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Preparation and Performance Analysis of Graphite Additive/Paraffin Composite Phase Change Materials

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Abstract: In the thermal energy storage system, the thermal properties of phase change materials (PCM) have a great influence on the system performance. In this paper, paraffin-based composite phase change material with different graphite additive (expanded graphite, EG; graphene, GR; and graphene oxide, GO) and different concentrations (0.5 to 2.0%) are manufactured by a two-step method combining mechanical agitation and ultrasonic vibration. The characteristics of charge/discharge processes are studied, and the thermophysical properties are measured by T-history method. The experimental results show that the thermal conductivity and heat charge rate of the composite PCM are effectively improved by adding the graphite additive to the PCM, and the addition of additives can improve the melting point of the material. When the same graphite additive is added, the higher the concentration, the higher the thermal conductivity of the composite PCM, and the latent heat decreases with the increase of concentration. When adding the same amount of carbon additives, the graphene/paraffin composite PCM has the highest heat charge/discharge rate and thermal conductivity. It is finally concluded that graphene is the most promising candidate for heat transfer enhancement of paraffin among three carbon additives even though the EG-based composite PCM gives relatively high latent heat.

Keywords: composite phase change material; nanomaterials; thermal management; thermal conductivity; graphite additives

1. Introduction

Thermal energy storage system has attracted much attention due to their low cost and high flexibility, but it has the disadvantage of low heat storage density. Because of the higher energy storage density and the smaller temperature change, using phase change material (PCM) becomes an effective way to solve this problem. However, the lower thermal conductivity limits the application of PCM. In order to overcome this problem, the scholars have attempted to add the high thermal conductive materials into the PCM to make new composite phase change materials (CPCM) to improve the thermal performance of thermal energy storage system [1–3]. CPCM can not only overcome the defects of single organic or inorganic PCM but also improve the application effect and application scope of PCM [4]. The phase transition temperature of CPCM, which has a good stability, can be chosen in a larger temperature range, and the latent heat of CPCM is greater than that of traditional PCM. The main additives are nanomaterials [5–8], metal particles [9–13] and carbon-based materials [14–16].

Among them, carbon-based materials have been extensively studied because of their high thermal conductivity and complex heat transfer mechanisms [17]. For the preparation of composite PCM using

carbon-based materials as additives, local and foreign scholars have also conducted in-depth research. Mehrali et al. [18] studied the preparation, characterization, and thermal properties of graphene oxide sheets/paraffin CPCM prepared by vacuum impregnation. The experimental results showed that the latent heat of the CPCM is 63.76 kJ/kg. The thermal conductivity of the new form-stable CPCM had a high increase from 0.305 to 0.985 (W/m·K). Xiao et al. [19] prepared graphite foam/paraffin composite with vacuum assistance. The structure and thermophysical properties of the CPCM were characterized. The results indicated that the thermal conductivity and effusivity of the CPCM are much higher than those of pure paraffin. Xia et al. [20] prepared and characterized an expanded graphite/paraffin CPCM in which the additive amount of expanded graphite (EG) is 0–10% by weight. The test system shows that the heat charge and discharge rate of EG/paraffin composites are greatly improved compared with pure paraffin. Chen et al. [21] prepared a dodecane/expanded graphite CPCM. The experimental results show that the latent heat of the CPCM decreases with the increase of expanded graphite, and the thermal conductivity increases with the mass fraction of the expanded graphite. Kant et al. [22] carried out the numerical investigation for the melting of graphene nanoparticles CPCM. They found that the addition of additive increased the charge rate, but can also hinder the heat convection of the CPCM. Zou et al. [1] found that compared with MWCNT, the addition of graphene has a greater increase in thermal conductivity of CPCM. In addition, the variation of latent heat is different from the theoretical calculation, which is lower than the theoretical value.

In recent years, although extensive research has been carried out on graphite-based additive CPCM, there is a lack of comparative analysis of the thermal properties of different graphite-based CPCM at a relatively low load, and the analysis on the melting point of phase change materials is also rare. Therefore, the objectives of this article are to analyze the heat charge/discharge characteristics of paraffin–graphite additive (expanded graphite, EG; graphene, GR; and graphene oxide, GO) CPCM and to analyze the thermophysical properties of CPCM containing different concentrations (0.5%, 1.0%, 1.5%, and 2.0%) and different types of graphite materials (EG, GR, and GO). The following studies are carried out: (1) different types of paraffin/graphite additive CPCM are prepared by a two-step method; (2) the characteristics in charge and discharge processes of CPCM are analyzed; (3) the thermal physical parameters of CPCM are tested by the T-history method, and their thermal conductivity, melting point, and latent heat are specially analyzed.

2. Materials and Experimental Methods

2.1. Material and Preparation

Industrial paraffin is used as PCM in this study. The additives are expanded graphite (EG), graphene oxide (GO) and graphene (GR). The properties of materials are shown in Table 1, and the SEM photographs are shown in Figure 1.
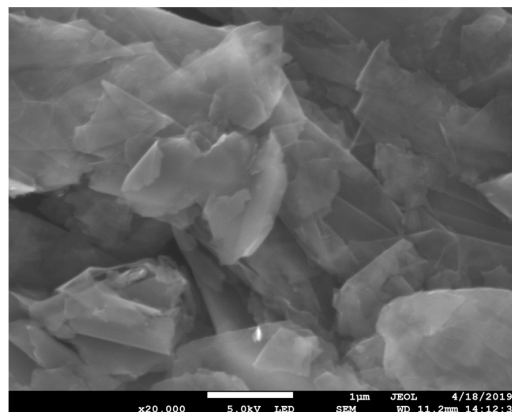
Table 1. The properties of materials.

Paraffin	Thermal conductivity (W/m·K)	0.2
	Melting point (°C)	52
	Density (kg/m^3)	800
	Latent heat (J/g)	160.8
Expanded graphite	Carbon content (%)	≥99.5
	Particle size (mesh)	325
	Expansion ratio (times)	40~425
Graphene	Thickness (nm)	0.55~3.8
	Specific surface area (m^2/g)	50~100
	Thermal conductivity (W/m·K))	~5000
Graphene oxide	Purity (%)	95~99
	Thickness (nm)	0.8~1.2
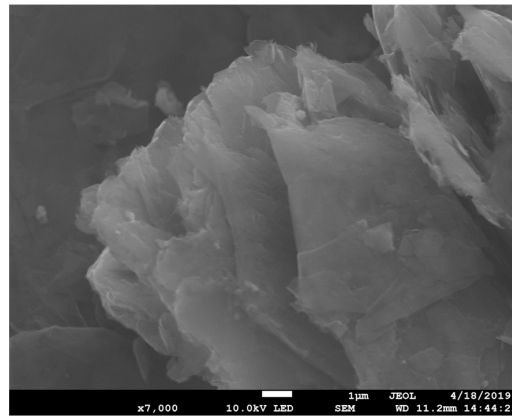
(**a**)
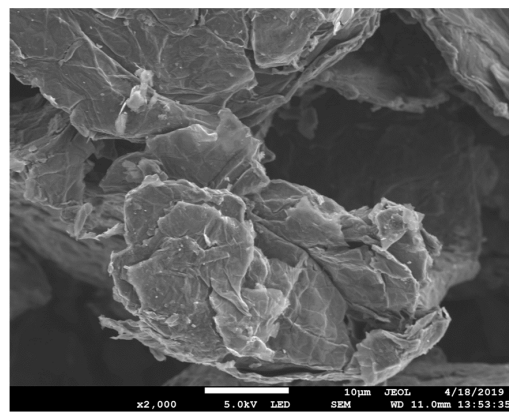
(**b**)

(**c**)

Figure 1. SEM images of (**a**) expanded graphite (EG), (**b**) graphene oxide (GO), (**c**) and graphene (GR) used in this study.

As shown in Figure 2, the carbon additive/paraffin CPCM are synthesized using a combination of mechanical agitation and ultrasonic vibration in a two-step process [23]. First, the paraffin is heated and completely melted in a 70 °C water bath, and weighed graphite additives are added into it at a stirring speed of 700 r/min for 1 h. Second, the mixtures are vibrated ultrasonically for 3 h in a thermostatic water bath to ensure complete mixing. Then, the samples are poured into the test tubes, in which thermocouples are inserted and cooled to solid of 20 °C to obtain the CPCM. The mass fraction

of graphite additive added are 0.5%, 1.0%, 1.5%, and 2.0%, respectively. The mixed samples are shown in Figure 3.
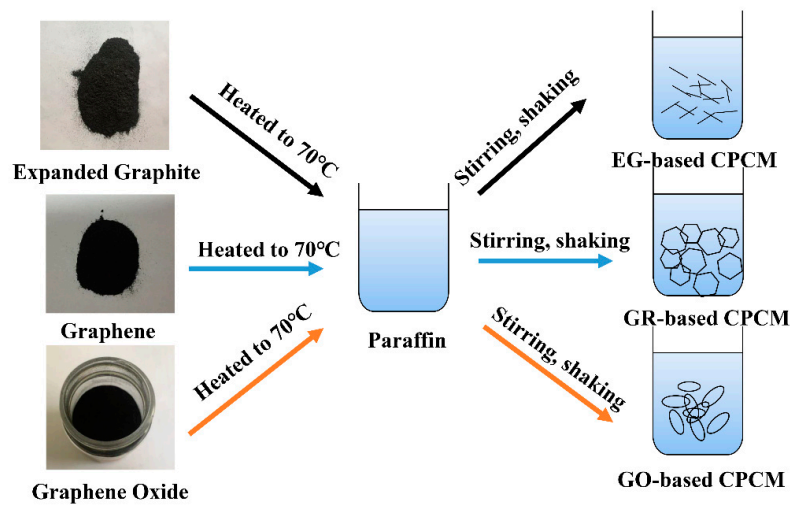

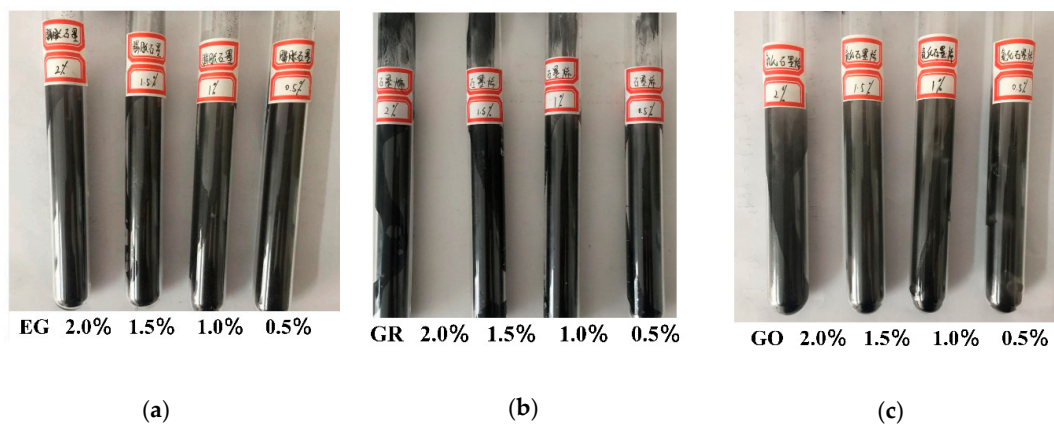**Figure 2.** The preparation system of composite phase change materials (CPCM).

Figure 3. Photographs of CPCM with (**a**) EG, (**b**) GR, and (**c**) GO.

2.2. Test System of Heat Transfer Performance

The experimental unit used in this paper is shown in Figure 4. The heat transfer performance test system consists mainly of two water baths (high-temperature, low-temperature), a data acquisition device, a computer, and some test tubes filled with thermocouples and samples. The thermostatic water baths are set at 70 °C and 20 °C. All thermocouples are calibrated with a measurement error of less than 0.1 °C. The temperature changes of all samples are recorded by a data acquisition device (34970A, Agilent Technologies, the United States). In this experiment, there are five samples in each group, each of which have a mass of 10 g.
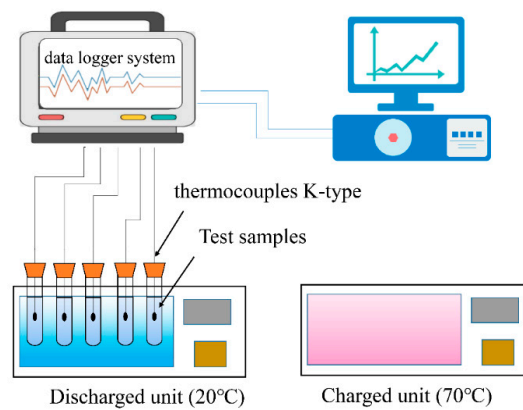
Figure 4. Experiment system.

2.3. Experimental Preparation

(1) Preparing stage: Number the test tubes filled with CPCM with different additives (GO, EG, and GR) and different loadings (0.5%, 1.0%, 1.5% and 2.0%). Put the test tubes into the low-temperature water bath for 2 h to unify their temperatures.

(2) Charge stage: Transfer the test tubes to the high-temperature water bath. The charge processes and the temperature changes of the CPCM are recorded by the digital camera and the data acquisition system, respectively. When the temperatures collected by the thermocouples are stable at 70 °C, the charge stage is deemed to have been completed.

(3) Discharge stage: After the charge stage, the test samples are rapidly transferred to the low-temperature water bath. The temperature changes of the CPCM are recorded by the data acquisition system until they are stable at 20 °C.

2.4. Measurements of the Thermophysical Properties

The thermophysical properties of the CPCMs are calculated by using the T-history method [24]. The formulas of T-history method can be expressed as:

$$c_{p,s} = \left(\frac{m_0 c_{p,0} + m_w c_{p,w}}{m} \right) \times \left(\frac{A_3}{A_2'} \right) - \frac{m_0}{m} c_{p,0} \tag{1}$$

$$c_{p,1} = \left(\frac{m_0 c_{p,0} + m_w c_{p,w}}{m} \right) \times \left(\frac{A_1}{A_1'} \right) - \frac{m_0}{m} c_{p,0} \tag{2}$$

$$k_s = \frac{1}{1 + \frac{c_{p,s}(T_m - T_\infty)}{H_m}} \cdot \frac{1}{4 \left[\frac{t_f (T_m - T_\infty)}{\rho R^2 H_m} - \frac{1}{2hR} \right]} \tag{3}$$

$$H_m = \left(\frac{m_0 c_{p,0} + m_w c_{p,w}}{m} \right) \times \left(\frac{A_2}{A_1'} \right) \times (T_0 - T_{m,1}) - \frac{m_0 c_{p,0}(T_{m,1} - T_{m,2})}{m} \tag{4}$$

where A is the area of the different regions surrounded by the cooling curve and the coordinate axis, the cooling curves of different materials are shown in Figure 5. T_0 is the initial temperature, T_∞ is the environment temperature, m is the mass of the PCM, m_0 is the mass of tube, m_w is the mass of water, $c_{p,w}$ is the specific heat of water, $c_{p,0}$ is the specific heats of the tube, h is the heat transfer coefficient of air, R is the radius of tube, t_f is the time required for full solidification of the molten PCM, $c_{p,l}$ is the specific heat of the liquid PCM, $c_{p,s}$ is the specific heats of the solid PCM, and a is the thermal diffusivity of PCM. T_m, k, ρ, and H_m are the melting temperature, thermal conductivity, density, and the latent heat of CPCM, respectively. The specific heat, latent heat, and thermal conductivity of CPCM can be calculated by using the above formulas.
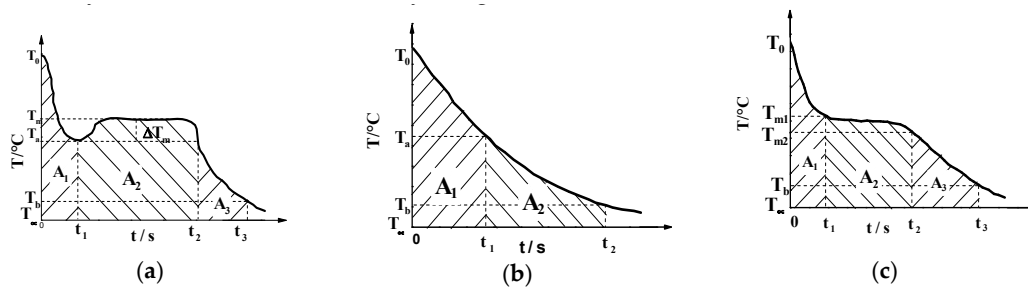
Figure 5. Cooling curve of phase change material (PCM) with (**a**) undercooling, (**b**) water, and (**c**) without undercooling.

3. Results and Discussions

3.1. Compatibility of Composite PCM

The gravity sedimentation method is used to verify the dispersion stability of the nano-suspension liquid. If the prepared nano-suspension has poor dispersion stability, the nanoparticles will quickly agglomerate into flocs and precipitate out. Finally, the agglomerated nano-particle floc sinks to the bottom of the test tube and there is a clear visible boundary line between the supernatant and the precipitation. The nanoparticle suspension with good dispersion effect is quite stable, without obvious flocculation and precipitation, and the structure is relatively stable.

In this experiment, different concentrations (0.5%, 1%, 1.5%, and 2%) of graphite additive/paraffin CPCM are prepared. To ensure the reliability of the experiment, the prepared PCM prepared must be a uniform and stable mixture. However, the preparation of CPCM often leads to precipitation, so sedimentation observations must be made for the prepared CPCM. Firstly, the prepared solution of the CPCM is placed in a constant temperature bath to be observed after standing for 4 h. Then the solution is cooled at room temperature, and the sedimentation is observed until it solidifies completely. Figure 6 shows the final state of the CPCM. There are pure paraffin, 2% expanded graphite/paraffin, 2% graphene oxide/paraffin and 2% graphene/paraffin from left to right in the photo. It can be seen that the CPCM have good dispersion stabilities.

Figure 6. The comparison of dispersion stability of CPCM. Left to right: pure paraffin, 2% EG/paraffin, 2% GO/paraffin, 2% GR/paraffin.

3.2. Analysis of the Charge Process

The temperature changes of CPCM during the charge process are shown in Figure 7. It can be seen that the charge process can be divided into three stages [23].
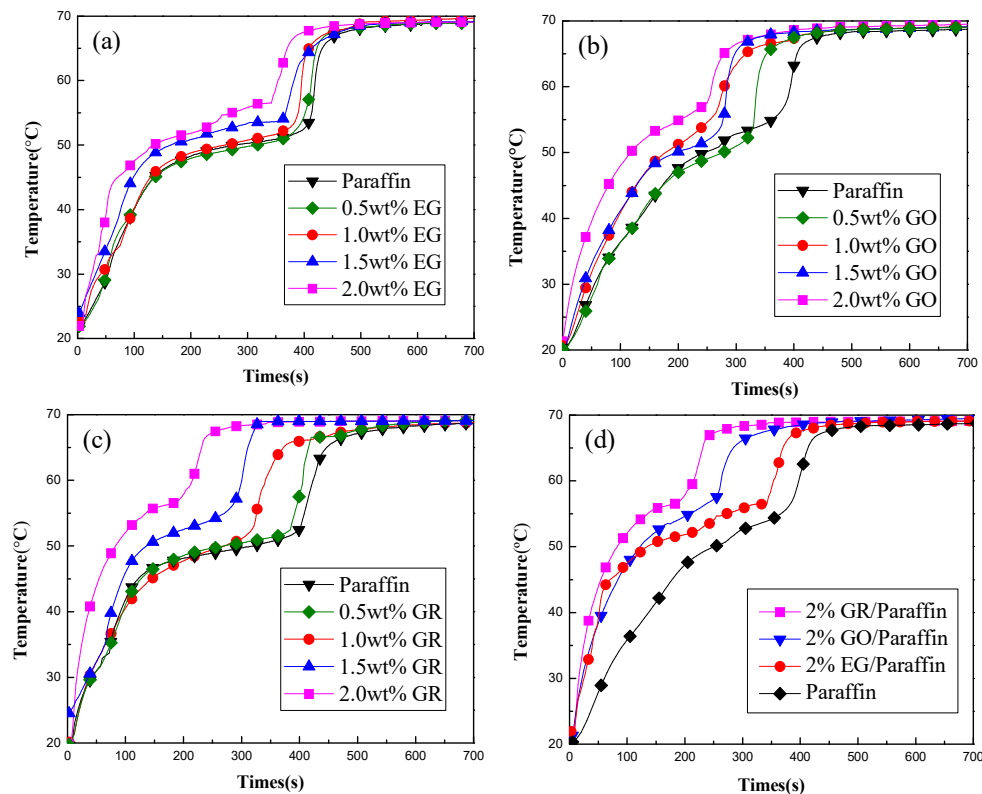

Figure 7. Melting curves of pure PCM and composite PCM with (**a**) EG as additives, (**b**) GO as additives, (**c**) GR as additives, (**d**) different additives at a concentration of 2%.

In the first stage, the temperatures of CPCM increase quickly from the initial temperature to the melting point. The temperature of all CPCM increases rapidly in an approximately linear trend. In this stage, heat conduction is the dominant way of heat transfer. When the concentration of additive is relatively low, the graphite-based additive increases the thermal conductivity of CPCM in the solid state, but the low additive density leads to an increase of the viscosity, which seriously hinders the natural convection of the CPCM. Therefore, the temperature curve at this stage is basically coincident with pure paraffin.

During the second stage, the CPCM begins to melt. Because of the latent heat, the temperature of the CPCM increased slowly. With the heat charge process progressed, the proportion of liquid CPCM gradually increase, and the heat transfer mode is transformed into the combination of natural convection and heat conduction.

During the last stage, the CPCM has been completely melted, and the temperature of the material increases dramatically due to the increase of natural convection intensity. The addition of additives greatly increased the viscosity of the CPCM [25]. The addition of graphite additives not only increased the thermal conductivity of the CPCM but also inhibited the natural convection of PCM due to the increase of viscosity. Because of this context, the heat transfer performance of CPCM depends on the interaction between viscosity and natural convection. At the beginning of the experiment, the CPCM is a solid stage with heat conduction as its main component. With the passage of time, the CPCM absorbs heat and heats up, and the portion close to the tube wall begins to melt gradually to form a thin liquid layer. As the phase change process progresses, a large number of solids melt into a high-temperature liquid. Due to the difference in density, the hot liquid PCM begins to move up. The edge portion of

the unmelted CPCM is washed during the upwelling process. The natural convection of the CPCM becomes the dominant mode of heat transfer and becomes more and more intense.

After the graphite additive is added to the PCM, the heat charge rate of the CPCM increases with the increase of addition amount. Whereas graphene heating rate is the fastest, expanded graphite is the lowest, and graphene oxide is located between the two. For example, for pure paraffin, the complete melting time is about 400 s. For the graphene-based CPCM, when the addition amounts are 0.5%, 1.0%, 1.5%, and 2.0%, respectively, the times are 390 s, 320 s, 295 s, and 195 s, respectively. In the case when the graphene oxide is added in the same amount, the times are 405 s, 340 s, 290 s, and 210 s, respectively. When the expanded graphite is added in the same amount, the times are 395 s, 390 s, 350 s, and 330 s, respectively. As shown in Figure 7d, compared with pure paraffin when the addition amount is 2%, the complete melting time of GR, GO, and EG are shortened by 51.25%, 47.5%, and 17.5%, respectively. Therefore, it can be seen from the heat storage curve that GR is the most effective additive for heat transfer enhancement of paraffin among the three additives in the present study.

Generally speaking, the mechanisms of thermal conductivity enhancement of nano-addition mainly includes the micro convection cell model, network structure, Brownian motion, and liquid layering theory. The size of the nano-addition plays a vital part in heat conduction in the micro state. [23]. The larger specific surface area of graphene two-dimensional sheet structure produces a lower interface thermal resistance. This sheet-like structure formation leads to a seepage model [26]. According to this model, the low heat flow resistance formed by the graphite-based additive of the sheet structure through the percolation network can explain the improvement in thermal conductivity [27].

3.3. Analysis of the Discharge Process

Figure 8 shows the discharge process of graphene CPCM at different concentrations. Like the charge process, the discharge process of CPCM can also be divided into three stages. As Figure 8 shows, the CPCM with different additive contents has a higher heat discharge rate compared with the pure paraffin during the entire discharge process.
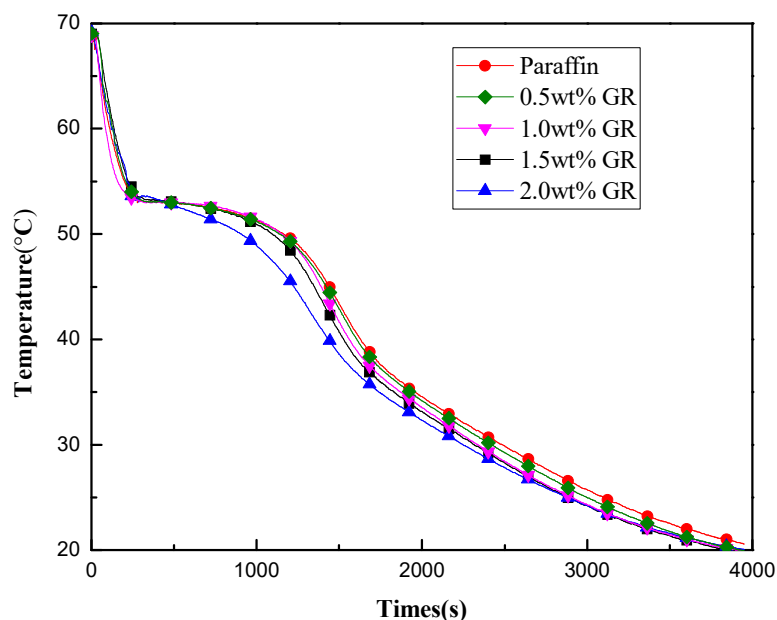

Figure 8. Melting Curves of GR/Paraffin CPCM with Different Concentrations.

At the beginning of the discharge process, the heat discharge rate is very fast, and their temperature reduction speed is not much different. This is due to the larger heat transfer area and temperature difference and the liquid state of PCM have heat convection. Therefore, the addition of graphene at this stage has a relatively small effect on heat transfer. Subsequently, the liquid PCM begins to solidify, and

its temperature is slowed down due to the latent heat of CPCM. When the PCM is completely solidified, their temperature reduction rate becomes larger, and the heat transfer mode is heat conduction. As the solid phase region continues to increase, the thermal resistance also increases, and the solidification process proceeds more slowly, so the PCM reaches full solidification after 1000 s. As shown in Figure 8, during the discharge process, the CPCM with high thermal conductivity transfers the stored heat in a shorter time, so that the temperature is reduced faster and the complete solidification time is the shortest when the additive concentration is 2%. As the concentration decreases, the solidification time becomes slower. This trend confirms the above conclusion: the higher the graphene concentration, the better the heat transfer performance. However, unlike the charge process, the temperature change rate of CPCM with different graphene concentrations during the discharge process is basically the same.

3.4. Thermophysical Properties

In this paper, the thermophysical properties of the graphite additive/paraffin CPCM are tested and calculated by the T-history method. The thermal conductivity, latent heat, specific heat capacity, and other parameters of the CPCM are shown in Table 2.

Table 2. The thermophysical properties of CPCM.

CPCM	Melting Point (°C)	Latent Heat (J/g)	Liquid Specific Heat Capacity (J/kg·K)	Solid Specific Heat Capacity (J/kg·K)	Thermal Conductivity (W/m·K)
Pure paraffin	52 °C	146.97	2773.758	3886.135	0.201
0.5% EG	51 °C	137.56	2683.367	3623.256	0.224
1.0% EG	51 °C	125.47	2522.432	3418.309	0.239
1.5% EG	52 °C	113.98	2384.305	3178.919	0.252
2.0% EG	53 °C	101.84	2240.271	3085.336	0.272
0.5% GO	51 °C	129.41	2632.573	3609.049	0.244
1.0% GO	51 °C	119.57	2338.629	3240.265	0.266
1.5% GO	52 °C	108.69	2215.676	3189.368	0.286
2.0% GO	55 °C	100.76	2102.365	3055.634	0.309
0.5% GR	52 °C	131.60	2409.717	3224.259	0.266
1.0% GR	52 °C	118.51	2144.818	3153.062	0.289
1.5% GR	55 °C	106.12	2031.645	3021.319	0.312
2.0% GR	56 °C	98.33	1931.629	3010.212	0.348

3.4.1. Thermal Conductivity

Figure 9 shows the relationship between graphite additive concentration and effective thermal conductivity. It can be seen from Figure 9 that at a certain concentration (≤2.0%), the thermal conductivity of CPCM increases linearly with graphite additive. The increase of thermal conductivity can accelerate the heat charge and discharge rate of CPCM and improve the thermal efficiency of CPCM. Under the same amount of addition, GR has the largest increase in thermal conductivity, EG has the smallest, and GO is between the two. For example, adding 2.0% of graphite additive, the ratios of thermal conductivity of the three CPCM to pure paraffin are 1.73, 1.35, and 1.54, respectively. Besides this, it can be seen from Figure 9 that the thermal conductivity of GR/paraffin CPCM is higher than that of other samples, especially at high concentrations. In addition, it can be seen from Table 2 that the addition of graphite additives can increase the melting point of CPCM to a certain extent. In this experiment, adding 2.0% GR will increase the melting point of the CPCM from 52 °C to 56 °C.
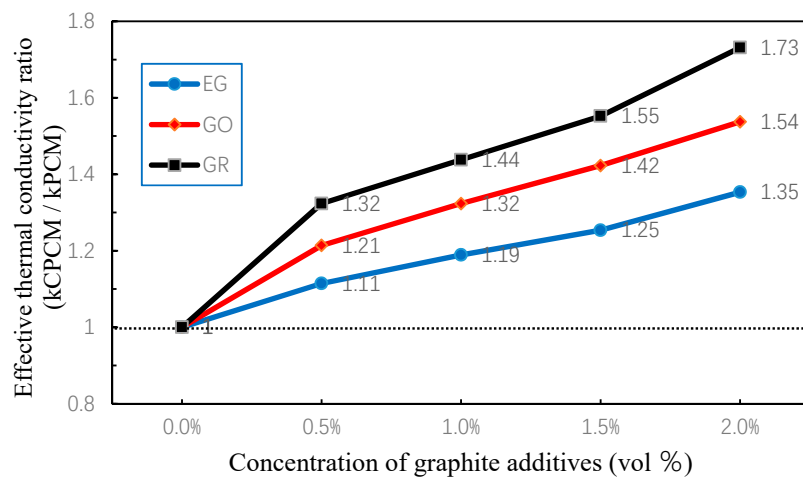
Figure 9. Effective thermal conductivity ratio as a function of the concentration of graphite additives.

3.4.2. Latent Heat

Figure 10 shows the latent heat of different graphite additive-based CPCM. As shown in Figure 10, the latent heat of CPCM decreases linearly with the increases of graphite additive. Since graphite additives (EG, GO, and GR) do not have latent heat, the latent heat of samples is theoretically the latent heat of the paraffin contained therein.

$$L_{\text{s,cal}} = L_p \cdot (1 - \varnothing) \tag{5}$$

where $L_{\text{s,cal}}$ is the calculated value of the latent heat of the CPCM, L_p is the calculated value of the latent heat of paraffin, and $\varnothing$ is the mass fraction of the graphite additive in the CPCM.
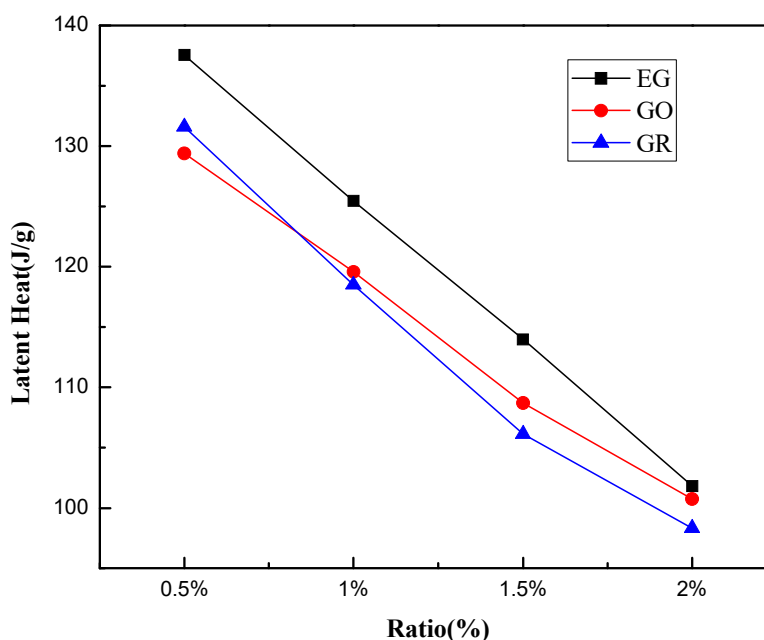

Figure 10. The latent heat of CPCM.

It can be seen from the above formula that the latent heat of the CPCM decreases with the mass fraction of additive, and the experimental results also show the same trend. We also find that the theoretical values are not completely consistent with the experimental values. On one hand, local agglomeration may occur in composite materials. On the other hand, the addition of additives,

especially EG, increases the molecular potential energy of the whole material. Within the mass fraction range of this experiment, the higher the addition amount, the more obvious the increase of the overall molecular potential energy of the material. Meanwhile, the porous structure of EG can make the two materials mix more evenly, so the latent heat of expanded graphite is slightly different from that of the other two additives. As Figure 10 shows, under the same added concentration, the latent heat of PCM with different additive are not the same. The latent heat of EG is higher than that of GO, while GO and GR are almost the same.

4. Conclusions

In this paper, a variety of graphite additives were added into paraffin respectively to improve the heat transfer performance of CPCM. The change and discharge processes of CPCM were collected and analyzed, and the physical parameters were measured by the T-history method. The conclusions are as follows:

The charge process of CPCM can be divided into three stages. When the amount of additive is the same, the graphene CPCM has the fastest charge rate, expanded graphite has the lowest, and graphene oxide is located between the two. The discharge process can also be divided into three stages, but compared with the charge process, the CPCM with different graphene concentrations have less influence in the discharge process.

In a certain range (≤2.0%), the higher concentration of the CPCM, the better the heat transfer performance. When adding the same concentration of additive, the heat transfer performance from highest to lowest is that of GR, GO, then EG. At the same time, the addition of additives can increase the melting point of CPCM, among which 2.0% GR increases the melting point the most.

Graphite additives do not have latent heat, so the latent heat of CPCM decreases linearly with the concentration of additives. However, since EG increases the molecular potential energy of the whole material, the latent heat of EG is higher than that of the other two materials.

It is finally concluded that GR is the most promising additive for heat transfer enhancement of paraffin among three carbon additives tested in the present study, even though EG gives the highest heat latent.

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