To minimize the decrease of heat capacity, it's of significant importance to determine the optimal percentage of EG in the CPCM. Song et al. (2019) prepared EG/dodecane CPCM by vacuum infiltration method. The scanning electronic microscope (SEM) images of EG and CPCM illustrate that the pore structures of EG are filled with dodecane and the vermiform structure of EG still keeps intact without agglomeration. The thermal conductivity of the shape-stabilized CPCM (SSCPCM) was as high as 2.2745 W/(m K) based on the hot-wire method, which was 15 times higher over that of dodecane (only 0.14 W/(m K)). A paper was used to study the formability of the CPCMs with different contents of EG. The results demonstrate that when mass fraction of EG is higher than 16 %, the PCM will be restricted in the porous matrix without leakage.

The long-term effectiveness of CPCM in thermal properties after hundreds of melting/freezing cycles has attracted great concern due to its critical importance in practical applications. To explore the effects of thermal cycles on properties of EG/paraffin CPCM, five EG/paraffin composites with different mass ratios were prepared and subjected to thermal cycling (Wang et al., 2019). The thermal conductivity greatly decreases after 100 thermal cycles. The thermal cycling also has a significant influence on electrical conductivity. Aljehani et al. (2020) established a numerical model to investigate the transient heat transfer in EG/n-tetradecane CPCM. A CPCM with aluminium nitride additives as heat transfer promoters was fabricated and exhibited desirable thermal management for a battery module (Zhang et al., 2020).

2.2 Graphene aerogel

Compared to EG, graphene aerogels (GAs) present higher conductivities, lower densities and much larger specific surface areas, though the fabrication of GAs is much more complicated than EG. The main drawbacks of organic PCMs, such as low thermal conductivity and leakage, can be effectively improved by combining with graphene aerogels.

Tang et al. (2019) fabricated GA/paraffin shape-stabilized PCM (SSPCM) by a vacuum impregnation method. Then they observed the morphology characteristics of the CPCM by SEM. The CPCM exhibited desirable thermal conductivity as high as 1.432 W/(m K). To further enhance the thermal properties of CPCM, the combination of GA with phosphorylated polyvinyl alcohol (PPVA) as a "double network" supporting material was carried out by a one-step method with polyethylene glycol (PEG) as PCM (Shen et al., 2019), and a polyvinyl alcohol (PVA)/GA/PEG CPCM was fabricated for comparison. The leakage tests demonstrated that the addition of PPVA and PVA greatly improved the liquid leakage for PEG-based CPCM, and the efficiency of PPVA was more significant than PVA. Since phase change process of paraffin in GA skeleton with micro-sized pores is difficult to be observed directly, numerical simulation was applied to investigate the temperature distribution and the solid-liquid interface distribution for the phase transition processes (Feng et al., 2020). The results showed that GA obviously accelerated the melting and solidification processes.

2.3 Carbon foam

Carbon foam is another member of the carbon-based porous nanomaterials featuring excellent thermal conductivity, ultralight weight, desirable adsorption capability for organic PCM, and robust mechanical integrity to improve the leakage issue during liquid state.

Carbon foams (CFs) can be synthesized from low-cost melamine foams (MFs) (Maleki et al., 2020). In this work, CF/PEG, CF/paraffin (PA), and CF/palmitic acid (PAA) CPCMs were prepared by vacuum impregnation method. The observation investigation for heat transfer rate illustrates that the presence of the carbon-induced network in PCMs brings to a rapid thermal energy storage/release rate during the phase transition process. A CPCM consisting of liquid paraffin wax and 2 wt% graphene nanoplatelets was adsorbed by carbon foam matrix via vacuum infiltration method (Ola et al., 2019). The carbon foams exhibited excellent mechanical properties to keep the shape stability of the CPCM without destroying the structure of the CPCM mixture. In addition to the work aforementioned, Kim et al. (2019) added Ag, Al, carbon nanotubes, and graphene to CF/erythritol CPCM as nanoadditives to enhance heat transfer. Wilson et al. (2019) prepared CF/paraffin CPCM with various wax loadings (50.5 % to 82.6 %) and investigated the melting/freezing characteristics as well as the form stability.

3. Application

The demand for PCMs with high thermal conductivity, anti-leakage property and acceptable decrease of heat capacity is becoming more and more urgent with the extensive application of PCMs in thermal storage systems for heating and cooling applications. According to the previous research, CPCM consisted of organic PCM and 3D porous carbon-based matrix has greatly promoted the development of PCM in industry, including thermal management (Sheng et al., 2020), light-thermal conversion (Wu et al., 2020) and electricity-thermal conversion (Cao et al., 2019). The list of applications of CPCM are shown in Table 2. Carbon-based CPCMs have shown great potential in thermal management and thermal conversion, but it is still far from full utilization. It is worth exploration to develop CPCM for energy saving in buildings, for cooking in regions rich of sunlight, for shape flexibility in certain industries.

3.1 Thermal management

For industry application, it is of great importance to keep the temperature varying in an allowable range, especially for electronic products, the lifespan of these products highly relies on the operation temperature. To cool down the batteries more rapidly, an aluminum nitride/paraffin/expanded graphite/epoxy resin CPCM was fabricated for 18650 $LiFePO_4$ battery module (Zhang et al., 2020). This battery with CPCM thermal management system exhibits excellent heat dissipation capability and temperature uniformity with less than 1 ℃ temperature difference even at a high discharge rate of 3 C. A paraffin with low thermal conductivity was combined with textile-structured carbon scaffolds to enhance the heat transfer (Sheng et al., 2020). Reduced graphene oxide aerogel beads/1-tetradecanol paraffin exhibits great potential for thermal management applications attributing to its ultrahigh latent heat up to 230.3 J/g and superior thermal stability with 96.6 % efficiency after 50 melting/solidification cycles (Zhao et al., 2019). Graphene oxide-based SSPCM shows an improvement in light-thermal conversion performance and thermal management capacity (Zhao et al., 2019).

3.2 Energy conversion

N-alkanes are regarded as the most attractive PCMs due to their superior properties, including desirable heat capacity, nontoxicity and nonsubcooling, whilst the main drawbacks of n-alkanes are the low thermal conductivity and liquid leakage. An introduction of carbon nanotubes (CNTs) into nanofibrillated cellulose/noctacosane CPCM effectively increases the solar-thermal conversion and storage efficiency up to 83.4 % attributing to the function of CNTs as perfect photo captors and molecular heaters (Su et al., 2020). Carbon nanotubes combined with hexadecyl acrylate also exhibit better light-thermal and electricity-thermal performances (Cao et al., 2019). The CNT/polystyrene fabricated by Maleki et al. led to an increase in the light absorbance efficiency as well as the light-thermal conversion efficiency (90 %) (Maleki et al., 2019). Carbon foam is another porous supporting material that can cause a significant increase in light-thermal energy conversion efficiency and a decrease in the temperature fluctuation (Maleki et al., 2020). A paraffin-based CPCM with reduced graphene oxide working as a light absorption medium also has a great potential in solar-thermal conversion and storage application (Wu et al., 2020). Zhao et al. (2019) prepared a novel CPCM emulsions based on paraffin.

Table 2: List of applications of CPCM

Application	Efficiency	Reference
Thermal management	Null	Maleki et al., 2019
Thermal management for batteries	Null	Zhang et al., 2020
Light-thermal conversion	83.4 %	Du et al., 2020
Light-thermal conversion	~ 90 %	Maleki et al., 2019
Light-thermal conversion	88.1 %	Maleki et al., 2020
Light-thermal conversion	81.62 %	Tang et al., 2019
Light-thermal conversion	Null	Wu et al., 2020
Light-thermal conversion	Null	Zhao et al., 2019
Light-thermal conversion	80.1 %	Zhou et al., 2019
Electricity-thermal conversion	84.4 %	Zhou et al., 2019

4. Conclusions

The main drawbacks of organic PCMs are low thermal conductivity and liquid leakage. The development of carbon-based porous matrix provides a desirable solution to this issue. This paper reviews the newest development of CPCM with 3D porous carbon matrix as supporting material and organic material as PCM. The fabrication and thermal performance of the CPCM are illustrated in detail, and the applications of the CPCM are discussed. According to the literatures previously published, carbon- based scaffold can significantly enhance the heat transfer and improve the leakage without lower the latent heat too sharply. It exhibits great potential to promote the energy conversion and storage efficiency, including light-thermal conversion, electricity-thermal conversion and light-electricity conversion.

A lot of work has been focused on experiments, but the phenomena inside cannot be observed visibly, such as the inner fluid flow during the melting process and the cooling process. Simulation offers a perfect solution to this issue. Among all the simulation methods, molecular dynamics is one of the future hot topics.

To further enhance heat transfer, many novel materials will be introduced into this field. Metal organic frameworks and covalent organic frameworks are the two novel materials showing superior capability to conduct heat and electricity, and their potential to optimize CPCM is being explored.

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