

## Research Article

# Effects of Nanofilled Particle Forms and Dispersion Modes on Properties of Carbon-Based Energy Storage Composites

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How to improve the thermal conductivity of phase change materials (PCMs) is always the key to thermal control technology. At present, the thermal conductivity of PCMs has two ways to improve: one is to fill the matrix with high thermal conductivity and the other is to fill nanoparticles. After combining the two methods, the choice of filled nano-SiO<sub>2</sub>, carbon nanotubes (CNTs), or graphene (GNPs) has different effects on the performance of carbon-based energy storage composites. Filling paraffin with foamed carbon increased the thermal conductivity of pure paraffin from 0.25 W/(m·K) to 8.3083 W/(m·K), an increase of 33.2 times. When the nanoparticle mass fraction is 5%, the enthalpy of GNP composites is 10 J·g<sup>-1</sup> less than that of SiO<sub>2</sub> composites. Under the same mass fraction, compared with the thermal conductivity enhancement effect of SiO<sub>2</sub> composites, the thermal conductivity increase effects of CNTs and GNP composites are 6.7 and 15.8 times the thermal conductivity increase of SiO<sub>2</sub> composites, respectively. The comparison of theoretical and experimental values shows that different nanoparticle forms and dispersion modes have different effects on the performance of carbon-based energy storage composites, among which GNPs have the greatest improvement in the thermal conductivity of carbon-based composites.

## 1. Instruction

Phase change energy storage materials are the basis of research on phase change energy storage technology [1, 2]. The selection of appropriate PCMs is of paramount importance. At present, paraffin with the advantages of chemical stability, no overcooling, no toxicity, high latent heat of phase change, cheapness, etc. has become a common PCM. However, paraffin has problems such as low thermal conductivity during the phase change process, which makes it poor in thermal conductivity. It cannot crystallize and release heat in time at the phase change temperature, which affects the performance of the phase change thermal control system. Seriously, it can even cause thermal control system failure [3].

Therefore, an important basic research work on energy storage technology is the development of phase change energy storage materials [4]. Using nanoparticles to fill phase change materials has made many achievements in improving thermal conductivity now. The first method is to fill the nanoparticles with PCMs [5, 6]. Amin et al. [7] studied the effect of nanoparticles on the physical and thermal properties

of materials. DSC analysis showed that the latent heat of 0.3 wt.% beeswax/GNP increased by 22.5%. The thermal conductivity of 0.3 wt.% beeswax/GNP is 2.8 W/(m·K). The latent heat and thermal conductivity of beeswax were improved by the presence of GNP nanoflakes. Therefore, beeswax/GNP has the potential to reduce building energy consumption. Jesumathy et al. [8] designed an energy storage system using embedded nano copper oxide (CuO) particles to study the thermal properties of paraffin. In this study, 40 nm CuO particles with average particle sizes of 2%, 5%, and 10% are dispersed in PCMs. The results show that with the increase of the mass fraction of CNEPs, the thermal conductivity increases by 6.7% and 7.8% in the liquid state and 5.14% and 30% in the dynamic viscosity. The thermal conductivity of composite materials is increased by 1.3 times. At the maximum flow rate, the heat transfer coefficient during solidification increased by about 78%. This shows that adding copper oxide nanoparticles to paraffin promotes conduction and natural convection very effectively in composites and paraffin. The second method is to fill PCMs into a highly thermally conductive matrix (such as graphite foam (CF20)

[9, 10], “vascular” resin-based composites [11]). Huang et al. [12] prepared a myristyl alcohol (MA)/metal foam composite phase change material by vacuum infiltration. MA is used as phase change material, and metal foam is used as a skeleton. Comparisons are made using foamed nickel and copper foam. The influence of the pore size of foam metal on its thermal properties is analyzed. Compared with pure MA, the latent heat of fusion of metal foam composite phase change materials (CPCMs) is reduced by 3-29%. In addition, the composite material has good thermal stability and the foam metal has a good adsorption effect on PCM. The thermal conductivity of the composite is tested with a thermal conductivity meter (TCM), and the results show that the MA/foam metal composite has good thermal conductivity. Both of the above methods have effectively improved the performance of PCMs.

The combined use of the two methods synergistically increases the thermal conductivity of composite PCM [13]. Nada et al. [14] added multiwalled CNTs to a paraffin-CF20 matrix to achieve a synergistic increase in thermal conductivity. However, little research has been done on the effect of nanoparticles in different particle forms on the thermal conductivity of composite PCMs. The mechanism by which nanoparticles enhance synergistic PCM thermal conductivity is still unclear. Here, we explore this scientific issue by analyzing the effect of nanoparticle form and dispersion on the thermal conductivity of composite PCMs. It is found that under the same mass fraction, the solidification and melting enthalpy of SiO<sub>2</sub> composite and GNP composite are different and that SiO<sub>2</sub>, CNTs, and GNPs have very different thermal conductivity enhancement of the composite, and with the increase of the content, the increase in the respective thermal conductivity is also significantly different. Therefore, under the same mass fraction, the different particle forms and dispersion modes of the nanoparticles will have a great impact on the change of the thermal conductivity of the composite material, which will serve as a reference for future research.

## 2. Materials

After the paraffin with purity of 99% is melted, 0 wt.%, 0.5 wt.%, 1 wt.%, 2 wt.%, and 5 wt.% nanoparticles are filled therein, and the modified PCM is obtained by shaking and mixing uniformly. The template method is used to prepare porous foamed carbon with a porosity of 85% and an average pore size of 400 μm. The modified paraffin is well impregnated into CF20 under the vacuum and the adsorption of the foamed carbon pores, and the paraffin-nanoparticles are prepared by solidification of CF20 samples, namely, shaped phase change energy storage composites.

The relevant parameters of the selected nanomaterials are shown in Table 1.

The dispersion of nanoparticles is characterized by field emission scanning electron microscopy. The thermal conductivity of the composite is measured by the laser method. Nanoparticle materials are mixed with each other evenly.

## 3. Results and Discussion

*3.1. Effect of Particle Form on Properties of Carbon-Based Composites.* The effect of nanoparticle form on the properties of paraffin-CF20 composites is mainly reflected in thermal conductivity and solidification melting enthalpy. Figure 1(a) shows that the microstructure of SiO<sub>2</sub> is mainly spherical. CNTs are mainly two-dimensional tubular structures in Figure 1(b). Theoretically, heat flow will rapidly propagate in the radial direction of the nanotube, similar to the rapid transmission of current along the direction of copper wires with low resistance.

GNP has a sheet structure with wrinkles in Figure 1(c). Compared to CNTs, both the front and back of GNPs are in contact with a large number of GNP molecules, which greatly reduces the contact thermal resistance. The GNPs normally will wrinkle in the composites [15, 16]. GNP is easy to form a connected network structure in PCM and has good dispersion, which helps to improve the thermal conductivity of the composite. The direction of high thermal conductivity of GNPs is along the layer extension direction. This means that heat can be transferred quickly on GNPs, which greatly improves thermal conductivity. But in fact, although their theoretical thermal conductivity is very high, the thermal conductivity measured after filling with paraffin/CF20 (uniform) is not as high as expected. One of the reasons is the difference in particle form. In order to illustrate the effect of particle form on thermal conductivity, MD (molecular dynamics) simulations are performed.

First, a paraffin molecular model is established, as shown in Figure 2. The melting point of paraffin is 58-60°C. Therefore, C<sub>27</sub>H<sub>56</sub>, an alkane with a carbon number of 27, is created as a simulation molecule in the material studio, with a space volume of 35 Å \* 5 Å \* 5 Å.

Then, Packmol software was used to create a box of appropriate size to fill the molecules. For paraffin molecules, the appropriate box is a 5 nm cube unit cell (both the length, the width, and the height are greater than the original molecules). When creating a mixture system, the nanoparticles should be first created in a certain area in this cubic space; the experiments done this time are rectangular coordinate systems, and the nanoparticles are placed in the center area of the entire cubic unit cell.

After the crystal cell box is determined, the density of C<sub>27</sub>H<sub>56</sub> is 0.88 g/cm<sup>3</sup>, so the number of paraffin molecules in the 5 nm cube of the solid is calculated:

$$N = \frac{880 \times (5 \times 10^{-9})^3}{1.66 \times 10^{-27} \times 380} \approx 174. \quad (1)$$

In this paper, the hot wire method is used to measure the thermal conductivity when the test piece is solid, and when PCM melts into liquid, the flow of liquid will greatly improve the thermal conductivity, which is beyond the scope of micro heat transfer simulation. Therefore, this paper uses the molecular dynamics of solid paraffin to simulate.

Building nanoparticle models was continued, as shown in Figure 3.

TABLE 1: Selected materials and material data.

Material	The average particle size (nm)	Purity	Particle shape	Loose density ( $\text{g}\cdot\text{cm}^{-3}$ )	Density ( $\text{g}\cdot\text{cm}^{-3}$ )
Nano-SiO <sub>2</sub>	15	99.5%	Spherical	0.05	2.2
CNTs	15	99.5%	Chain	0.05	2.2
GNPs	5 $\mu\text{m}$	$\geq 99.5\%$	Two-dimensional flake	$0.065 \pm 0.020$	2.0

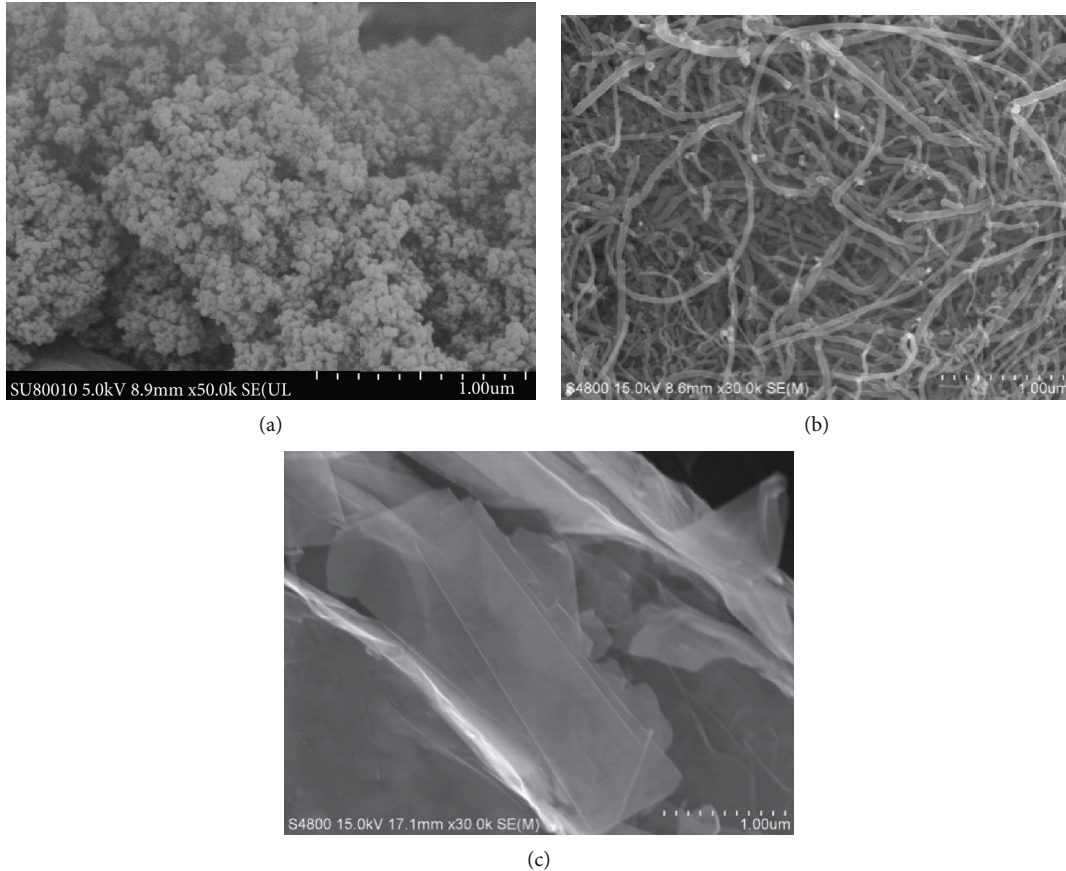
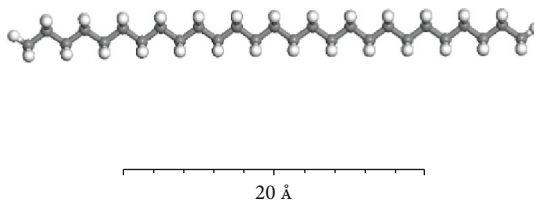
FIGURE 1: (a, b, and c) are electron micrographs of nano-SiO<sub>2</sub>, CNTs, and GNPs, respectively.

FIGURE 2: Paraffin molecular model.

To build a 5 nm cube, it needs to extend in the  $X$ -direction again. CNTs or GNP molecular crystal nanoparticles have a length of more than 30 Å. In order to avoid molecules exceeding the box size in subsequent experiments, the simulation box size must be expanded to 150 Å \* 50 Å \* 50 Å.

The mixture model is obtained after extension, as shown in Figure 4.

The two sides of the simulated box are, respectively, equipped with a constant temperature heat source and con-

stant temperature cold source. The heat of particles simulated by this nonequilibrium state will be transferred along the  $X$ -direction, generating an irreversible heat flow. To calculate such directional heat flow, use the definition of thermal conductivity of Fourier [17, 18]:

$$k = -\frac{1}{V_c} \frac{Q(\omega)}{\nabla T} = -\frac{Jx}{\partial T/\partial x} = \frac{\phi A}{(T_h - T_1)/Lx}, \quad (2)$$

where  $J$  is the heat flow density in the  $X$ -direction,  $T$  is the temperature,  $Lx$  is the thickness,  $\phi$  is the heat flow, and  $A$  is the cross-sectional area perpendicular to the heat flow.

The thermal conductivity of the previously created box is simulated in Lammmps with an integration step of 0.5 timesteps and a total time of 2000000 timesteps. The simulation temperature of the PCM is 300 K (solid state) at room temperature. After 500000 timesteps, the calculated thermal conductivity tended to be stable, which proved that the relaxation



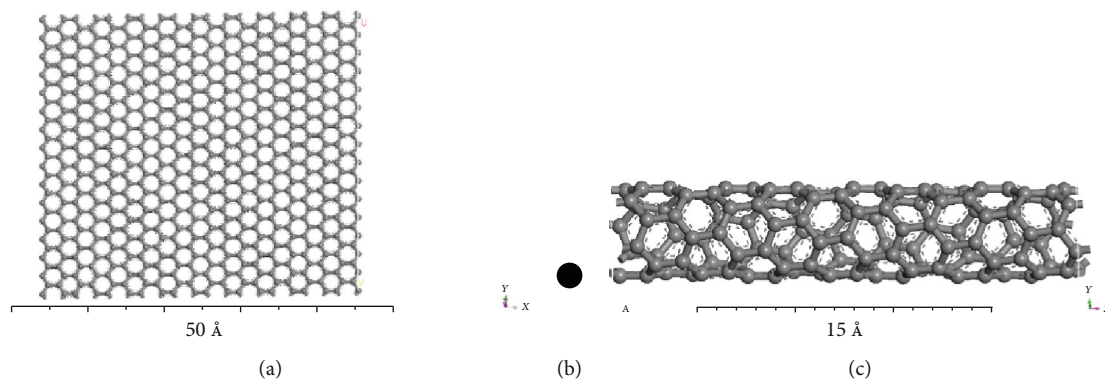


FIGURE 3: (a, b, and c) are simulation diagrams of GNP molecules, SiO<sub>2</sub> molecules, and CNT molecules, respectively.

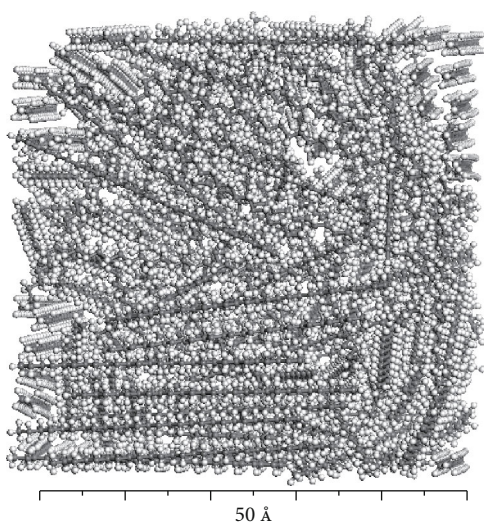


FIGURE 4: Mixture model.

equilibrium particle energy method with 500000 timesteps is feasible. According to the calculation of 2000000 timesteps, the thermal conductivity of paraffin is 0.1306 W/(m·K), and that of CNTs and GNPs is 0.2733 W/(m·K) and 0.3425 W/(m·K) at 5 wt.%, respectively.

A hot area is set on the left side of the simulation box, and a cold area is set on the right side. The temperature distribution will not converge if the temperature difference is too low. In this simulation, the temperature difference between the left and right sides is 85 K, the left side is 190.8 K, and the right side is 115.5 K. Paraffin can maintain solid shape at this temperature, and the system temperature tends to be stable after 2000000 timesteps.

The results of thermal conductivity obtain by applying a heat source at one end are simulated in Figure 5.

The simulated paraffin-GNP thermal conductivity is 1.5 times the thermal conductivity of paraffin-CNTs and 2.5 times the thermal conductivity of paraffin-SiO<sub>2</sub> at 5 wt.%, indicating that the specific particle form affects the thermal conductivity of paraffin-nanoparticles.

On the other hand, the filling of nanoparticles also affects the phase change enthalpy of PCMs during solidification and

melting. The traditional nucleation theory [19, 20] is based on the Gibbs theory which describes the formation of new phases in a homogeneous single phase. This theory has been successful in explaining many experimental phenomena.

Critical nucleus radius  $r^* = 2\sigma M/\rho RT \ln(c/c_0)$ , and nucleation energy  $\Delta G^* = 16\pi\sigma^3 M^2/3R^2 T^2 \rho^2 (\ln c/c_0)$ . It shows that different particles have different molar masses  $M$ , different saturation concentrations  $c_0$ , and different densities. The required nucleation energies are also different. Relatively, the enthalpy of the composite PCM during solidification or melting will be changed. Here, we compare the enthalpy values of SiO<sub>2</sub> and GNP fill composites with the same mass fraction to illustrate the effect of different particle forms on the performance of composite phase change energy storage materials.

In the DSC curve, the upper curve is the material melting process, and the lower curve is the material solidification process. The “pure” in the icon represents pure PCM, and the percentage represents the composite phase change material containing the nanoparticles of the mass fraction. The melting enthalpy of pure paraffin is 207.2 J/g, and the enthalpy of solidification is 206.0 J/g. Figures 6(a) and 6(b) show that the melting enthalpy and solidification enthalpy of the GNP-filled composites are the smallest, respectively, 176.2 J/g and 178.9 J/g.

Figures 6(c) and 6(d) show the DSC curves of the paraffin-nanoparticle-GNP samples and the composite PCM still maintain good melt-solidification properties. With the addition of CF20, the phase change latent heat of the sample is significantly reduced, and with the increase of nanoparticle content, the phase change latent heat further decreases. Without nanoparticles, the melting enthalpy of the paraffin-CF20 samples is 155.6 J/g, and the freezing enthalpy is 153.1 J/g. After adding nanoparticles, the melting enthalpy and solidification enthalpy of the composites filled with GNPs are the lowest, which are 146.3 J/g and 145.9 J/g, respectively.

The results show that when paraffin is filled into CF20, the enthalpy of the composite energy storage material is reduced by about 50 J/g, because CF20 does not participate in energy storage. Similarly, after filling nano-SiO<sub>2</sub> and GNPs, the enthalpy of the composite material will also be reduced, but when the mass fraction is 5%, the enthalpies of paraffin-GNPs-CF20 are lower than the two; the values

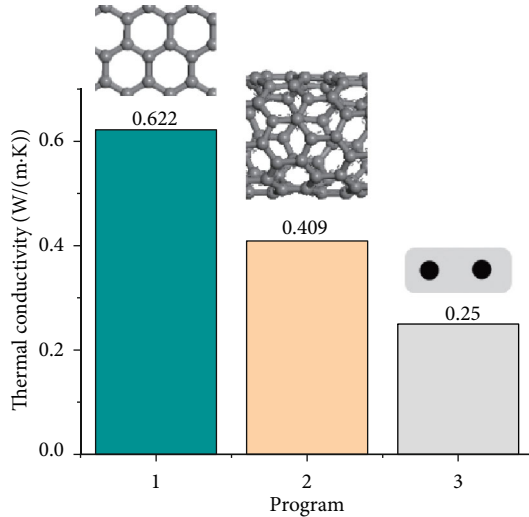


FIGURE 5: Simulation scheme of nanoparticles and their thermal conductivity simulation results. The thermal conductivity is simulated by the Lammmps program. Programs 1, 2, and 3 stand for four small GNP molecules and one long nanotube, and the homogeneous distribution of SiO<sub>2</sub> particles after mixing with paraffin is simulated. This is the simulation result of their thermal conductivity at 5 wt.%.

are 10 J/g. The paraffin-GNPs-CF20 enthalpy decreases even more. This is because when the composite PCM is solidified, the nanoparticles will crystallize and need to absorb energy; when the composite PCM is melted, the nanoparticle crystals disperse and also need to absorb energy. Because the two participate in the process of solidification or melting of PCM, the required nucleation energy is different, and GNPs need more energy to nucleate paraffin.

**3.2. Effect of Dispersion Modes on Thermal Conductivity of Carbon-Based Composites.** The dispersion of nanoparticles in modified PCM/CF20 under the same mass fraction is mainly reflected in the uneven distribution of particles and the distribution of different particles. In order to explain the effect of the dispersion mode on the thermal conductivity of paraffin-nanoparticle-CF20, we chose the “island-network” thermal conductivity composite model and the Maxwell model [21, 22] to predict the thermal conductivity of the paraffin-nanoparticle-CF20 composite. The “island-network” model equation formula is as follows:

$$\frac{1}{\lambda} = \frac{V}{\lambda_f} + \frac{1-V}{\lambda_p}. \quad (3)$$

In the formula,  $\lambda_p$  is the thermal conductivity of polymers,  $\lambda_f$  is the thermal conductivity of filler,  $\lambda$  is the thermal conductivity of composite materials, and  $V$  is the volume fraction of the particle. For low-filling thermally conductive polymer-based composites, it has no interaction between the fillers and they are distributed in the matrix in the form of “islands,” which is the “island” model.

The Maxwell model formula is as follows:

$$\lambda = \lambda_1 \frac{\lambda_2 + 2\lambda_1 + 2V(\lambda_2 - \lambda_1)}{\lambda_2 + 2\lambda_1 - V(\lambda_2 - \lambda_1)}. \quad (4)$$

In the formula,  $\lambda_1$  is the thermal conductivity of polymers,  $\lambda_2$  is the thermal conductivity of filler,  $\lambda$  is the thermal conductivity of composite materials, and  $V$  is the volume fraction of the particle. The Maxwell-Eucken model characterizes the thermal conductivity of composites formed by particles that have no interaction with each other and are uniformly dispersed in the matrix. This model is suitable for predicting the thermal conductivity of composites with filler contents below 30 vol.%. The volume of the composite energy storage material is measured to be 50 cm<sup>3</sup>, the density of paraffin is 0.88 g·cm<sup>-3</sup>, the density of foamed carbon is taken to be 2.2 g·cm<sup>-3</sup>, and the density of the nanoparticle material is shown in Table 1. The experimental data and prediction data obtained are as follows.

It is clearly seen in Figures 7(a) and 8(a) that the two models are more accurate for predicting the thermal conductivity of nano-SiO<sub>2</sub> composites. However, the predicted values of paraffin-SiO<sub>2</sub>-CF20 and paraffin-CNTs-CF20 in Figure 7(b) do not match, and both are higher than the experimental values. The experimental values of paraffin-GNPs-CF20 are higher. This is because the “island-network” model (tandem model) considers that it has no interaction between the fillers and that the distribution is uniform, and it is basically used to predict the lower limit of the thermal conductivity of the composite. Because of the presence of paraffin and CF20 in the composite, the overall structure is more complicated. From the model formula, it shows that with the increase of  $\lambda_f$  and the decrease of  $V/\lambda_f$ , the predicted value of the thermal conductivity of the composite material will be more biased to the calculation result of  $1-V/\lambda_p$ , so the predicted increase of the thermal conductivity of paraffin-GNPs-CF20 has no actual increase in thermal conductivity of modified GNPs.

In Figure 8(b), the thermal conductivity of composites predicted by the Maxwell-Eucken model is obviously higher than that of the experimental value. This is because the Maxwell-Eucken model is applicable to the thermal conductivity model without uniform distribution of homogeneous spheres in the homogeneous matrix, but in practice, the distribution of nanoparticles is not uniform, and the interaction between nanoparticles and CF20 also exists. For example, when CNTs are filled in paraffin and then immersed in CF20, the CNTs with their irregularly distributed cloth will entangle themselves, as shown in Figure 8(b), which greatly reduces the prediction effect of the Maxwell-Eucken model.

The CF20 structure increases the thermal conductivity of paraffin wax from 0.25 W/(m·K) to 8.3083 W/(m·K), an increase of 33.2 times. The thermal conductivity of paraffin-SiO<sub>2</sub>-CF20 in Figure 7(b) decreases by 0.02% from 0 wt.% to 5 wt.% and then increases with the increase of mass fraction. This is because when SiO<sub>2</sub> nanoparticles are just added, a small number of particles will increase the contact thermal resistance, and the gap between the particles

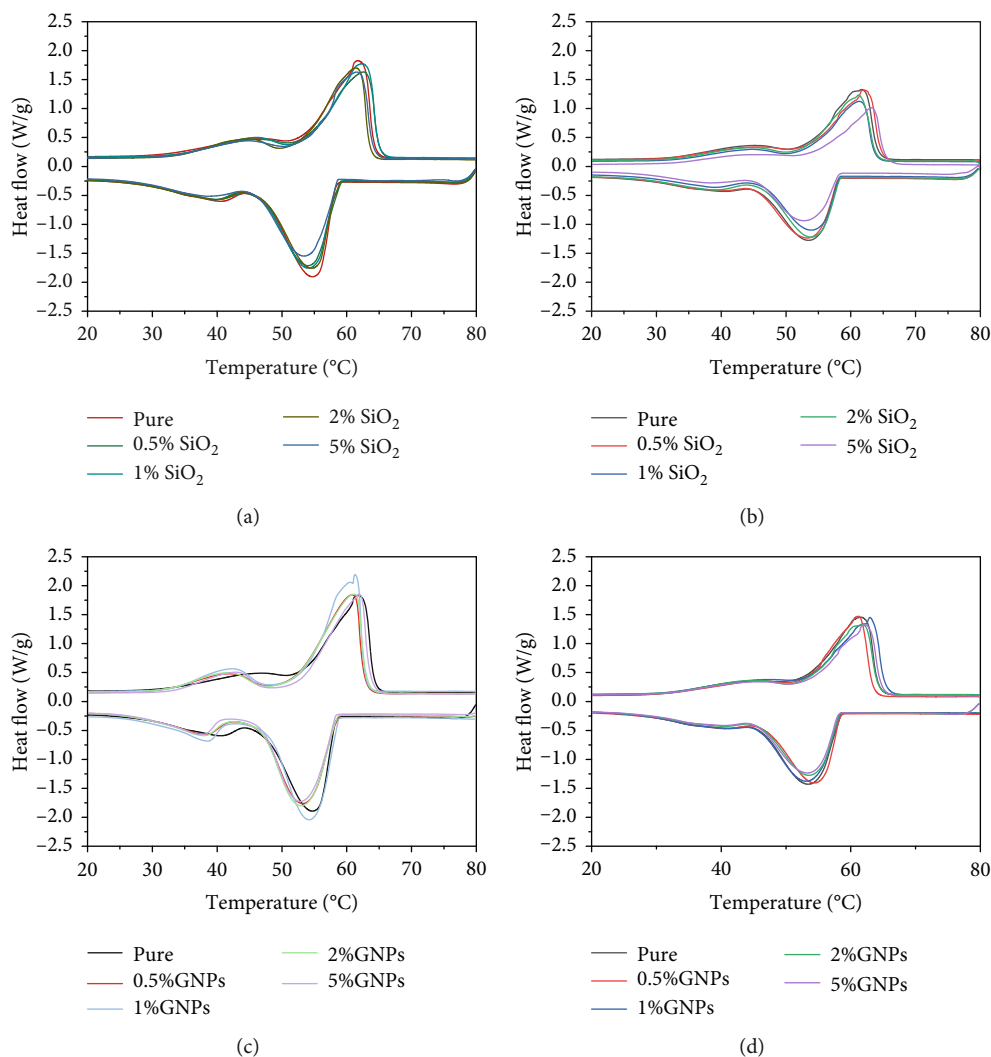


FIGURE 6: (a, b, c, and d) are DSC curves of paraffin-SiO<sub>2</sub>, paraffin-SiO<sub>2</sub>-CF20, paraffin-GNPs, and paraffin-GNPs-CF20, respectively.

becomes large, which does not achieve the effect of increasing the thermal conductivity. As the mass fraction increases, the gap becomes smaller, the contact thermal resistance decreases, and the thermal conductivity starts to increase, but the thermal conductivity of the final composite material does not increase significantly due to the low thermal conductivity of the nano-SiO<sub>2</sub> particles. With the increase of CNTs from 0 wt.% to 2 wt.%, the thermal conductivity increases by 0.0142 W/(m·K), but when the content is increased from 2 wt.% to 5 wt.%, the thermal conductivity only increases by 0.0107 W/(m·K). The theoretical thermal conductivity of GNPs should be very high, but the actual percentage increase in thermal conductivity is only 4.71% on the premise that Figure 7(b) changes from 0 wt.% to 5 wt.%.

#### 4. Conclusion

Based on the composite energy storage material paraffin-CF20, different nanoparticles are filled in to obtain three new composite materials. The experimental data and the

simulation data of the two models are compared to obtain conclusions that under the same mass fraction of nanoparticles, different particle forms and dispersion modes have different effects on the properties of the composite.

**4.1. Particle Form.** With simulation MD, the simulated paraffin-GNP thermal conductivity is 1.5 times the thermal conductivity of paraffin-CNTs and 2.5 times the thermal conductivity of paraffin-SiO<sub>2</sub> at 5 wt.%, indicating that the specific particle form affects the thermal conductivity of paraffin-nanoparticles. At the same time, the nucleation theory is used to explain the effect of particle form on the enthalpy of composite phase change energy storage materials. When the mass fraction is 5%, the enthalpies of paraffin-GNPs-CF20 are lower than the other two; the values are 10 J/g.

**4.2. Diffusion Modes.** The different distribution modes lead to uneven distribution and different distribution conditions, and their effects on thermal conductivity are also different. Filling paraffin with foamed carbon increases the thermal

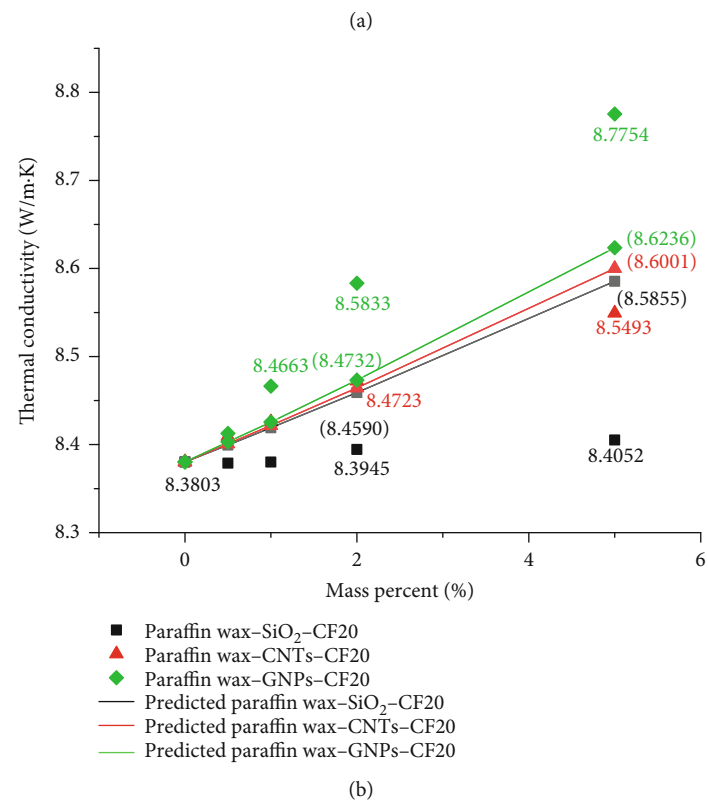
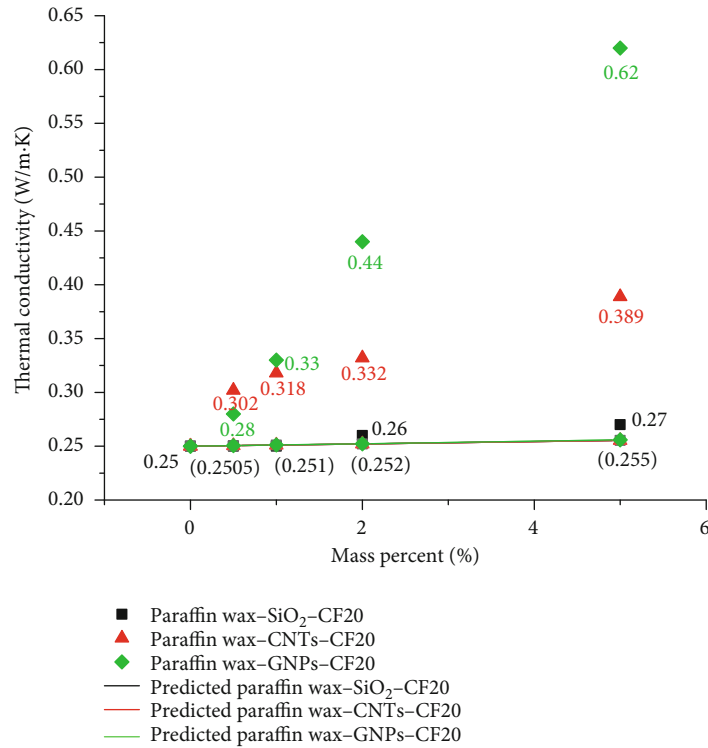
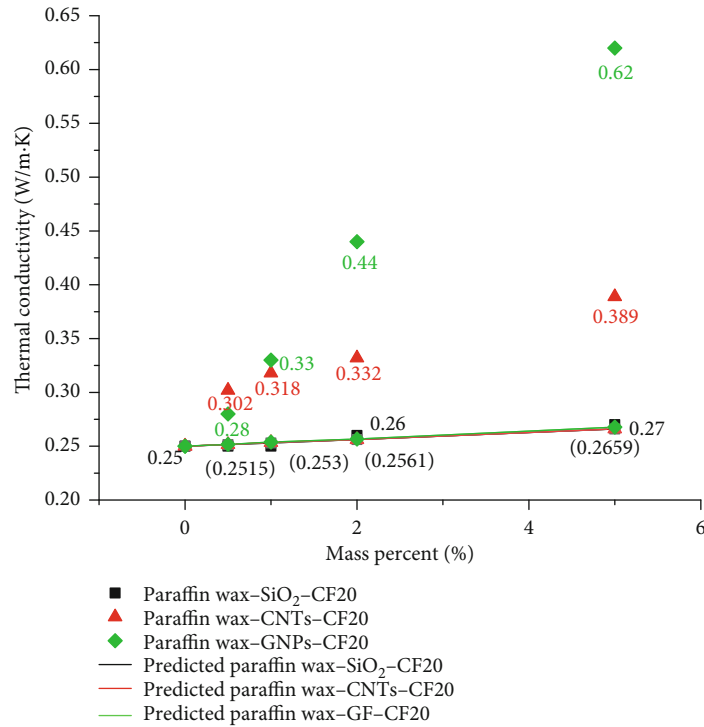


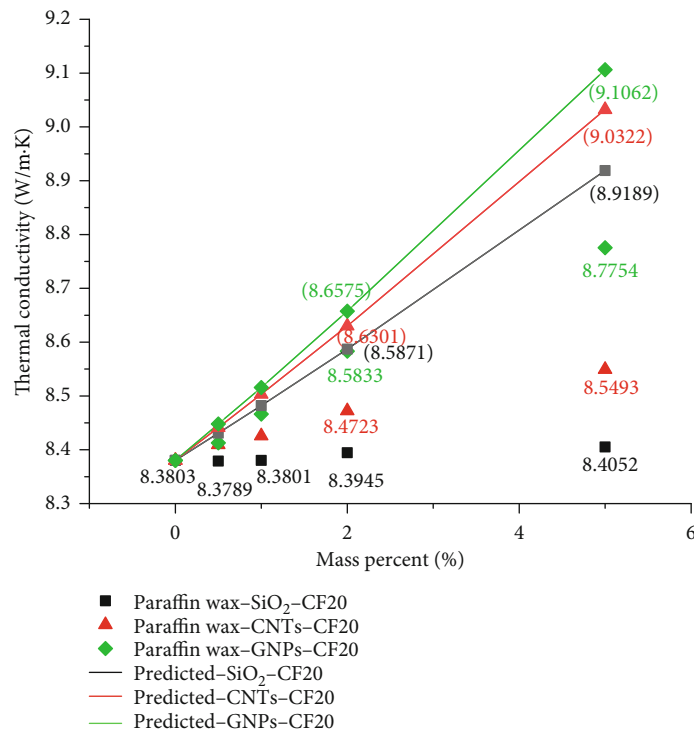
FIGURE 7: (a and b) are comparison diagrams of predicted values and experimental values of the thermal conductivity of the “island-network” model. The material in (a) is three kinds of modified paraffin, and the material in (b) is three kinds of modified paraffin-CF<sub>2</sub>O.

conductivity of pure paraffin from 0.25 W/(m·K) to 8.3083 W/(m·K), an increase of 33.2 times. The thermal conductivity of paraffin-SiO<sub>2</sub>-CF<sub>2</sub>O decreases by 0.02% from 0 wt.% to 5 wt.% and then increases with the increase of mass

fraction. Under 5 wt.%, compared with the thermal conductivity enhancement effect of SiO<sub>2</sub> composites, the thermal conductivity increase effects of CNTs and GNP composites are 6.7 and 15.8 times the thermal conductivity increase of



(a)



(b)

FIGURE 8: (a and b) are the comparison diagrams of the thermal conductivity predictions and experimental values of the Maxwell model; the materials in (a) are three modified paraffin waxes, and the materials in (b) are three modified paraffin-CF<sub>2</sub>O.

SiO<sub>2</sub> composites, respectively. The experimental data of thermal conductivity of SiO<sub>2</sub>, CNTs, and GNP composites are different from the data obtained from the “island” model and Maxwell model theory. GNP composites have the high-

est thermal conductivity and are higher than the theoretical thermal conductivity of the two models, indicating that the different ways of dispersing nanoparticles have a significant effect on thermal conductivity.



## Data Availability

All data used to support the findings of this study are included within the article.

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

## Authors' Contributions

Hui Li put forward the idea that different dispersion modes of nanoparticles will affect the thermal conductivity of the PCM composite. Jia Yu proposed that different forms of nanoparticles would also affect thermal conductivity. Hui Li and Qingshan Zhu collected relevant literature. Hui Li and Li Kong contacted and obtained SEMs of these nanoparticles. Hui Li, Li Kong, and Hongji Zhu designed the PCM with SiO<sub>2</sub>, CNTs, and GNPs and measured the thermal conductivity experiments. The experiment was carried out by Hui Li, Haoqing Wang, and Qingshan Zhu. Hui Li and Haoqing Wang verified the experimental data. Hui Li collated the experimental data and analyzed them to get the conclusion of the experiment. Finally, Hui Li wrote this paper. Jia Yu reviewed the paper and agreed to submit it.

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