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Lauric Acid Hybridizing Fly Ash Composite for Thermal Energy Storage

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Abstract: Fly ash includes different mineral phases. This paper reported on the preparation of a novel lauric acid (LA)/fly ash (FA) composite by vacuum impregnation as a form-stable phase change material (PCM) for thermal energy, and especially investigated the effect of the hydrochloric acid-treated fly ash (FAh) on the thermal energy storage performance of the composites. The morphology, crystalline structure, and porous textures of the samples were characterized by scanning electron microscopy (SEM), X-ray diffraction (XRD), Brunauer–Emmett–Teller (BET), X-ray fluorescence (XRF), and differential scanning calorimetry (DSC). The results indicated that hydrochloric acid treatment was beneficial to the increase of loading capacity and crystallinity of LA in the LA/FAh composite, which caused an enhanced thermal storage capacity with latent heats for melting and freezing of LA/FAh (80.94 and 77.39 J/g), higher than those of LA/FA (34.09 and 32.97 J/g), respectively. Furthermore, the mechanism of enhanced thermal storage properties was investigated in detail.

Keywords: fly ash; mineral phase; lauric acid; hybridization; thermal energy storage

1. Introduction

The development of energy storage devices is very important all over the world. Thermal energy storage (TES) has been considered as one of the potential key technologies for energy storage devices because TES with phase change materials (PCMs) has attracted interest due to its significant role in solar thermal applications [1–6]. TES includes sensible heat storage, reversible chemical reaction heat storage, and latent heat storage; the latter is regarded as the most effective of the three based on its higher energy-storage density and good isothermal properties [7]. Additionally, solid–liquid PCMs are most suitable for TES [8]. PCMs mainly have organic or inorganic types, and inorganic materials or porous silicate minerals were also applied as supports to stabilize organic PCMs, including gypsum boards [9–11], concrete [12–14], bentonite [15–17], expanded vermiculite [18], clay [19], and porous silica [20–22]. Furthermore, lauric acid has been considered as a latent heat of fusion storage material due to its availability in a wide temperature range. Fly ash (FA) is a by-product of coal combustion in thermal power plants, including complex and abundant anthropogenic matter [23–26]. FA can cause ecological pollution [27]; recently, more efforts have been undertaken to recycle and utilize FA [28,29].

In this paper, FA has been used to form stable LA in an LA/FA composite, and the LA/FA composite was prepared by vacuum impregnation. To increase the maximum loadage of LA of

the composite, FA was subjected to chemical treatment with solution of HCl. It is of interest that the maximum loadage of LA of the LA/FAh composite is larger than the LA/FA composite. The samples were characterized by using X-ray fluorescence (XRF), scanning electron microscopy (SEM) and Brunauer–Emmett–Teller (BET).

2. Materials and Methods

The typical chemical composition of pristine fly ash (FA) from the East Hope power plant, Xinjiang, China is (mass %): SiO₂, 53.63; Al₂O₃, 17.78; CaO, 13.47. Lauric acid (LA, CH₃(CH₂)₁₀COOH) was from Tianjin Kemiou Chemical Reagent Co., Ltd., Tianjin, China. LA/FA composite was synthesized via vacuum impregnation technology. In a typical process, 15.0 g of LA and 10.0 g of FA placed in conical flask were vacuum evacuated to −0.1 MPa for 10 min. The conical flask was then put in a thermostat-controlled water bath at 80 °C for 20 min. The vacuum pump was turned off and air was entered into the conical flask for 5 min. After cooling, the LA/FA composite was thermally filtered at 85 °C for 48 h to remove excess LA. To increase the maximum loadage of LA and improve the thermal storage capacity of the composite, the pristine fly ash was mixed with 2 M solution of HCl and stirred by electric blender for 24 h, and then filtered off and washed with distilled water until pH = 7 [30]. The obtained material (FAh) was dried at 120 °C for 24 h. FAh was then used to prepare LA/FAh composite. The chemical compositions of FA and FAh were analyzed, respectively (Table 1), the result of XRF revealed that the major elements in FA and FAh are Ca, Si, Al, Mg, Fe, Ti, Na and K. To determine the maximum LA content, 5.00 g of LA/FA and LA/FAh composites were heated at 500 °C for 12 h then cooled to room temperature, respectively. The boiling point of LA reaches 299 °C without mass loss of FA or FAh in the composite at 500 °C for 12 h, thus, the LA loadage capacity could be calculated [31].

Table 1. Chemical composition of FA and FAh (wt %).

Samples	SiO ₂	Al ₂ O ₃	CaO	MgO	Fe ₂ O ₃	TiO ₂	Na ₂ O	K ₂ O	Others
FA	53.63	17.78	13.47	2.36	6.08	0.82	1.90	1.09	2.87
FAh	70.07	14.69	2.23	0.81	7.32	0.90	1.79	0.99	1.29

The structure of the samples was analyzed by X-ray diffraction (XRD, DX-2700 diffractometer, Haoyuan, Dandong, China) at a scanning rate of 5°/min with 2θ range from 5° to 80°. Sample morphology was visualized by scanning electron microscopy (SEM, Quanta-200 scanning electron microscope, FEI, Hillsboro, OR, USA) at an acceleration voltage of 5 kV. Differential scanning calorimetry (DSC) analyses were operated using a DSC Q10 instrument at a heating rate of 5 °C/min under a constant stream of argon at atmospheric pressure (accuracies of ±0.10 °C and ±1% for phase change temperature and ΔH capacity, respectively). The sample was cooled in liquid nitrogen during the freezing process. N₂ adsorption–desorption isotherms were measured using an ASAP 2020 unit at 77 K, while the materials should be first dehydrated at 200 °C for 10 h. The specific surface area was calculated by the Brunauer–Emmet–Teller (BET) method using the adsorption and desorption isotherms, respectively. The total pore volume was acquired from the maximum amount of N₂ adsorbed at partial pressure. According to the generally accepted methods [32–34], thermal storage and release tests were performed to investigate the thermal storage/release properties of LA, LA/FA and LA/FAh composites. The samples were, respectively, put into a uniform test tubes. In order to analyze the temperature changes of the samples, one temperature indicator (precision of ±0.10 °C) was placed at the center of each tube. The test tubes were first entered into a thermostat-controlled water bath at the temperature equilibrium of 25 °C, and quickly put into a thermostat-controlled water bath at 80 °C, then put back into a 25 °C water bath until the temperature reached 25 °C.

3. Results and Discussion

SEM was performed to investigate the effect of acid treatment. The morphology of FA is shown in Figure 1a, where FA has a smooth sphere structure. After treatment with HCl, the smooth sphere structure of FA became clearly distinguishable (Figure 1b). The modified FA with high specific surface area and unique porous structure was selected as a supporting material. The decrease of CaO and MgO in FAh was due to the reaction of CaO and MgO compounds with HCl (Table 1). Herein, excessive LA in the composite was removed by thermal filtration, and the residual LA was considered to reach saturation. The LA loadage was calculated from the loss in the composite mass after being heated for 12 h, with a 19.30% mass loss. Thus, the maximum LA content of the composite was 19.30%. The morphology of FA is shown in Figure 1a, where FA has a smooth sphere structure. After impregnation with LA, the smooth sphere structure of FA became clearly distinguishable (Figure 1c). The FA surfaces in the composite were fully covered with a continuous layer of LA. Therefore, LA was impregnated into FA, and the surfaces and pores of FAh were entirely occupied by LA (Figure 1d). HCl acid-treated FA (FAh) mainly included SiO_2 (Table 1); its XRD pattern could be that of amorphous silica according to our previous work [35–38]. The disappearance of the weak mullite diffractions indicated the possible formation of a new phase, which was probably related to the interfacial aspects between LA and FA (Figure 2).

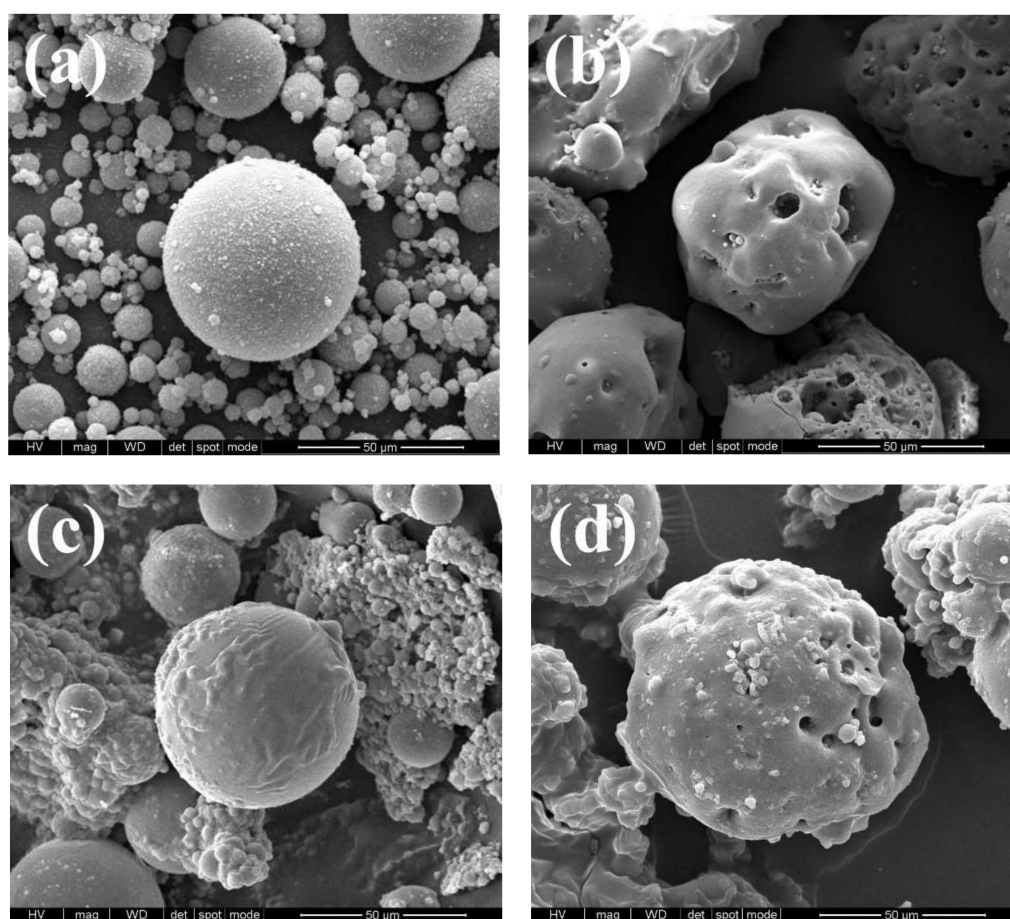


Figure 1. SEM images of (a) FA, (b) FAh, (c) LA/FA and (d) LA/FAh.

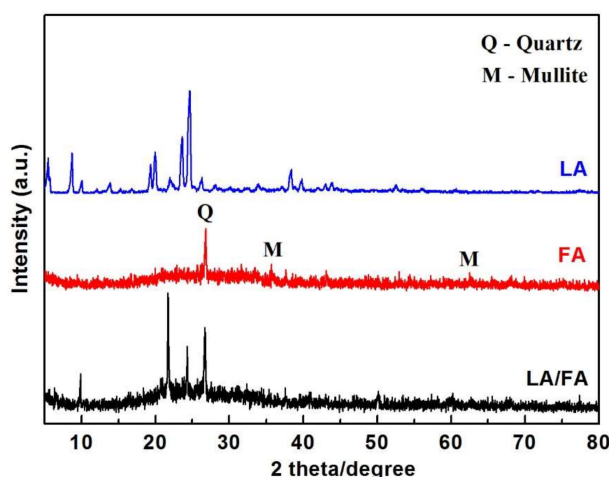


Figure 2. XRD patterns of LA, FA and LA/FA.

LA/FAh composite was prepared using FAh by the same method as described *vide supra*. In addition, the maximum loadage of LA for LA/FAh was determined by the ignition loss method and was 38.12%; it is larger than that of LA/FA (19.30%). The pore structures of FA before and after HCl treatment are shown in Figure 3, and the porous textures of FA and FAh are also given (Table 2). The BET surface area of FAh (52.961 m²/g, after HCl treatment) was higher than that of FA (3.958 m²/g, before HCl treatment) and demonstrated that FAh had more adsorption sites than FA. The total pore volume of FAh (0.036 cm³/g) was larger than that of FA (0.010 cm³/g) and indicated that FAh had more hold space than FA. So, FAh could possess more loading capacity than FA. The thermal storage and release performances of LA/FA and LA/FAh composites were assessed by comparison to those of pure LA. The thermal storage and release curves of pure LA and the composites indicated that the thermal storage ability of LA/FAh is better than LA/FA (Figure 4). By comparing the freezing time of LA/FAh with that of LA/FA, it can be seen that the thermal storage and release rates are 1.85 times higher in LA/FAh than that in LA/FA. The freezing times determined from Figure 4 were also obviously reduced from 930 s for pure LA to 220 s for the LA/FAh composite.

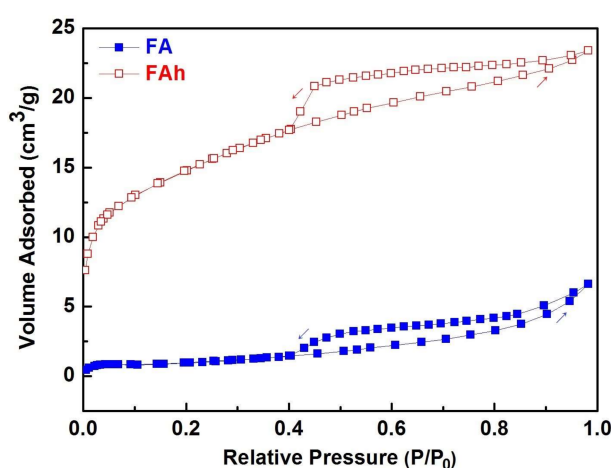
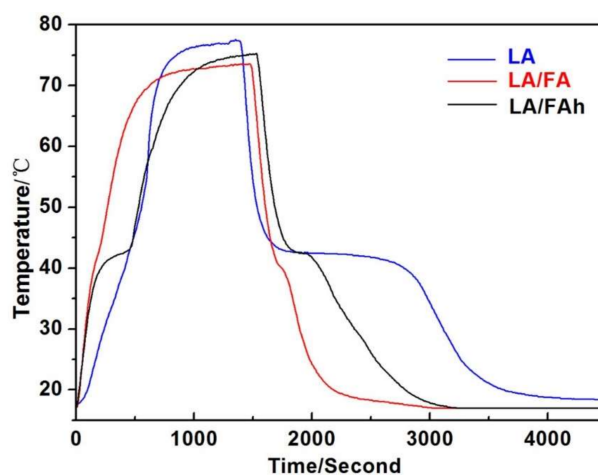


Figure 3. N₂ adsorption-desorption isotherms of FA and FAh.

Table 2. Porous characteristics of FA and FAh.

Samples	BET Surface Area S_{BET} (m^2/g)	Total Pore Volume (cm^3/g)
FA	3.958	0.010
FAh	52.961	0.036

**Figure 4.** Melting and freezing curves of LA and corresponding composites.

DSC thermal analysis was employed to evaluate the latent heat and onset temperature of LA and the composites (Figure 5). Thermal energy storage properties of LA, LA/FA and LA/FAh are given in Table 3. Pure LA has a melting temperature (T_m) of 40.10 °C in the endothermic curve and a freezing temperature (T_f) in the exothermic curve of 40.76 °C; its latent heats of melting (ΔH_m) and freezing (ΔH_f) of pure LA are 235.90 and 206.00 J/g, respectively, but, the latent heats of melting and freezing of the composites seem lower, according to the curves for LA/FA and LA/FAh. The latent heats of melting are determined as 34.09 and 80.94 J/g for LA/FA and LA/FAh, respectively, while the latent heats of freezing are 32.97 and 77.39 J/g for LA/FA and LA/FAh, respectively, but these values are less than their theoretical values (for the LA/FA, ΔH_m : $235.90 \text{ J/g} \times 19.30\% = 45.53 \text{ J/g}$, ΔH_f : $206.00 \text{ J/g} \times 19.30\% = 39.76 \text{ J/g}$; for the LA/FAh, ΔH_m : $235.90 \text{ J/g} \times 38.12\% = 89.93 \text{ J/g}$, ΔH_f : $206.00 \text{ J/g} \times 38.12\% = 78.53 \text{ J/g}$). The lower fraction of LA showed no obvious effect on the decrease of the latent heats of the composites. The crystallinity of LA in the composites should be considered because the interactions between LA and supports would hinder LA from crystallizing and decreased the latent heats of the composites. The crystallinity of LA (F_c) in the composite was calculated by: $F_c = (\Delta H_{\text{composites}} / \Delta H_{\text{pure}} \cdot \beta) \times 100\%$, where $\Delta H_{\text{composites}}$ and ΔH_{pure} are the latent heat of the composite and pure LA, respectively, and β represents the loadage of LA in the composites [39]. The crystallinity of LA in the LA/FA is 74.88% (Table 3), while that in LA/FAh reaches 90.00%, higher than that in LA/FA. This is why the latent heat of melting is 2.37 times greater in LA/FAh than that in LA/FA, while the maximum loadage is 1.98 times higher in LA/FAh than the in LA/FA. In summary, the crystallinity of LA in the composite shows important effects on the latent heats of the composite.

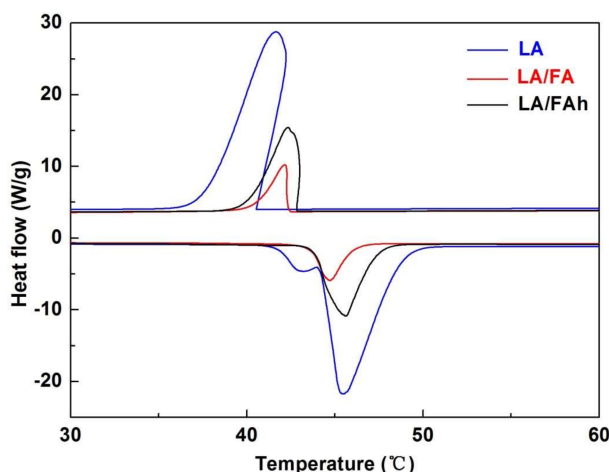


Figure 5. DSC curves of pure LA, LA/FA and LA/FAh.

Table 3. Thermal properties of pure LA, LA/FA and LA/FAh composites.

Samples	Loadage (β , %)	Melting Temperature (T_m , °C)	Freezing Temperature (T_f , °C)	Latent Heat of Melting (ΔH_m , J/g)	Latent Heat of Freezing (ΔH_f , J/g)	Crystallinity of LA (F_c , %)	Extent of Supercooling ($T_m - T_f$, °C)
LA	100	40.10	40.76	235.90	206.00	100	−0.66
LA/FA	19.30	41.34	42.75	34.09	32.97	74.88	−1.41
LA/FAh	38.12	42.34	43.50	80.94	77.39	90.00	−1.16

4. Conclusions

An innovative form-stable phase change material was prepared with lauric acid and modified FA. High specific surface area and unique porous structure of the modified FA was suitable as a supporting material. The composite PCM would retain its morphology in the solid state without seepage of the melted lauric acid due to the new unique porous structure. The latent heat of LA/FAh composite for melting and freezing (80.94 and 77.39 J/g) was higher than that of LA/FA composite (34.09 and 32.97 J/g), respectively. The maximum loadage of LA for LA/FAh was 38.12%, larger than that of LA/FA (19.30%). The obtained composite PCM had a good latent heat and proper melting temperature (42.34 °C) for building thermal energy storage, and LA/FAh composite could have potential applications in the energy storage fields.

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Conflicts of Interest: The authors declare no conflict of interests.

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