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An experimental investigation on the control mechanism and heat transfer enhancement of composite phase change materials

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ABSTRACT

In order to obtain phase change materials with suitable melting point, latent heat and thermal conductivity, this paper employed the liquid paraffin with a melting point of 17°C and solid paraffin with a melting point of 41°C in different proportions to fabricate PCM composites. By adjusting the mixing ratio of short-chain hydrocarbons and long-chain hydrocarbons in paraffin wax, phase change materials with different phase change ranges were obtained to meet the requirements of practical applications. A variety of copper foam /paraffin composite phase change materials with different porosities and pore densities were prepared by the melt dipping method. The influence of the porosity and pore density on the thermal conductivity of copper foam/paraffin composite was explored. The experimental results show that mixed paraffin can control the melting point and latent heat of composite phase change materials. Copper foam can greatly increase the thermal conductivity of composite phase change materials with the porosity has a greater impact and the pore density has a less effect. The thermal conductivity of composites can be increased by a maximum 20 times.

Keywords: Phase change materials (PCM); Composites; Control mechanism; Copper foam; Heat transfer enhancement

1. INTRODUCTION

Phase change materials (PCMs) absorb or release large amounts of latent heat during phase transitions, thereby they are widely used in building energy saving, indoor warming, temperature adjustable textiles, military and aerospace, etc. [1-3]. Phase change heat storage materials mainly include inorganic phase change materials and organic phase change materials. Inorganic materials are mainly crystalline water and salts, molten salts, metals and alloys, etc. Inorganic materials have high heat storage density, fixed melting point, higher thermal conductivity, and they are generally neutral [4, 5], such as $CaCl_2 \cdot 6H_2O$, $Na_2CO_3 \cdot 10H_2O$, $NaSO_4 \cdot 5H_2O$. Crystal water and salts suffer from problems such as supercooling and phase separation, affecting their widespread applications [6]. Metals and metal alloys usually have high phase transition temperatures, which are characterized by large thermal conductivity and fast heat storage and exothermic speeds [7]. Organic materials are mainly paraffin, fatty acids, and polyols [8]. Compared with inorganic materials, organic materials have better molding effect during curing, and generally do not appear supercooling and phase separation phenomena. In addition, organic materials have stable performance, low toxicity and low cost. However, there are also problems in organic materials such as low thermal conductivity, small energy storage density, and easy volatility [9-12].

Low thermal conductivity, slow heat storage and release rates limit the practical applications of PCMs. Therefore, intensive investigations have been carried out to improve the thermal conductivity of PCMs [13-17]. Li et al. [18] mixed the expanded graphite (EG) with capric acid (DA) to prepare a capric acid / expanded graphite composite phase change material (DA/EG-PCMS) with high stability and good thermal conductivity, the thermal conductivity of composites increased with the increase of expanded graphite. Zeng et al. [19] added Ag nanoparticles to myristyl alcohol, and the thermal conductivity increased with the increase of Ag nanoparticles, and the phase transition temperature and latent heat decreased slightly. Wu et al. [20] prepared

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nanocomposite PCM by mixing paraffin with multi-walled carbon nanotubes (MWCNTs). The results showed that as the loading of carbon nanofillers increased, the thermal conductivity of the nanocomposite PCM increased, and the phase change enthalpy gradually decreased. Yang et al. [21] compounded copper foam and bottom fins into PCM in order to improve the heat transfer performance. High temperature water flowed through the copper tube as a heat transfer fluid (HTF). The temperature variations of the selected points inside PCM and the solid–liquid interface evolution in axial plane of symmetry for three samples, including that of pure paraffin, paraffin–copper foam composites without and with the bottom fin, were recorded under the different heating temperatures and flow rates of HTF. The experimental results showed that under the same operating conditions, the time required to completely melt the PCM in the composite material was 1/3 less than that of pure paraffin. Yang et al. [22] designed a visual experiment test bench to investigate the influence of metal foam on the melting process of paraffin wax. Experimental results showed that the open cell metal foam could significantly improve the efficiency of thermal energy storage. Compared with pure PCM, the complete melting time of composite PCM was reduced by 64 %, and the temperature distribution was more uniform.

The paraffin phase change process is stable, non-toxic, non-corrosive and low cost. However, the melting point of ordinary solid paraffin is generally around 40-70°C, while the melting point of liquid paraffin is relatively low. In order to obtain a more appropriate phase transition temperature and phase transition latent heat, it is necessary to modulate the paraffin wax. Since the longer the number of chain hydrocarbons, the longer the melting point of the corresponding paraffin wax material. It is able to obtain phase change materials with different phase change ranges by adjusting the mixing ratio of short-chain hydrocarbons and long-chain hydrocarbons in the paraffin wax to meet the requirements of practical applications. Although common pure alkane phase change materials can find the right temperature for practical applications, the cost is too high to be used alone. Therefore, in this paper, 17°C and 41°C pure paraffin were applied in different proportions to fabricate composite PCMs. After complete mixing, the melting point and latent heat change were measured by DSC to explore the control mechanism of organic phase change materials. In addition, copper foam with a porosity of 95% and a pore density of 15 ppi, 30 ppi, and 50 ppi and copper foam with a porosity of 85% and a pore density of 50 ppi were integrated with mixed paraffin to obtain PCM composites. Four copper foam /paraffin composite phase change materials were prepared by the melt impregnation method. The effect of copper foam on the heat transfer enhancement of PCM composites were investigated.

2. EXPERIMENTAL APPROACH

2.1 Paraffin wax blending experiment

In the experiment, the melting point of 17 °C and 41 °C paraffin supplied by Shanghai Joule Company was selected for blending. Among them, at a room temperature of 25°C, paraffin wax with a melting point of 17°C is a transparent liquid, and paraffin wax with a melting point of 41°C is a white solid particle, as shown in Fig. 1. Differential scanning calorimetry (German Netzsch DSC-200PCPhox) was used to characterize the phase transition behaviour of two pure paraffin waxes at a heating rating of 5°C/min.

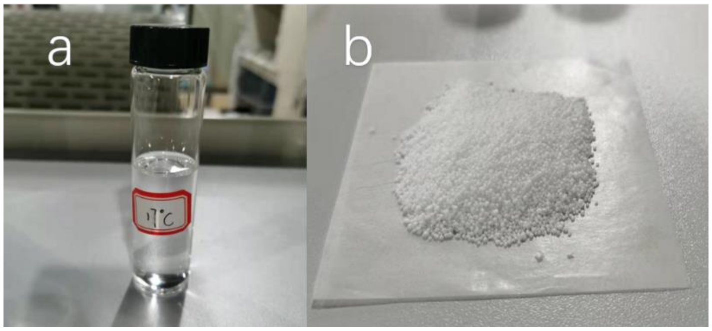

Fig.1. Paraffin wax with the melting point of (a) 17°C and (b) 41°C.

The measured DSC curves of the pure paraffin waxes are shown in Figure 2. The two paraffin waxes are accurately weighed and mixed. Taking into account the accuracy of the experiment, the mass of each sample was set at 40g. The preparation ratio is shown in Table 1, and the mass required for each paraffin wax was weighed with an electronic balance. The two sets of samples were heated in a water bath with an ultrasonic tip to make sure the samples are thoroughly mixed, the beakers were sealed during the process. Finally, the samples were put in the refrigerator to cool and solidify.

2.2 Heat transfer enhancement experiment of composite phase change materials

The copper foam used in this experiment was provided by Suzhou Jia Shi De Metal Foam Co., Ltd. Four types of copper foam with different specifications were selected, three of which had a porosity of 95% and a pore density of 15 ppi, 30 ppi and 50 ppi. The other has a porosity of 85% and a pore density of 50 ppi, as shown in Figs. 3 and 4. As metal foam has the advantages of high porosity and low density, it has little effect on the heat storage capacity of phase change materials. As a porous material, there are mainly two ways to characterize its structure: porosity and pore density. Porosity refers to the percentage of the pore volume to the total volume, and the pore density (pores per inch, ppi) is defined by the number of pores per inch of the porous material [23-26]. It can be seen from the intuitive diagram that as the pore density increases, the pore diameter of the copper foam decreases, and the diameter of the metal skeleton decreases. In addition, it can be seen from the visual diagram that the metal foam is an isotropic material, and its pore diameter and skeleton diameter in all directions are the same, which is determined by the metal foam production process.

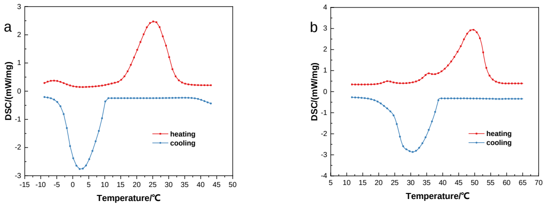

Fig.2. DSC curves of paraffin wax with melting points of (a) 17°C and (b) 41°C.

Table 1 Sample preparation ratio of group A.

Sample number	17°C	Mass（g）	41°C	Mass（g）
A1	20%	8	80%	32
A2	35%	14	65%	26
A3	50%	20	50%	20
A4	65%	26	35%	14
A5	80%	32	20%	8

Fig. 3. Copper foam with a porosity of 95% and a pore density of 15 ppi, 30 ppi and 50 ppi.

Fig. 4. Copper foam with a porosity of 85% and a pore density of 50 ppi.

As shown in the flowchart in Fig. 5, a sample of copper foam with a size of 100×100×5 mm was completely immersed in the molten mixed paraffin liquid and placed in the refrigerator to solidify. After the sample was completely solidified, it removed the excess paraffin by cutting. The prepared copper foam /paraffin composite phase change materials are shown in Figs. 6 and 7.

The experiment used a thermal conductivity meter produced by Nanjing Dazhan Electromechanical (DZDR-S) to measure the thermal conductivity. The instrument has a specified accuracy of ± 3 % depending on the sample size and conductivity. The five samples are: 80% 17°C+20% 41°C (B1) mixed paraffin wax; 15 ppi copper foam /paraffin (B2), 30 ppi copper foam /paraffin (B3), 50 ppi copper foam /paraffin (B4) with a porosity of 95%; 50 ppi copper foam /paraffin (B5) with a porosity of 85%, as shown in Table 2. Each sample was measured 5 times, and the interval between each test was 5 minutes. During the test, the nickel metal probe was clamped between the two samples, the smooth surface of the sample was in contact with the probe, and the two were clamped to reduce the contact thermal resistance.

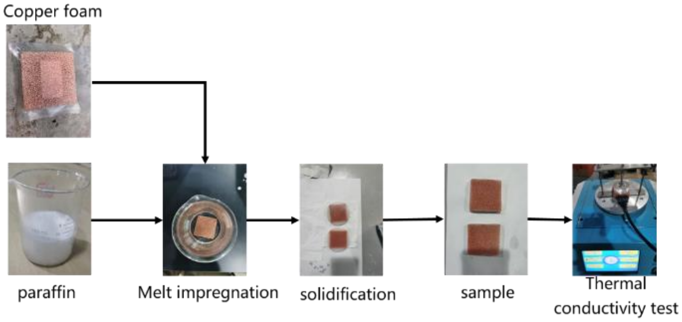

Fig. 5. Preparation process of copper foam/paraffin PCM composites.

Fig. 6. Three copper foam /paraffin composite phase change materials with a porosity of 95% and a pore density of 15 ppi, 30 ppi and 50 ppi from left to right were prepared.

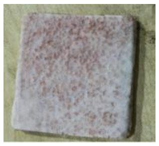

Fig.7. Copper foam /paraffin composite phase change material with a porosity of 85% and a pore density of 50ppi was prepared.

Table 2 Prepared five composite phase change materials.

Element	B1	B2	B3	B4	B5
Paraffin	80% 17°C+20% 41°C	80% 17°C+20% 41°C	80% 17°C+20% 41°C	80% 17°C+20% 41°C	80% 17°C+20% 41°C
Porosity	_	95%	95%	95%	85%
Pore ensity	_	15 ppi	30 ppi	50 ppi	50 ppi

3. RESULTS AND DISCUSSION

3.1 DSC test results

A sample size of 20 mg for samples A1-A5 were placed in aluminum crucible for DSC measurement. The temperature measurement range is -20°C to 100°C and both the heating and cooling rate are 5°C /min. The DSC curves of samples A1-A5 are shown in Figs. 8 and the thermal properties of samples are summarized in Table 3.

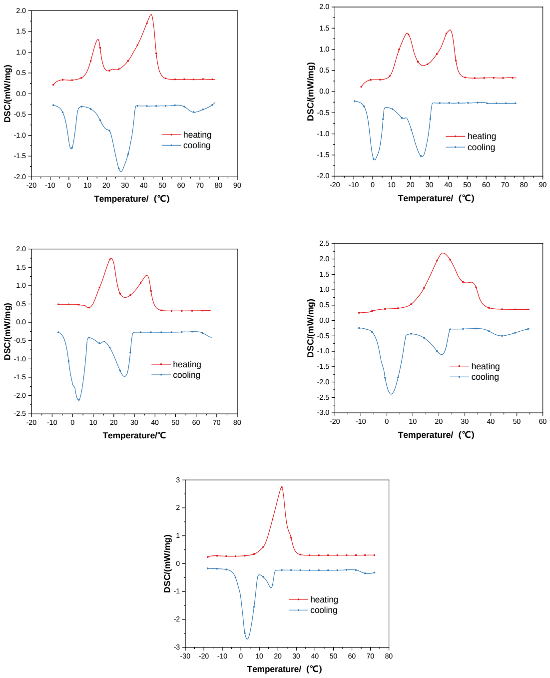

Fig. 8. DSC curves of sample A1-A5.

It can be seen from Figs. 8 that sample A1 (20% 17°C+80% 41°C), sample A2 (35% 17°C+65% 41°C) and sample A3 (50% 17°C+50% 41°C) have two melting peaks. The starting points of the low-temperature melting peaks are 9.3°C, 9.2°C and 10.4°C respectively, which are all lower than the melting point of pure paraffin wax at 17°C. The starting points of the high temperature melting peaks are 32.1°C, 34.8°C and 30.0°C, which are all lower than the melting point of pure paraffin wax at 41°C. This is also in line with Schroeder's formula and low-temperature eutectic theory [27-29]. The composite of phase change materials is regarded as an ideal solution model, and the melting temperature of the composites is always lower than that of the pure compound. Under the interaction of long- and short-chain hydrocarbons, the temperature of the starting point of the high and low melting peaks will decrease. This might provide ideas for reducing the melting temperature of phase change materials with specific melting points.

Table 3 Thermal properties of paraffin and samples A1-A5.

Type	Melting area (J/g)	Solidification area(J/g)	Peak(°C)		Starting point （°C）	
			Low peak	High peak	Left	Right
17°C paraffin	151.6	-142.6	25.8		15.8	
41°C paraffin	204	-210.4	49.4		40.2	
A1: 20% 17°C+80% 41°C	165.2	-151.9	12.9	41.9	8.1	32.9
A2: 35% 17°C+65% 41°C	149.3	-140.6	15.2	39.6	8.5	31.9
A3: 50% 17°C+50% 41°C	199.4	-192.8	17.6	35.5	9.2	28.5
A4: 65% 17°C+35% 41°C	159.6	-154.6	21	32.5	11.4	26.5
A5: 80% 17°C+20% 41°C	177	-179.2	19.9		11.2	

It can be seen from Figs. 8 that sample A4 (65% 17°C + 35% 41°C) and sample A5 (80% 17°C + 20% 41°C) have only one melting peak. This is actually a combination of high and low melting peaks. In the two sets of samples, 41°C paraffin accounts for 20% and 35%, respectively, and 17°C paraffin accounts for more. The melting points of the two are quite different and they are evenly mixed. Long-chain hydrocarbons are wrapped by short-chain hydrocarbons, thereby the low-temperature melting peak does not appear. It can be seen from the figure that there is a bulge on the right side of the melting peak, sample A4 is more obvious, and sample A5 has almost completely merged into one peak. This is attributed to the proportion of 41°C paraffin in sample A5 is lower. As shown in Table 3, as the proportion of paraffin wax at 17°C increases, the proportion of paraffin wax at 41°C decreases. The starting point of the low temperature melting peak of mixed paraffin wax gradually increases, and the starting point of the high temperature melting peak gradually decreases, at the ratio of 80% 17°C +20% 41°C, it completely merges into one peak. At this time, the melting point is 11.2 °C, and the latent heat is 177J/g, which is between 17°C and 41°C.

According to the above results, it can be concluded that after the two pure paraffin are mixed, under the interaction of long and short chain hydrocarbons, the temperature of the starting point of the high and low melting peaks will decrease, but the latent heat of the mixture is between the two pure paraffin.

3.2 Thermal conductivity test results

Figure 13 shows the thermal conductivity test results of composite PCMs. The average value of the five thermal conductivity tests of mixed paraffin is 0.22w/m·k. After adding 15 ppi copper foam with a porosity of 95%, the thermal conductivity of the composite material is 0.79 w/m·k. The thermal conductivity of 30 ppi copper foam with 95% porosity is 1.21 w/m·k, the thermal conductivity of 50 ppi copper foam with 95% porosity is 1.34 w/m·k, and the thermal conductivity of 50 ppi copper foam with 85% porosity is 4.35 w/m·k.

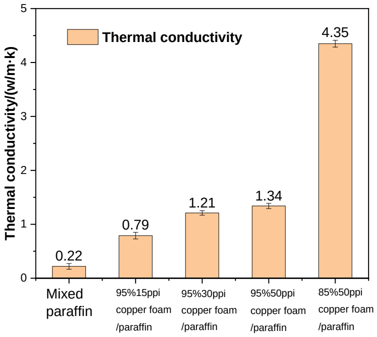

Fig.13. Thermal conductivity test results of copper foam/paraffin composite phase change materials.

It can be seen from the figure that the copper foam can greatly increase the thermal conductivity of the organic phase change material paraffin wax. The thermal conductivity of copper foam with 95% porosity and 50 ppi is 1.34 w/m·k and the thermal conductivity of copper foam with 85% porosity and 50 ppi is 4.35 w/m·k. therefore, the porosity plays a major role and it has great effect on the heat transfer enhancement of composites. Compared with the mixed paraffin, the thermal conductivity of composites with 85% porosity and 50 ppi is increased by 20 times. When the porosity is 95%, the pore density will also affect the thermal conductivity of the copper foam/paraffin composite material, and it will increase as the pore density increases. However, the effect is not as obvious as the porosity. Compared with pure paraffin, the thermal conductivity of samples B2-B4 are increased by 4 to 6 times.

With the same pore density, the porosity decreases from 95% to 85%, and the thermal conductivity increases significantly. This is because when the porosity decreases, the paraffin that can be filled in the porous structure decreases, and the proportion of copper will increases. Although the thermal conductivity is greatly improved, the heat storage capacity of the composite phase change material will be reduced. When the porosity is the same, the thermal conductivity will increase as the pore density increases, but the change is relatively small. This is due to increasing pore density leads to the pore diameter of the copper foam decreases and the density of the metal skeleton decreases, thereby the copper foam with a higher density can achieve a better heat transfer enhancement.

4. CONCLUSIONS

It can be concluded that after the two pure paraffin are mixed, under the interaction of long and short chain hydrocarbons, the starting point temperature of the high and low melting peaks of the mixed paraffin is lower than the melting point of the two pure paraffin. The latent heat of the mixture is between the two pure paraffin waxes. When 17°C paraffin wax was mixed with 41°C paraffin wax, as the proportion of 17°C paraffin wax increased, the proportion of 41°C paraffin wax decreased. The starting point of the low temperature melting peak of mixed paraffin wax gradually increased, and the starting point of the high temperature melting peak gradually decreased. At the ratio of 80% 17°C+20% 41°C, it completely merges into a peak. At this time, the melting point is 11.2°C, and the latent heat is 177J/g.

Copper foam can greatly increase the thermal conductivity of composite phase change materials. The porosity plays a major role, and the pore density will also affect the thermal conductivity of the copper foam /paraffin composite material, and it will increase with the increase of the pore density. When the porosity is 95%, the thermal conductivity of composites is 4 to 6 times higher than that of mixed paraffin. When the porosity is 85%, the thermal conductivity of composites is 20 times higher than that of mixed paraffin.

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