Study of Phase-Transition Characteristics of New Composite Phase Change Materials of Capric Acid–Palmitic Acid/Expanded Graphite

Hua Fei,* Wenqing Du, Qian He, Qingjun Gu, and Linya Wang

ABSTRACT: A new composite phase change material of capric acid–palmitic acid/expanded graphite (CA–PA/EG) with the optimum mass ratio of EG equated to 8:1 was prepared by the physical adsorption method. It was observed that the eutectic point of CA–PA binary system was reached at 22.1 °C, and CA–PA was uniformly distributed into the pores of EG by physical interaction. The melting and freezing temperatures of CA–PA/EG obtained by differential scanning calorimeter (DSC) were 23.05 and 20.82 °C, respectively, while the corresponding latent heats were 139.7 and 131.8 J/g, respectively. It had good thermal and chemical stability, and there was almost no leakage of liquid binary phase change materials after 1000 melting–freezing cycles. According to the experimental results of the thermogravimetry (TG) analyzer as well as heat storage and release, CA–PA/EG has excellent thermal reliability and heat resistance and the high thermal conductivity of EG promotes the thermal energy storage and release rate of CA–PA. Thus, CA–PA/EG is suitable as a phase change energy storage material for building energy conservation.

1. INTRODUCTION

With the rapid development of the economy and the continuous improvement of people’s living standards, the global energy demand is also increasing, resulting in the rapid consumption of fossil fuel resources.1–10 It is necessary to find an effective way to reduce the dependence on fossil fuels and meet the increasing global energy demand. In addition, it has become an essential research topic in the field of energy to improve energy efficiency to achieve the purpose of energy saving.11–14 Phase change material (PCM) is an efficient energy storage material that can not only reduce the peak of power consumption and transfer part of the load from the maximum demand period but also improve the energy efficiency and alleviate energy crisis.15–18 As a typical latent heat energy storage material, it has received extensive attention from researchers in recent years.19–24

Fatty acids have shown many superior properties, such as high heat capacity, melting consistency, nontoxicity, good thermal stability, nonflammability, small volume change during phase transition, and so on. However, some studies have found that the phase change materials of fatty acids have some defects, such as inappropriate temperature, low thermal conductivity, and easy leakage of liquid phase for solid–liquid phase transition,19–22 which affect its application scope, development prospects, energy storage, and release efficiency.23–26 Therefore, researchers have carried out relevant studies and proposed corresponding solutions.27–30 If the phase-transition temperature and the thermal conductivity are not appropriate, it can be considered to set a suitable multielement system and optimize material.27 A binary eutectic mixture was formed by the compound of octanoic acid (OA) and myristic acid (MA).28 The results show that the phase-transition temperature of OA–MA is 7.13 °C, which is about 9 and 45 °C lower than that of OA and MA, respectively. On the other hand, the researchers found that expanded graphite (EG) has a unique network pore structure, large specific surface area, high surface activity, and strong adsorption capacity. The heat transfer performances of diatomaceous earth, organobentonites, calcium ores, and expanded graphite of high thermal conductivity media added into the phase change materials were analyzed.29 The results show that EG has a better heat transfer effect. A composite phase change material of d-mannitol/expanded graphite was obtained. The d-mannitol/expanded graphite has a maximum sorption capacity. D-Mannitol is evenly dispersed in the micropore of EG. Therefore, expanded graphite can not only increase the thermal conductivity of phase change materials.
but also restrain the liquid phase leakage of phase change materials.\textsuperscript{30–33}

The phase change energy storage materials obtained in this work are suitable for the field of building energy conservation, capric acid–palmitic acid (CA–PA) binary eutectic mixture for phase-transition temperature with 21.78 °C is used as phase change materials, and expanded graphite is used as the matrix, the preparation of a new type of CA–PA/EG composite phase change materials was obtained in this work. The structure and properties of CA–PA/EG were analyzed by scanning electron microscopy (SEM), Fourier transform infrared (FT-IR), differential scanning calorimetry (DSC), and thermogravimetry (TG), which provided a guide for the application of CA–PA/EG in the field of building energy conservation.

2. RESULTS AND DISCUSSION

2.1. Determination of the Ratio of CA–PA Binary Eutectic Mixture. To determine the proportion of CA–PA binary eutectic mixtures with different mass ratios were prepared with the mass fractions of CA of 0, 20, 40, 60, 80, 85, 86, 87, 90, and 100%. According to the step-cooling curve and the $T$–$X$ phase diagram of the binary system, the low eutectic point of CA–PA can be determined, and the mass ratio corresponding to this point is the best ratio of low eutectic.

Figure 1 shows the cooling curves of different components of the CA–PA binary mixture, and Figure 2 shows the $T$–$X$ phase diagram of the CA–PA binary mixture. It can be found that the eutectic point of the CA–PA binary system can be reached at 22.1 °C when the mass fraction of CA is 86%.

![Figure 1](https://example.com/figure1.png)

Figure 1. Cooling curves of different components of the CA–PA binary mixture.

![Figure 2](https://example.com/figure2.png)

Figure 2. $T$–$X$ phase diagram of the CA–PA binary mixture.

Figure 3 shows the DSC curves of a binary low melting mixture of CA–PA and CA–PA/EG composite phase transition material. As can be seen from Figure 3, the binary low melting mixture of CA–PA has only one heat absorption peak and exothermic peak, indicating that CA and PA have good eutectic properties for the mass fraction of CA of 86%. The melting temperature ($T_{m}$) and solidification temperature ($T_{h}$) of CA–PA are 21.78 and 17.51 °C, respectively. It is consistent with the results from the step-cooling curve, and its phase-transition temperature is much lower than those of CA and PA, which is in accordance with the requirements of the field of building energy saving. The melting latent heat ($H_{m}$) and the solidification latent heat ($H_{h}$) are 154.7 and 148.0 J/g, respectively. Therefore, the CA–PA binary eutectic mixture is suitable for developing energy conservation.

2.2. Determination of the Optimum Ratio of CA–PA/EG. A series of different proportions of CA–PA/EG composite phase change materials were prepared, and the proportions of CA–PA and EG are 7:1, 8:1, 9:1, 11:1, 13:1, 15:1, 17:1, and 19:1, respectively. The samples of 0.200 g of CA–PA were weighed on a filter paper, placed in a drying oven at 65 °C for 1.5 h, then taken out, and cooled to room temperature. The experimental data are shown in Table 1 and Figure 4. It can be seen that they have an obvious seepage circle in M3–M4 and the mass-loss rate exceeds 1.5%. This indicates that the content of CA–PA exceeds the adsorption capacity of EG, and CA–PA leaks out during solid–liquid phase transition, but not the exudation ring for M1–M2. The mass-loss rates of M1 and M2 changed little during heat treatment, both of which are 1%.

![Figure 3](https://example.com/figure3.png)

Figure 3. DSC curves of CA–PA and CA–PA/EG.

| sample | CA–PA:EG | before heat treatment (g) | after heat treatment (g) | mass change (g) | mass loss (%) |
|--------|----------|---------------------------|--------------------------|----------------|--------------|
| M1     | 7:1      | 0.200                     | 0.198                    | 0.002          | 1.0          |
| M2     | 8:1      | 0.200                     | 0.198                    | 0.002          | 1.0          |
| M3     | 9:1      | 0.200                     | 0.197                    | 0.003          | 1.5          |
| M4     | 11:1     | 0.200                     | 0.196                    | 0.004          | 2.0          |
| M5     | 13:1     | 0.200                     | 0.195                    | 0.005          | 2.5          |
| M6     | 15:1     | 0.200                     | 0.194                    | 0.006          | 3.0          |
| M7     | 17:1     | 0.200                     | 0.192                    | 0.008          | 4.0          |
| M8     | 19:1     | 0.200                     | 0.187                    | 0.013          | 6.5          |
However, the latent heat of phase change of CA−PA/EG composite phase change material increases with the increase of CA−PA content. Therefore, the optimal mass ratio of CA−PA to EG should be 8:1.

2.3. Microscopic Morphology and Structural Properties of CA−PA/EG. The micromorphology of EG and CA−PA/EG composite phase change materials is shown in Figure 5. As can be seen from Figure 5a, EG has a honeycomb pore structure, with the interior composed of graphite flakes and a large number of irregular pore canals, which can provide enough space for CA−PA and inhibit the leakage of liquid CA−PA. It can be seen from Figure 5b that the pores of EG are filled with CA−PA. Therefore, the adsorption capacity of EG reaches a certain degree for the CA−PA-to-EG mass ratio of 8:1, and the CA−PA/EG composite phase change material does not have liquid phase leakage. As can be seen from Figure 5c, the original structure of the CA−PA/EG composite phase change material remains unchanged after 1000 accelerated cooling and heating cycles. There is almost no leakage of liquid CA−PA, which has a good working performance.

Figure 6 shows the FT-IR diagrams of CA−PA, EG, and CA−PA/EG composite phase change materials. As can be seen from Figure 6, the FT-IR curves of EG show the bending vibration peaks of H−O−H, the asymmetric stretching vibration peaks of −CH2, and the bending vibration peak of water adsorbed at 3415, 2917, and 1625 cm−1, which might be because EG absorbs a small amount of water. The characteristic absorption peaks of CA−PA at 3418 cm−1 are generated by the stretching vibration of −OH and the asymmetric stretching vibration absorption peaks and the symmetric stretching vibration absorption peaks of −CH2 occur at 2928 and 2858 cm−1, respectively. The characteristic absorption peaks at 1710 cm−1 are caused by the stretching vibration of C=O and the asymmetric bending vibration of −CH2 causes
the characteristic peak at 1458 cm\(^{-1}\). The characteristic absorption peaks of 1287 and 938 cm\(^{-1}\) are the stretching vibration peak of C–O and the bending vibration peak –OH, respectively. The characteristic absorption peaks at 723 cm\(^{-1}\) are caused by the out-of-plane bending vibration of C–H. The FT-IR curves of CA–PA/EG composite phase change materials also show characteristic absorption peaks at 3433, 2925, 2858, 1710, 1459, 1288, 936, and 725 cm\(^{-1}\). A comparison of the FT-IR curves of CA–PA and CA–PA/EG composite phase change materials shows that there are characteristic absorption peaks of CA–PA and EG in the curves of CA–PA/EG, and the new characteristic absorption peaks do not appear, indicating that the CA–PA/EG composite phase change materials cannot produce new substances and the chemical reaction does not occur between CA–PA and EG, which rely on the surface tension and capillary force.

2.4. Thermal Properties of CA–PA/EG. Figure 3 shows the DSC curves of CA–PA and CA–PA/EG composite phase change materials. As can be seen from Figure 3, the melting temperature (\(T_m\)) and the melting enthalpy (\(H_m\)) of the CA–PA/EG are 23.05 °C and 139.7 J/g, respectively, and the freezing temperature (\(T_f\)) and the freezing enthalpy (\(H_f\)) are 20.82 °C and 131.8 J/g, respectively. By comparing the DSC curves of CA–PA, it can be seen that the melting and freezing temperatures of CA–PA/EG increased by 1.27 and 3.31 °C compared with those of CA–PA. This may be because CA–PA overcomes the force produced by EG voids during melting and freezing. In addition, the phase change enthalpy of CA–PA/EG is less than that of CA–PA, which is due to the fact that phase transition does not occur in EG, and the phase-transition enthalpies of CA–PA/EG composite phase change materials are proportional to the mass fraction of CA–PA. The theoretical phase change enthalpies of CA–PA/EG can be calculated according to formula 1. The melting enthalpies and the solidification enthalpies are 137.5 and 131.5 J/g, respectively. Compared with the actual phase change enthalpies of CA–PA/EG, it can be found that the actual phase change enthalpies of CA–PA/EG are slightly larger than the theoretical phase change enthalpies of CA–PA/EG, which is because EG strengthens the phase change characteristics of CA–PA. Compared with the composite phase change materials in Table 2, CA–PA/EG composite phase change materials have similar thermal properties to those of other composite phase change materials, but the phase change enthalpies of CA–PA/EG are higher. Therefore, CA–PA/EG is a potential phase change energy storage material, which could be suitable for building energy saving, air conditioning condensation heat recovery, low-temperature solar thermal storage, and other fields.

\[
H_n = \eta H_m
\]

where \(H_n\) is the theoretical phase-transition enthalpy of CA–PA/EG, \(\eta\) is the mass fraction of CA–PA, and \(H_m\) is the phase-transition enthalpy of the CA–PA binary eutectic mixture.

2.5. Thermal Cycling Stability of CA–PA/EG. The thermal cycling stability of composite phase change materials affects service life. In this paper, the experiments of 1000 accelerated cold and hot cycles were carried out on CA–PA/EG composite phase change materials, and the stability of thermal and chemical properties of CA–PA/EG after the cycles was measured. The experimental results are shown in Figures 7 and 8. It can be seen from Figure 7 that the melting

![Figure 7. DSC curves of CA–PA/EG before and after thermal cycles.](https://dx.doi.org/10.1021/acsomega.0c03665)

and solidification temperatures of CA–PA/EG composite phase change materials after accelerated cooling and heating for 1000 times are 22.95 and 20.74 °C, respectively, and the latent heats of melting and solidification are 141.4 and 132.8 J/g, respectively. Compared with the phase transformation

![Figure 8. FT-IR spectra of CA–PA/EG before and after thermal cycles.](https://dx.doi.org/10.1021/acsomega.0c03665)

Table 2. Thermal Properties of Some Other Composites in the Literature

| composite phase change materials | melting temperature (°C) | solidification temperature (°C) | enthalpy (J/g) | reference |
|----------------------------------|--------------------------|---------------------------------|----------------|-----------|
| lauric acid–myristic acid/expanded graphite | 32.70 | 151.7 | 34 |
| decanoic acid–stearic acid/waste gas filling block | 26.76 | 57.84 | 35 |
| stearic acid–acetamide/expanded graphite | 66.94 | 58.02 | 36 |
| decanoic acid/kaolin | 30.71 | 28.21 | 37 |
| decanoic acid/bentonite | 29.31 | 21.89 | 38 |
| capric acid–palmitic acid/expanded graphite | 23.05 | 20.82 | 39 |

Table 2. Thermal Properties of Some Other Composites in the Literature

| composite phase change materials | melting temperature (°C) | solidification temperature (°C) | enthalpy (J/g) | reference |
|----------------------------------|--------------------------|---------------------------------|----------------|-----------|
| palmitic acid/expanded graphite | 23.05 | 20.82 | 39 |
| palmitic acid/lauric acid | 28.25 | 21.89 | 39 |
| palmitic acid/acetamide | 30.71 | 28.21 | 39 |
| lauric acid/expanded graphite | 32.70 | 151.7 | 39 |
| lauric acid/lauric acid | 32.70 | 151.7 | 39 |
| lauric acid/acetamide | 30.71 | 28.21 | 39 |
| stearic acid/lauric acid | 30.71 | 28.21 | 39 |
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| lauric acid/expanded graphite | 32.70 | 151.7 | 39 |
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| lauric acid/acetamide | 30.71 | 28.21 | 39 |
| stearic acid/lauric acid | 30.71 | 28.21 | 39 |
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| lauric acid/lauric acid | 32.70 | 151.7 | 39 |
| lauric acid/acetamide | 30.71 | 28.21 | 39 |
| stearic acid/lauric acid | 30.71 | 28.21 | 39 |
| stearic acid/acetamide | 30.71 | 28.21 | 39 |
| stearic acid/expanded graphite | 23.05 | 20.82 | 39 |
properties of CA–PA/EG before and after cold–heat cycles, it can be found that the melting and solidification temperatures of CA–PA/EG after cold–heat cycles are reduced by 0.1 and 0.08 °C respectively, and the latent heat of melting and solidification are increased by 1.2 and 0.7%, respectively, which may be caused by the test error of DSC. However, the changes in phase change temperature and latent heat are very small, indicating that the composite phase change materials of CA–PA/EG have good thermal stability.

As can be seen from Figure 8, the FT-IR curves of the CA–PA/EG composite phase change material do not show change in the peak shape and produce new characteristic absorption peaks for 1000 accelerated cooling and heating cycles. Therefore, CA–PA/EG has good chemical stability. Based on the research results of CA–PA/EG composite phase change materials before and after cooling and heating cycles, it can be seen that CA–PA/EG has good thermal stability and chemical stability after 1000 accelerated cooling and heating cycles. It has application and popularization value in building energy conservation and other fields.

2.6. Heat Resistance and Thermal Reliability of CA–PA/EG. The TG curves of EG and CA–PA/EG are shown in Figure 9. As can be seen from Figure 9, the mass loss is almost zero when EG rises from room temperature to 400 °C, indicating that EG has superior heat resistance. CA–PA/EG shows a weightlessness peak in the whole heating process, which is due to the evaporation of CA–PA, and the weight loss rate reaches 89% from 118 to 222 °C, which is consistent with the results of DSC. In addition, it can be found that the mass loss of CA–PA/EG almost does not occur below 80 °C, and the temperature reaches 142 °C when the weight loss is 5%; it shows that the composite phase change material of CA–PA/EG has good heat resistance at the working temperature (<80 °C).

The mass 1 g of CA–PA/EG composite phase change materials on the filter paper was put into the drying box at 80 °C for 48 h, then removed, and cooled to room temperature. The quality of the composite phase change material before and after heat treatment was compared. The results show that there is not obvious exudation ring on the filter paper, and the mass of CA–PA/EG after heat treatment is 0.993 g, which is consistent with that of the original sample. Therefore, CA–PA/EG has good thermal reliability at working temperature.

2.7. Heat Storage and Heat Release Properties of CA–PA/EG. The heat storage and heat release properties are important properties of composite phase change materials. Figure 10 shows the relationship between temperature and time of CA–PA binary eutectic mixture and CA–PA/EG composite phase change materials in heat storage and release processes. As can be seen from Figure 10, the temperature and time curves of CA–PA and CA–PA/EG show the three stages of rapid rise and fall, melting or solidification, and rapid change until they are consistent with the ambient temperature. In the heat storage process, it takes 1590 s for CA–PA to rise from 5 to 40 °C and CA–PA/EG only needs 880 s to reach the same temperature range, which is 45% less than that for CA–PA. CA–PA needs 330 s for the solid–liquid phase transition, while CA–PA/EG only needs 110 s, which is 67% shorter than that for CA–PA. CA–PA and CA–PA/EG reduced from 40 to 5 °C in 1600 and 770 s in an exothermic process, respectively, and the time required by CA–PA/EG is 52% shorter than that required for CA–PA. The time required by CA–PA and CA–PA/EG for the liquid–solid phase transition is 550 and 160 s, respectively, and the time consumed by CA–PA/EG is reduced by 71%. Therefore, the storage and heat release rates of the CA–PA/EG composite phase change materials are much higher than that of the CA–PA binary eutectic mixture, which has excellent storage and heat release performance, indicating that the EG with a high thermal conductivity improves the storage and heat release rates of CA–PA.

3. CONCLUSIONS

The physical adsorption method is adopted to the CA–PA binary eutectic phase change materials, EG is used as a carrier material, the CA–PA/EG composite phase change material with optimum mass ratio was prepared. The melting and solidification temperatures are 23.05 and 20.82 °C respectively, and the latent heat of melting and solidification are 139.7 and 131.8 J/g, respectively. The results show that the binary eutectic mixture of CA–PA depending on the surface tension and capillary force is uniformly distributed in the porous structure of EG. After 1000 accelerated cooling and heating cycles, CA–PA/EG composite phase change materials still maintain a good heat storage performance, and there is no leakage of liquid CA–PA mixture. EG can significantly improve the thermal conductivity of CA–PA eutectic mixtures and promote heat storage and release rate of CA–PA. The best mass ratio of the CA–PA/EG composite phase change material has good thermal stability, chemical stability, thermal

![Figure 9. TG curves of EG and CA–PA/EG.](image-url)

![Figure 10. Temperature–time curves of CA–PA and CA–PA/EG in heat storage and release processes.](image-url)
reliability, and heat resistance. It is a potential and promising phase change energy storage material in building energy conservation and other fields.

4. MATERIALS AND METHODS

4.1. Materials. Capric acid (CA, C₉H₁₈O₂, 172.27, 98.5%, Chemical Pure) and palmitic acid (PA, C₁₆H₃₁O₂, 256.43, Analytical Reagent) were supplied by Changzhou Haituo Experimental Instrument Co., Ltd. Expandable graphite (mesh 50, expansion ratio: 380 mL/g, carbon content: 98%) was purchased from Qingdao Risheng Graphite Co., Ltd. The chemicals were used as received without further purification.

4.2. Preparation of CA–PA/EG Composite Phase Change Materials. A certain amount of CA and PA were weighed and put into a beaker. The beaker sealed with a film was heated in a drying oven. After the fatty acids in the beaker had completely melted, the beaker was taken out and the contents stirred for 0.5 h to ensure that CA and PA were evenly mixed. Then, the mixture was cooled to room temperature for obtaining a two-component eutectic mixture of CA–PA. In addition, according to the microwave method, a certain amount of expandable graphite was weighed and put into a beaker, and the beaker sealed with a film was put into a drying oven at 80 °C for 24 h. Then, a certain mass of dried expandable graphite was weighed in an evaporation dish, and then the evaporating dish was placed in a microwave oven so that EG was prepared by expanding at 700 W for 40 s. EG was placed in different beakers with the same mass, and the CA–PA binary eutectic mixture weighed according to different mass ratios was added to EG respectively. After being evenly stirred by a glass rod, the beaker sealed with a film was heated in a drying oven for 24 h and stirred every 8 h to ensure that the CA–PA binary eutectic mixture was evenly adsorbed into the pore of EG. After cooling to room temperature, the CA–PA/EG composite phase change material was obtained.

4.3. Characterization of CA–PA/EG Composite Phase Change Materials. The phase change temperatures and latent heat of CA–PA and CA–PA/EG composite PCM were analyzed by DSC (TAQ200, TA Instruments) under nitrogen atmosphere at a flow rate of 50 mL/min. The temperature range was 10–80 °C, and the heating/cooling rate was set at 5 °C/min. The morphology and microstructure of EG and CA–PA/EG composite PCMs were observed by SEM (evo18, ZEISS, Germany). The FT-IR spectra of EG, CA–PA, and CA–PA/EG composite PCMs were obtained with a resolution of 4 cm⁻¹ and the resolution of 4 cm⁻¹. The thermal stability of the CA–PA/EG composite PCM was investigated by the TG using a thermal analyzer (STA6000, TA Instruments) in the temperature range from 20 to 400 °C at the heating rate of 10 °C/min in a nitrogen environment.

It is a common analytical method to test the thermal properties of materials using the step-cooling curve method. First, the test tubes containing the samples with different mass ratios were placed in a water bath at 80 °C. When the samples were completely melted, they were quickly removed and placed in a constant temperature and humidity incubator. As the temperature of the samples in the test tube reached 60 °C, the constant temperature and humidity incubator was immediately started at the target temperature of 10 °C. Then, the step-cooling curve could be drawn according to the data of the temperature change with time in the cooling process, and the crystallization temperature could be obtained by it. In addition, to determine the low eutectic point of CA–PA, the T–X phase diagram of the binary system could be drawn according to the crystallization temperature obtained from the step-cooling curve.

The thermal performances of the CA–PA and CA–PA/EG composite PCM during heat storage and release process were investigated to evaluate the effects of EG on the heat transfer behavior. One tube filled with 15 g of CA–PA binary eutectic mixture and the other filled with 15 g of CA–PA/EG composite phase change material were placed in a constant temperature and humidity incubator at 5 °C. When the temperature of the thermocouple was constant, it was quickly put into a water bath pot at 40 °C for melting experiments; after the sample temperature became constant, it was then quickly put in a constant temperature and humidity incubator of 5 °C for freezing experiments. The temperature changes of the samples in the whole storage and heat release period were recorded by the temperature patrol instrument.

The thermal stability of the CA–PA/EG composites was characterized by the 1000 accelerated melting–freezing cycle test. First, the test tubes filled with CA–PA/EG composite phase change materials were placed in a low-temperature constant temperature and humidity incubator. After the thermal resistance test temperature became constant, it was quickly put into a normal temperature water bath for the melting experiment. After the sample temperature did not change, it was quickly placed in a low-temperature constant temperature and humidity incubator for freezing experiment. The temperature changes of the samples in the whole storage and release cycle were recorded by the temperature inspection instrument. At the same time, the test tube containing the CA–PA/EG sample was placed in a high-temperature water bath. After the sample was completely melted, the test tube was taken out and put into a low-temperature, constant temperature and humidity incubator until the sample in the test tube was completely solidified; then, it was put into the high-temperature water bath. A total of 1000 cycles were carried out, and the thermal properties of the samples were tested after the end of each cycle.

AUTHOR INFORMATION

Corresponding Author
Hua Fei — The Laboratory of Architectural Environment and Energy Application Engineering, Jiangxi University of Science and Technology, Ganzhou 341000, Jiangxi, China; orcid.org/0000-0003-1800-8900; Phone: 86-797-8161560; Email: feihua0928@163.com

Authors
Wenqing Du — The Laboratory of Architectural Environment and Energy Application Engineering, Jiangxi University of Science and Technology, Ganzhou 341000, Jiangxi, China
Qian He — The Laboratory of Architectural Environment and Energy Application Engineering, Jiangxi University of Science and Technology, Ganzhou 341000, Jiangxi, China
Qingjun Gu — The Laboratory of Architectural Environment and Energy Application Engineering, Jiangxi University of Science and Technology, Ganzhou 341000, Jiangxi, China
Linya Wang — The Laboratory of Architectural Environment and Energy Application Engineering, Jiangxi University of Science and Technology, Ganzhou 341000, Jiangxi, China

Complete contact information is available at:
Notes
The authors declare no competing financial interest.

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