

## Article

# Shape-Stabilized Stearic Acid/Expanded Graphite/Chitin-Derived Carbon Phase Change Materials for Enhanced Thermal Storage Performance and Photothermal Conversion

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## Abstract

Melting leakage and low thermal conductivity of stearic acid (SA) restrict its application in thermal storage. In this work, a shape-stabilized phase change material (ECNX/SA) with enhanced thermal storage performance and photothermal conversion is designed based on expanded graphite/chitin-derived carbon (ECNX). Thermal storage performance, including phase change temperature, enthalpy, thermal conductivity and shape stability, of ECNX/SA is investigated. With this, the influence mechanism of ECNX on the thermal storage performance is characterized via N<sub>2</sub> isothermal adsorption–desorption, FTIR, XRD and SEM. Results show that the prepared ECN15/SA has ideal thermal storage performance, where its phase change enthalpy and thermal conductivity are 121.59 J/g and 1.573 W/(m·K), respectively, and possesses superior shape stability. Moreover, the thermal storage performance of ECN15/SA keeps stable even undergoing several thermal cycles, and its photothermal conversion is as high as 89.2%. Characterizations suggest that ECN15 with a hierarchical pore structure and a high graphitization degree to enhance the shape stability and thermal conductivity of SA. Therefore, the prepared ECN15/SA is potential using in thermal storage.



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**Keywords:** shape stabilized phase change material; thermal storage; stearic acid; photothermal conversion

## 1. Introduction

Energy transition and renewable energy promote the development of energy storage technology. Thermal storage technology has received much attention in recent years owing to the desirable application and high storage capacity. It can be classified into thermochemical storage, sensible thermal storage and latent thermal storage [1,2]. In comparison to other thermal storage technologies, latent thermal storage, using phase change materials (PCMs) as a medium, has advantages including high thermal storage density, constant phase change temperature and a simple process [3,4]. Thus, latent thermal storage is potentially employed in the system of solar energy, waste thermal recovery and thermal management. In a myriad of PCMs, organic PCM has been widely used in latent thermal storage by its lower supercooling degree, non-toxic and small thermal expansion [5,6]. However, the application potential of organic PCM is limited by the problems of melting leakage and low thermal conductivity.

An efficient method to overcome the abovementioned drawbacks is encapsulating organic PCM by porous carbon with high thermal conductivity to synthesize shape stabilized

PCM (SSPCM) [7]. Specifically, the melted PCM could be adsorbed into the pore structure through interactions (capillary force, surface tension and hydrogen bond) to prevent leakage on the one hand; the thermal transfer of PCM is improved by porous materials on the other hand [8–10]. Nguyen et al. [11] encapsulated glutaric acid into expanded graphite to obtain SSPCM (GA/EG), where GA/EG contains 90 wt.% glutaric acid without leakage during thermal storage, and the phase change enthalpy of GA/EG is as high as 167 J/g. Li et al. [12] employed carbon nanotube adsorbing paraffin to prepare CNTs/Paraffin, its thermal conductivity is 0.65 W/(m·K), which is 179% higher than pure paraffin. Despite these achievements, the high costs and unsustainability of porous carbon are unfavorable for the large field application of SSPCM.

Biomass possesses the characteristics of wide resources and eco-friendly, its derived carbon exhibits porosity and abundant functional groups that can induce interactions with organic PCM [13,14]. These features make biomass-derived carbon suitable as support for the preparation of SSPCM. For instance, Baniasadi et al. [15] prepared SSPCM using biochar, cellulose and polyethylene glycol (PEG). The biochar–cellulose possesses an efficient thermal transfer channel and developed pore structure to improve the thermal storage performance of PEG, where prepared SSPCM displays the melting enthalpy of 130 J/g and thermal conductivity of 0.41 W/(m·K). More importantly, the photothermal conversion of PEG is significantly improved by biochar, corresponding an efficiency of SSPCM as high as 85%. Among the biomass, chitin ( $\beta$ -(1,4)-2-acetylamino-2-deoxy-D-glucose), widely distributing in crustaceans, fungus and microorganisms, shows the characteristics of low price and abundant N content [16]. These advantages promote its potential using as the support for PCM. In our previous study [17], chitin was carbonized to obtain in situ N-doped carbon for encapsulating stearic acid (SA), where N-doped carbon can induce a hydrogen bond with SA that enhanced the shape stability of SSPCM. Due to the outstanding ability for the capture photon of in situ N-doped carbon, the photothermal conversion efficiency of SSPCM is high as 74.12%. It is concluded that SSPCM based on chitin-derived carbon is anticipated for the practical application of thermal storage, but it exhibits low graphitization degree and numerous micropore structures, making a poor thermal conductivity and PCM capacity in SSPCM. Therefore, it is essential to propose an efficient strategy to enhance the encapsulating capacity of chitin-derived carbon.

Expanded graphite (EG) is a typical support for PCM owing to its low cost, high porosity and thermal conductivity [18]. Nevertheless, the macropore structure and monotonous functional group of EG make it difficult to produce the capillary force and interaction to PCM. An ideal answer for this question is integrating EG and biomass-derived carbon. In this study, a novel support (ECNX) for PCM is proposed by adding EG into chitin-derived carbon. On the one hand, the high porosity and thermal conductivity in EG is potentially compensating for a low adsorbing capacity and thermal transfer of chitin-derived carbon. On the other hand, the developed micropore structure and N-doped surface in chitin-derived carbon could enhance the interaction with the PCM of EG. ECNX serves as the support of stearic acid (SA) to prepare an SSPCM (ECNX/SA) with enhanced thermal storage performance. The evolution of physicochemical properties in ECNX with EG addition is revealed, and the influence mechanism of ECNX on the thermal storage performance is investigated. Also, the photothermal conversion, thermal stability and thermal reliability of ECNX/SA is revealed further. This is expected to provide an efficient way for preparing an SSPCM with low cost and high performance.

## 2. Experimental

### 2.1. Materials

Stearic acid (SA) was sourced from Shanghai Macklin Biochemical Technology Co., Ltd., China. Chitin was acquired from Zhejiang Golden Shell Pharmaceutical Co., Ltd., China. Expanded graphite (EG, with size 75  $\mu\text{m}$ ) was obtained from Qingdao Cologne Carbon Material Co., Ltd., China.

### 2.2. Preparation of Support (ECNX)

Initially, chitin was mixed with the EG of designed mass fraction (0 wt.%, 5 wt.%, 10 wt.%, 15 wt.%, 20 wt.% and 25 wt.%) in deionized water and treated by ultrasonic vibration for 1 h until homogeneous. After that, the suspension was dried at 80  $^{\circ}\text{C}$  to prepare the precursor of support (EG/Chitin). Based on the recent literature, EG/Chitin underwent carbonization in a tube furnace under  $\text{N}_2$  atmosphere, where the carbonization temperature was designed from 20  $^{\circ}\text{C}$  to 800  $^{\circ}\text{C}$  with a heating rate of 5  $^{\circ}\text{C}/\text{min}$ . Then, it was cooled to room temperature naturally, resulting in the support of PCM, named ECNX, where X represents the mass fraction of EG.

### 2.3. Synthesis of SSPCM (ECNX/SA)

Considering that the shape stability of SSPCM could be destructed by the excessive SA, the mass fraction of SA encapsulating in the SSPCM is defined as 60 wt.% based on the basic experiments. Briefly, 3 g SA were heated at 90  $^{\circ}\text{C}$  until melting, and 2 g ECNX were immersed in the melted SA. The obtained mixture underwent ultrasonic treatment at 90  $^{\circ}\text{C}$  for 15 min, and was subsequently cooled to room temperature. To guarantee the homogeneity between SA and ECNX, the abovementioned process was repeated twice. Finally, the mixture was ground into powder and melted in 90  $^{\circ}\text{C}$  in a vacuum oven, following vacuum impregnation for 6 h to synthesize SSPCM, labeled as ECNX/SA.

### 2.4. Characterizations

Thermal properties, such as phase change temperatures ( $T_m/T_f$ ), phase change enthalpies ( $\Delta H_m/\Delta H_f$ ) and supercooling degree ( $\Delta T = T_m - T_f$ ), were evaluated using a differential scanning calorimeter of TGA/DSC 3+ (Mettler Toledo Co., Ltd., Switzerland) within a temperature range of 50  $^{\circ}\text{C}$  to 80  $^{\circ}\text{C}$  in  $\text{N}_2$  atmosphere.

Thermal conductivity was detected by thermal constant analyzer of TPS2500 (Hot Disk Co., Ltd., Sweden).

Shape stability was measured via heating test. The ECNX/SA was pressed into a tablet and placed on a filter, then heated at 90  $^{\circ}\text{C}$  for 2 h. The shape stability was evaluated by the shape change and leakage rate ( $\varphi$ ) of tablet after heating, the  $\varphi$  was calculated by Equation (1) [19],

$$\varphi = \frac{m_3 - m_1}{m_2 \cdot \eta} \quad (1)$$

where  $m_1$  and  $m_2$  represent the masses of filter paper and ECNX/SA, g;  $m_3$  denotes the mass of filter paper following the heating test, g;  $\eta$  indicates the mass fraction of SA in ECNX/SA, %.

Thermal stability of samples was assessed using a thermogravimetric analyzer of TGA/DSC 3+ (Mettler Toledo Co., Ltd., Switzerland). Mass loss of ECNX/SA in a  $\text{N}_2$  atmosphere was measured between temperatures of 50  $^{\circ}\text{C}$  and 400  $^{\circ}\text{C}$ , with a heating rate set at 5  $^{\circ}\text{C}/\text{min}$ .

Pore structure of ECNX was conducted by an automatic specific surface area analyzer of BELSORP-max (Microtrac Co., Ltd., Japan) at 77.3 K. Before the measurement, ECNX was degassed at 120  $^{\circ}\text{C}$  for 12 h. The parameters of the pore structure, including specific

surface area ( $S_{BET}$ ), total pore volume ( $V_{total}$ ) and average pore diameter ( $D_a$ ), are calculated based on depicted  $N_2$  isothermal adsorption–desorption using the method of Brunauer–Emmett–Teller (BET). Briefly, the  $S_{BET}$  is calculated by the slope of the adsorption curve in the range of  $P/P_0 < 0.3$ , while  $V_{total}$  is evaluated by total quantity adsorption. Further, the relative quantity adsorption at different pore diameter is estimated by the model of Barrett–Joyner–Halenda (BJH) to obtain the pore diameter distribution.

The surface functional group was detected via Fourier transform infrared spectroscopy of Tensor 27 (Bruker Co., Ltd., Germany) in the range of  $500\text{ cm}^{-1}$  to  $3500\text{ cm}^{-1}$  with a scanning rate of 16 s.

Crystal phase was characterized through the X-ray diffractometer of SmartLab SE (Rigaku Co., Ltd., Japan) in the angle range of  $5^\circ$  to  $50^\circ$  with a scanning rate of  $10^\circ/\text{min}$ .

The micromorphology of the samples was observed by scanning electron microscope of Regulus8100 (Hitachi Co., Ltd., Japan) at 5 kV.

Photothermal conversion was measured by simulated solar irradiation. Specifically, the samples were pressed into a tablet and irradiated under a xenon lamp of CEL-HXF300-T3 (Ceaulight Co., Ltd., China). A thermocouple pyrometer was adhered at the bottom of sample to record the temperature curves. The temperature curves of samples are measured three times to guarantee the accuracy. The photothermal conversion efficiency ( $\omega$ ) of samples based on temperature curves was estimated via Equation (2) [20],

$$\omega = \frac{m\Delta H_{absolute}}{PA(t_s - t_e)} \quad (2)$$

where  $m$  was the mass of ECNX/SA, g;  $\Delta H_{absolute}$  is the phase change enthalpy of ECNX/SA, J/g;  $P$  is the solar power,  $\text{W}/\text{m}^2$ ;  $A$  represents irradiation area,  $\text{m}^2$ ;  $t_s - t_e$  demonstrates the time of phase change stage, s.

### 3. Results and Discussion

#### 3.1. Thermal Storage Performance

DSC curves of SA and ECNX/SA are recorded in Figure 1. Both SA and ECNX/SA exhibit one endothermic/exothermic peak, indicating that thermal storage is predominantly stored by the phase change enthalpy of SA. The parameters of thermal property are calculated based on DSC curves and listed in Table 1. It can be seen that a lower melting temperature ( $66.71^\circ\text{C}$  to  $67.89^\circ\text{C}$ ) and a higher freezing temperature are observed in ECNX/SA compared with SA. This enhancement could be ascribed to the improved thermal transfer and heterogeneous nucleation facilitated by ECNX, which accelerates the phase change process of SA [21,22]. As a result, the supercooling degree of ECNX/SA is markedly lower than that of SA ( $3.52^\circ\text{C}$ ). In detail, as the EG mass fraction rises from 0 wt.% to 25 wt.%, the supercooling degree of ECNX/SA is decreased from  $0.89^\circ\text{C}$  to  $0.46^\circ\text{C}$ . This demonstrates that the addition of EG can efficiently suppress the supercooling characteristic of SA.

Phase change enthalpy is a crucial parameter for estimating thermal storage density. In Table 1, ECNX/SA shows melting enthalpy as high as  $120.30\text{ J/g}$  and freezing enthalpy as high as  $119.33\text{ J/g}$ , which is better than the reported literature [23,24]. However, it is obvious that the phase change enthalpy of ECNX/SA is lower than its theoretical values ( $132.50\text{ J/g}$  and  $134.40\text{ J/g}$ ). This may be due to the micropore structure in ECNX that induces nanoconfinement to prevent the phase change in part of the SA molecules, resulting in the enthalpy loss [25]. Moreover, it is worth noting that the phase change enthalpy of ECNX/SA remains stable as increasing EG mass fraction, proving that EG marginally impacts the phase change behavior of SA.

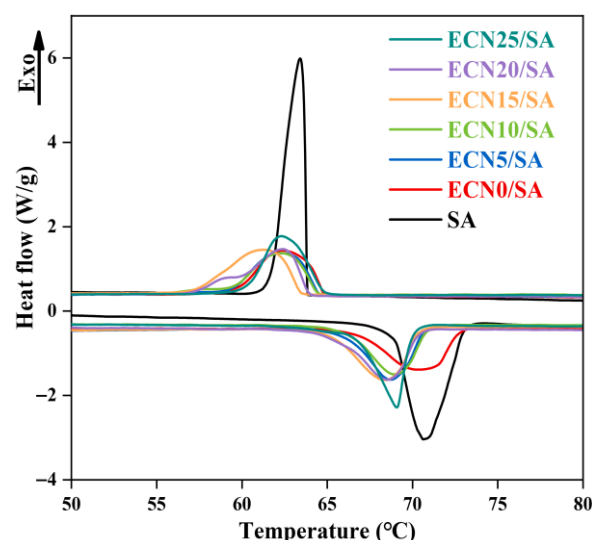


Figure 1. DSC curves of ECNX/SA.

Table 1. Parameters of thermal properties for SA and ECNX/SA.

| Sample   | $T_m$ (°C) | $T_f$ (°C) | $\Delta T$ (°C) | $\Delta H_m$ (J/g) | $\Delta H_f$ (J/g) |
|----------|------------|------------|-----------------|--------------------|--------------------|
| SA       | 68.53      | 65.01      | 3.52            | 220.83             | 224.00             |
| ECN0/SA  | 67.89      | 66.64      | 0.89            | 125.55             | 119.33             |
| ECN5/SA  | 67.27      | 66.64      | 0.63            | 121.45             | 119.59             |
| ECN10/SA | 66.71      | 66.14      | 0.57            | 124.41             | 124.14             |
| ECN15/SA | 67.68      | 67.06      | 0.62            | 121.59             | 119.80             |
| ECN20/SA | 67.63      | 67.16      | 0.47            | 120.30             | 119.95             |
| ECN25/SA | 67.79      | 67.33      | 0.46            | 123.76             | 122.55             |

As illustrated in Figure 2, ECNX can enhance the thermal conductivity of SA (0.180 W/(m·K)) remarkably that the thermal conductivity of ECNX/SA is higher than 0.406 W/(m·K). Moreover, a continuous increase in thermal conductivity from 0.406 W/(m·K) to 1.689 W/(m·K) is observed in ECNX/SA as EG mass fraction increased from 0 wt.% to 25 wt.%, thereby, the addition of EG is profitable for the thermal conductivity. Notably, the thermal conductivity of ECN15/SA is 1.573 W/(m·K), while the value fails to elevate greatly when adding EG further. This may be due to that excessive EG addition provides more cavities in ECNX/SA, which aggravates thermal resistance to hinder the enhancement of thermal conductivity [26].

The images of the heating test in Figure 3 indicate that SA is melted absolutely into a liquid, but ECNX/SA keeps a stable shape after heating. Thus, ECNX can improve the shape stability of SA. In addition, the leakage rate and area are decreased as EG mass fraction increase. From ECN0/SA to ECN10/SA, the leakage rate is decreased from 16.73% to 4.74%, and no leakage is observed in ECN15/SA, ECN20/SA and ECN25/SA.

Consequently, ECN15/SA possesses ideal thermal conductivity (1.573 W/(m·K)), shape stability and phase change enthalpy ( $\Delta H_m$ : 121.59 J/g,  $\Delta H_f$ : 119.95 J/g), it is considered an optimal sample in ECNX/SA. Compared with the reported literature in Table 2, it can be observed ECN15/SA has favorable than other biomass-based SSPCM. For instance, the adsorbing capacity of CSC15/SA based on coconut shell charcoal is merely 35 wt.% with the melting enthalpy of 76.69 J/g and the thermal conductivity of 0.75 W/(m·K), which is much lower than that of ECN15/SA. Further characterizations reveal the mechanism by which ECNX affects the thermal storage performance of SA.

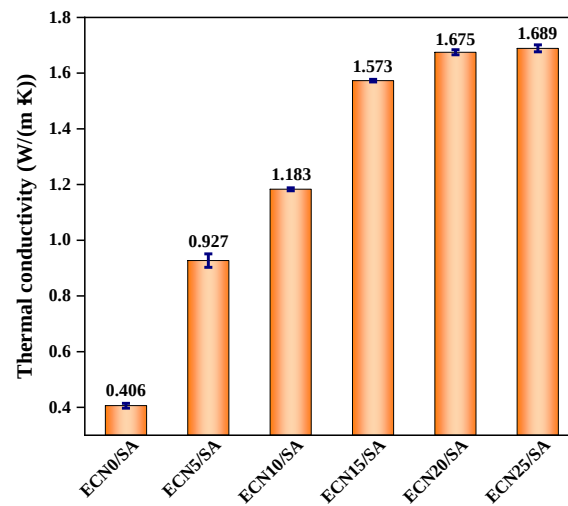


**Figure 2.** Thermal conductivity of ECNX/SA.

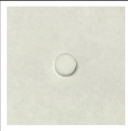
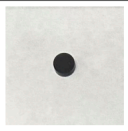
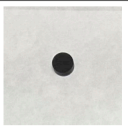
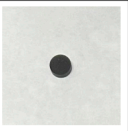
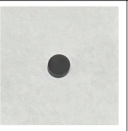
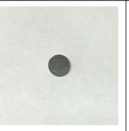
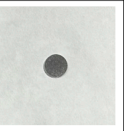
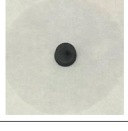
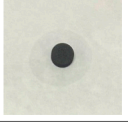
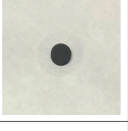
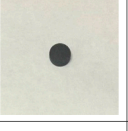
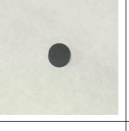
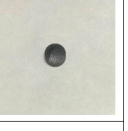
|            | SA                                                                                 | ECN0/SA                                                                            | ECN5/SA                                                                            | ECN10/SA                                                                            | ECN15/SA                                                                             | ECN20/SA                                                                             | ECN25/SA                                                                             |
|------------|------------------------------------------------------------------------------------|------------------------------------------------------------------------------------|------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------|
| 0 h        |   |   |   |   |   |   |   |
| 2 h        |  |  |  |  |  |  |  |
| $\phi$ (%) |                                                                                    | 16.73                                                                              | 9.12                                                                               | 4.74                                                                                | -                                                                                    | -                                                                                    | -                                                                                    |

**Figure 3.** Shape stability of ECNX/SA.

**Table 2.** Thermal properties of SSPCM in the reported literature.

| Sample     | PCM Mass Fraction (wt.%) | Melting Enthalpy (J/g) | Thermal Conductivity (W/(m·K)) | Ref.       |
|------------|--------------------------|------------------------|--------------------------------|------------|
| ECN15/SA   | 60                       | 121.59                 | 1.573                          | This study |
| CSC15/SA   | 35                       | 76.69                  | 0.75                           | [27]       |
| UMSCF16/SA | 75                       | 130.9                  | 1.725                          | [28]       |
| CC1/SA     | 65                       | 127.69                 | 0.261                          | [29]       |
| CC/CA-PA   | 50                       | 71.4                   | 0.74                           | [30]       |

### 3.2. Characterizations Analysis

The N<sub>2</sub> isothermal adsorption–desorption and pore diameter distribution of ECNX is depicted in Figure 4. ECNX exhibits the characteristic of type I-IV isotherms equipped with H3 hysteresis loop (Figure 4a), revealing its micropore and mesopore structure. Furthermore, a certain adsorption is detected at high relative pressure ( $P/P_0 > 0.9$ ) that suggests a macropore structure exists in the ECNX. Therefore, ECNX, with a hierarchical pore structure, can provide capillary force and a large space to encapsulate SA. It is observed that the absorption at high relative pressure is gradually elevated as increased EG mass fraction, indicating that the addition of EG can provide macropore structure for ECNX [31]. The pore diameter distribution (Figure 4b) of ECNX based on the BJH model also confirms this result that the micropores and mesopores are reduced below 10 nm and macropores are increased above 100 nm as the EG mass fraction increasing from 0 wt.% to 25 wt.%.

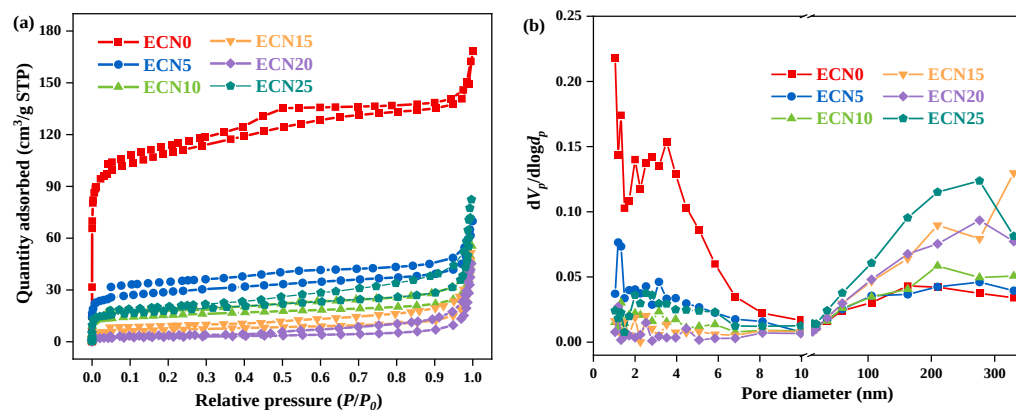


**Figure 4.** N<sub>2</sub> isothermal adsorption–desorption (a) and pore diameter distribution (b) of ECNX.

The pore structure parameters of ECNX are further calculated by the BET method and listed in Table 3. The specific surface area, total pore volume and average pore diameter of ECN0 are 406.16 m<sup>2</sup>/g, 0.233 cm<sup>3</sup>/g and 2.29 nm, respectively. Although a low pore diameter can provide a strong capillary force, it is undesirable for the adequate infiltration of PCM into the pore structure [10,32], so ECN0/SA has serious leakage. With increasing EG mass fraction, the specific surface area of ECNX is decreased from 406.16 m<sup>2</sup>/g to 9.41 m<sup>2</sup>/g and total pore volume is decreased from 0.233 cm<sup>3</sup>/g to 0.052 cm<sup>3</sup>/g, but average pore diameter is expanded from 2.29 nm to 32.54 nm. Although large specific surface area and small pore diameter generate surface tension and capillary force, it induces intensive mass transfer resistance preventing the adequate infiltration of SA into the pore structure [17]. The addition of EG expands the pore diameter of ECNX, which provides large storage space for adsorbing SA [33]. Therefore, ECN15 with the hierarchical structure of micropore, mesopore and macropore provide capillary force, surface tension and storage space for enhancing the shape stability of SA.

**Table 3.** Specific surface area, total pore volume and average pore diameter of ECNX.

| Sample | $S_{\text{BET}}$ (m <sup>2</sup> /g) | $V_{\text{total}}$ (cm <sup>3</sup> /g) | $D_a$ (nm) |
|--------|--------------------------------------|-----------------------------------------|------------|
| ECN0   | 406.16                               | 0.233                                   | 2.29       |
| ECN5   | 90.86                                | 0.083                                   | 4.62       |
| ECN10  | 47.90                                | 0.075                                   | 9.58       |
| ECN15  | 21.04                                | 0.066                                   | 37.56      |
| ECN20  | 9.41                                 | 0.044                                   | 39.03      |
| ECN25  | 19.82                                | 0.052                                   | 32.54      |

The FTIR spectra of ECNX are displayed in Figure 5a, the adsorption bands at 1529 cm<sup>−1</sup> and 1247 cm<sup>−1</sup> represent C=O and C-N, which are derived from the acetamido of chitin [34]. In Figure 5b, SA has the adsorption bands at 2914 cm<sup>−1</sup>, 2844 cm<sup>−1</sup> and 1701 cm<sup>−1</sup> attributing to -CH<sub>3</sub>, -CH<sub>2</sub> and C=O, respectively [35]. The adsorption bands located at 1468 cm<sup>−1</sup> and 1296 cm<sup>−1</sup> are characteristic of -OH. The adsorption bands of SA and ECNX are observed in ECNX/SA without additional bands, proving an ideal chemical compatibility between SA and ECNX. It is noted that the adsorption band of C=O at 1701 cm<sup>−1</sup> in SA is shifted to 1694 cm<sup>−1</sup> in ECNX/SA. This phenomenon is due to the hydrogen bond between SA and ECNX induced by C-N, this is beneficial to suppress the melting leakage of SA in ECNX/SA [36].

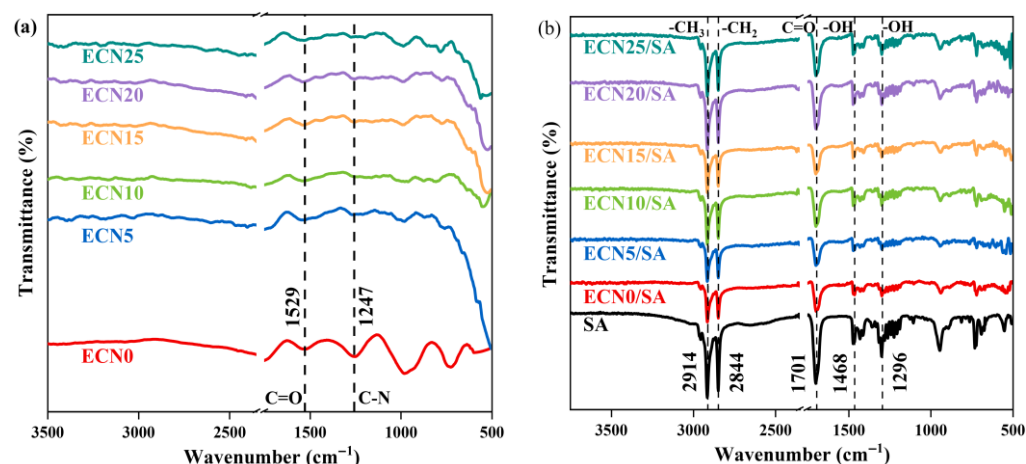


**Figure 5.** FTIR spectra of ECNX (a) and ECNX/SA (b).

Figure 6a depicts the XRD patterns of ECNX, where an intense diffraction peak at  $26.5^\circ$  represents the (002) facet of graphite structure [37]; it is originated from EG with a high graphitization degree. As the EG mass fraction rises, the intensity of (002) facet is increased, in accordance with the continuous enhancement of thermal conductivity of ECNX/SA. This is attributed to the ordered network of graphite structure would reduce phonon scattering thus accelerating thermal transfer [38]. Figure 6b provides the diffraction peaks of SA, in which the peaks at  $6.61^\circ$ ,  $21.62^\circ$  and  $24.31^\circ$  denote the (006), (118) and (200) facet, and their peaks are detected in ECNX/SA. This suggests that only a physical interaction exists in SA and ECNX. Nevertheless, the diffraction peaks of SA are shifted to a low angle in ECNX/SA. For instance, the (118) facet is decreased from  $21.62^\circ$  to  $21.48^\circ$  and the (200) facet from  $24.22^\circ$  to  $24.02^\circ$ . This phenomenon is generated by the hydrogen bond between ECNX and SA that expands the unit cell of SA [39], which is consistent with the FTIR results.

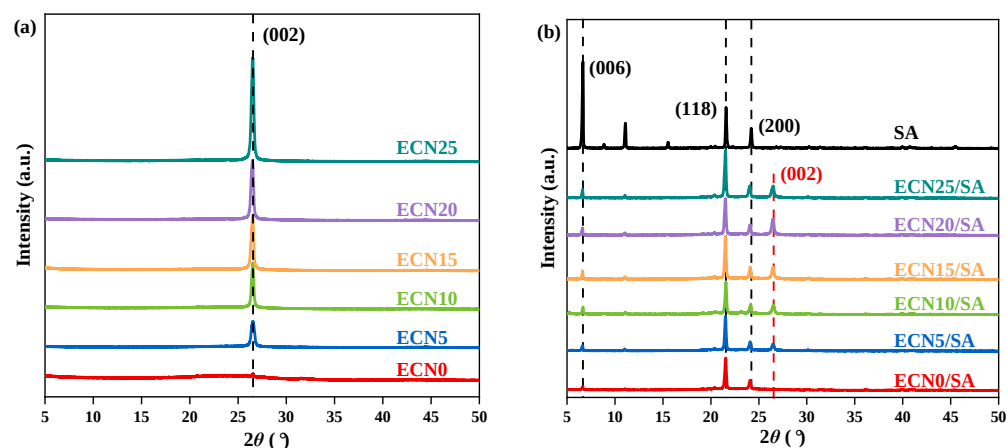


**Figure 6.** XRD patterns of ECNX (a) and ECNX/SA (b).

SEM images are employed to conduct the morphology of samples. In Figure 7a, a few macropores are observed on the surface of ECN0; this is due to the tight arrangement of chitin molecules difficult to generate pore structure during carbonization. In contrast, numerous macropores and worm-like structures are inspected in ECN15 (Figure 7b) owing to the EG addition, which can provide a larger space to encapsulate SA [40]. The images of ECN0/SA and ECN15/SA further illustrate this opinion. In Figure 7c, the surface of ECN0/SA shows free SA particles, where ECN0 is covered by excessive SA. This indicates that ECN0 with poor macropores fails to encapsulate SA completely. While SA is adsorbed

on the surface of ECN15 (Figure 7d), and be infiltrated into the pore structure. Therefore, ECN15 can prevent SA leakage efficiently than ECN0, making a superior shape stability in ECN15/SA.

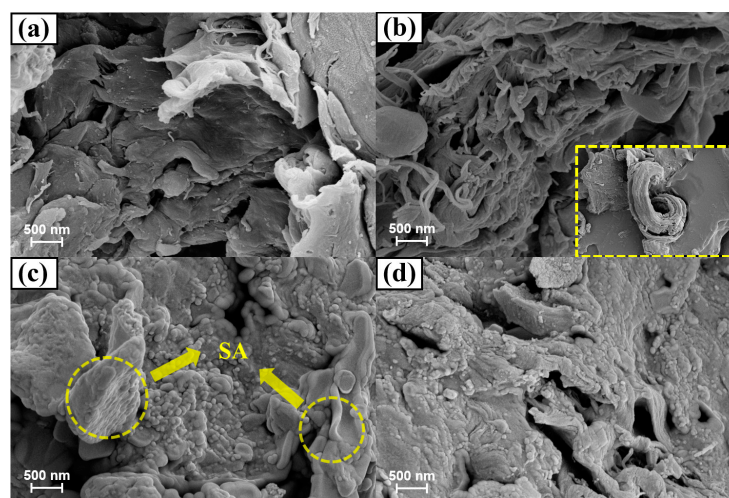


**Figure 7.** SEM images of ECN0 (a), ECN15 (b), ECN0/SA (c) and ECN15/SA (d).

Thermal stability is estimated by TG curves in Figure 8. It can be seen that SA, ECN0/SA and ECN15/SA appear one stage for mass loss (Figure 8a), corresponding to the evaporation of SA. ECN0/SA and ECN15/SA display the mass loss of 59.07% and 60.46% approaching the theoretical SA mass fraction, which demonstrates the accuracy of thermal storage performance. As demonstrated in Figure 8b, the peak temperature of SA in the DTG curve is 292 °C, but the peak of ECN0/SA shifts to a lower temperature of 289 °C. This may be attributed to the efficient thermal transfer provided by ECN0, accelerating the evaporation of SA. By contrast, the peak of ECN15/SA exhibits a higher temperature of 296 °C, revealing that the hierarchical pore structure delays the SA diffusion [41]. In summary, the thermal stability of SA is improved by ECN15, which ECN15/SA suggests ideal thermal stability.

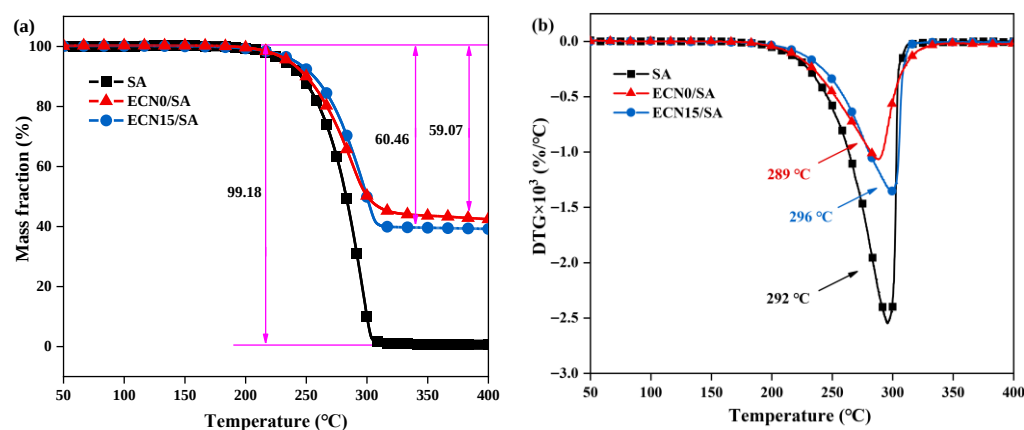


**Figure 8.** TG curves (a) and DTG curves (b) of SA, ECN0/SA and ECN15/SA.

### 3.3. Photothermal Conversion

This study also considers the photothermal conversion of samples in Figure 9. SA display two stages under radiation, where its temperature elevates from 30 °C to 45 °C within 0 s to 250 s, subsequently keeping stable. Obviously, the terminal temperature of SA is below its phase change temperature, reflecting a poor photothermal conversion. For ECN15/SA, it is detected three stages under radiation. In detail, the temperature

of ECN15/SA is elevated from 30 °C to 65 °C in the range of 0 s to 250 s; after that, the temperature arrives at plateau in a period of 250 s to 1530 s corresponding to the phase change process. Finally, the temperature of ECN15/SA climbs from 68 °C to 79 °C within 1530 s to 1750 s. This demonstrates an extraordinary photothermal conversion in ECN15/SA that the photon is captured by ECN15, preserving the phase change enthalpy of SA [42]. ECN0/SA exhibits a similar curve than ECN15/SA, however, ECN0/SA has a longer plateau in the range of 250 s to 1500 s. Therefore, EG addition is favorable for further improving the photothermal conversion of SA. It is ascribed to that high graphite degree can prevent photon scattering [43,44]. The photothermal conversion efficiency of ECN15/SA and ECN0/SA is calculated as 89.2% and 51.4%, reflecting a good photothermal conversion in ECN15/SA.

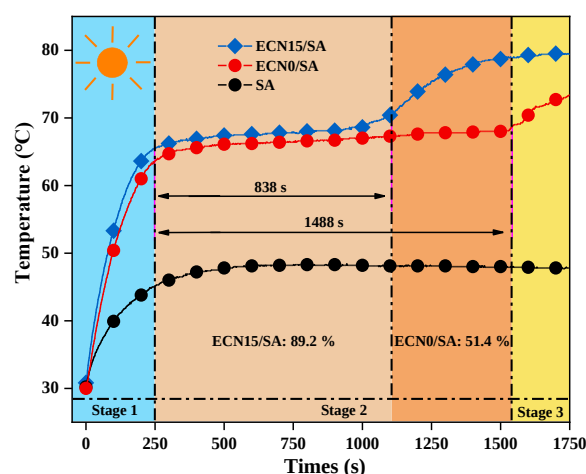


**Figure 9.** Temperature curves of ECNX/SA under simulated solar radiation.

### 3.4. Thermal Reliability

Thermal reliability is a pivotal parameter for the long-term utilization of SSPCM. ECN15/SA is repeated for 200 thermal cycles to determine thermal reliability owing to its optimized thermal storage performance, the ECN15/SA after cycle labeled as ECN15/SA-C. Figure 10a depicts the DSC curves of ECN15/SA and ECN15/SA-C, wherein the melting temperature of ECN15/SA-C is decreased by merely 0.42 °C, and the freezing temperature is decreased by 0.17 °C compared to ECN15/SA. The phase change enthalpy of ECN15/SA is reduced by 1.6% after thermal cycles. It can be concluded that ECN15/SA maintains stable thermal property. Meanwhile, the FTIR spectrum and XRD pattern of ECN15/SA in Figure 10b,c are similar with ECN15/SA, illustrating the unchanged chemical structure and crystal phase during thermal cycles [45]. Consequently, ECN15/SA is a potential medium for long-term thermal storage due to its superior thermal stability.

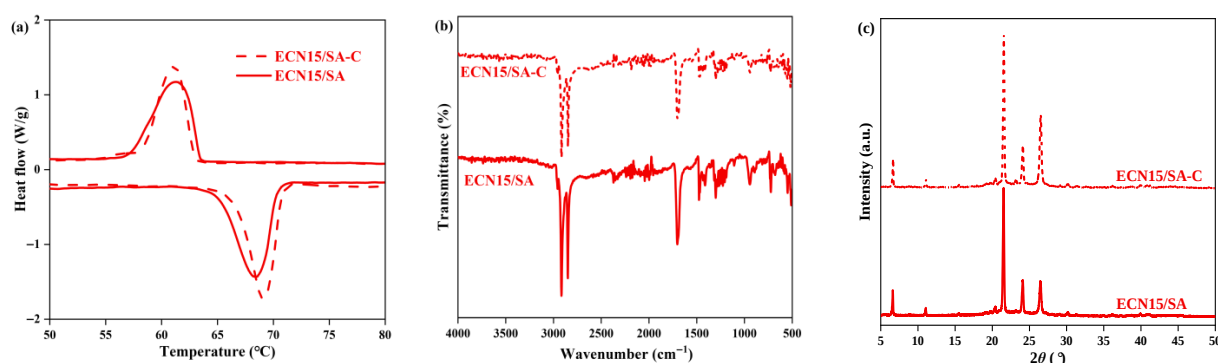


**Figure 10.** DSC curves (a), FTIR spectra (b) and XRD patterns (c) of ECN15/SA and ECN15/SA-C.

## 4. Conclusions

This study integrates EG and chitin-derived carbon to prepare a porous carbon with hierarchical pore structure and high thermal conductivity, it is served as support to adsorb SA for synthesizing SSPCM (ECNX/SA). Prepared ECN15/SA exhibits superior thermal storage performance that the phase change enthalpy and thermal conductivity are 121.59 J/g and 1.573 W/(m·K), respectively. It also has ideal shape stability without leakage at operation temperature. The synthesized ECN15 shows a hierarchical pore structure, providing a large space and capillary force to prevent melting leakage, and possesses a high graphitization degree constructing an efficient thermal transfer channel to elevate the thermal conductivity of SA. Furthermore, it is detected good chemical compatibility between ECN15 and SA that ECN15/SA keep stable thermal storage performance and physicochemical properties even after 200 thermal cycles. ECN15/SA also has enhanced photothermal conversion with an efficiency of 89.2%. Consequently, ECN15/SA is potential using in thermal storage, especially in solar energy conversion and utilization, owing to its high thermal storage density, photothermal conversion efficiency and low cost. In the future, ECN15/SA will be considered for the integration with existing solar energy system to balance its energy fluctuation. The thermal transfer characterization and thermal storage process of ECN15/SA should also be revealed by systemic experiment and numerical simulation.

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