

Stearyl Alcohol Modified Expanded Graphite / Paraffin Phase Change Composite

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Abstract: As a phase change material (PCM), paraffin has high latent heat, but low thermal conductivity. In this study, expanded graphite (EG) was added into paraffin to form composite material with both high thermal conductivity, and high latent heat. Moreover, the EG was organo-modified to improve the compatibility of EG and paraffin. The results reveal that thermal conductivity of paraffin (0.30 W/mK) was increased to 8.377 W/mK by adding 30% mass fraction of modified EG. The results also indicated that the heating and cooling times of modified EG/paraffin composites were increased significantly with respect to that of pure paraffin. The experimental results revealed that the use of modified EG can be considered an effective method to improve the thermal conductivity of paraffin.

Keywords: Paraffin; expanded graphite; thermal conductivity.

1. Introduction

Latent thermal energy storage (LTES) using phase change material (PCM) is one of the preferred forms of energy storage, which can provide high energy storage density, and nearly isothermal heat storage/retrieval processes. For such energy storage system, solid-liquid transition is preferred because of the small variation in volume, unlike liquid-gas or solid-gas transitions [1, 2, 3, 4].

Paraffin is one of the most commonly used phase-change materials (PCMs) in storing thermal energy. It is regarded as the most promising PCM for its large latent heat, low cost, stability, non-toxicity, and resistance to corrosion [2]. Paraffin however, has a low thermal transfer coefficient. That is, the melting and solidification of paraffin are slow. In other words, heat transfer through paraffin is slow. Therefore, for storing heat, paraffin is often mixed with a highly thermally conductive material to increase the heat transfer rate [3, 5, 6, 7, 8, 9].

Graphite is a good choice as a highly thermally conductive material to be added into paraffin [3, 5, 6, 7]. Graphite is refractory, highly electrically conductive, highly thermally conductive and highly plastic. Graphite is not metal mineral but exhibits properties of metal organic compounds because its lattice includes layers of atoms are arranged in a compact manner and electrons can easily travel in the lattice for conducting electricity and heat between the atoms. Therefore, graphite exhibits excellent electrical conductivity and thermal conductivity. If paraffin

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is mixed with graphite, the mixture would exhibit better thermal conductivity. Moreover, at the normal temperature, graphite is chemically inert and is stable in strong acids, strong alkalis and organic solutions. Expandable graphite (EG) exhibits a low density and a high heat transfer coefficient and is therefore commonly used as a thermally conductive material. But paraffin and graphite are organic or inorganic, respectively. Hence, it is difficult to mix expandable graphite with paraffin evenly. In this study, we provide a method for making organically modified expandable graphite, so that the expandable graphite will have the functional group of alkyl chain to increase its compatibility with paraffin.

2. Materials and methods

2.1. Materials

Paraffin (Sigma Aldrich) was used as PCM in the preparation of the composite PCMs. Expandable graphite (INCA Technology Co., EH00) was added as modifier to increase the thermal conductivity of composite PCMs. Dimethyl-acetamide (DMAC) was purchased from the Echo Chemical. Octadecanol and dicyclohexylcarbodiimide were supplied by the Alfa Aesar.

2.2. Preparation of EG/paraffin composite

In this research, the expanded graphite was obtained by heating the reactant (expandable graphite, EH00) to 1000°C for 15 s. Then the obtained expanded graphite was added to concentrated nitric acid, and reacted at 120°C for 3 hrs. After filtration, the solid was dried at 70°C for 48 hrs. And then this acid-treated EG powders were added to the mixture of dimethyl-acetamide (DMAC), stearyl alcohol and dicyclohexylcarbodiimide (DCC) to proceed with the organo-modified reaction (Scheme 1) for two days. After filtering the mixture, the solid was dried at 70°C for 48 hrs, the modified EG powders were obtained. Then, the paraffin was melt blended with modified EG powders for 15 min at 70°C in a counter-rotating internal mixer (Brabender PL2000, Duisburg, Germany) with a rotation speed of 60 rpm.

2.3. Characterization

2.3.1. Morphology

The morphology of the samples was observed with a Hitachi scanning electron microscope (Model S-3000N).

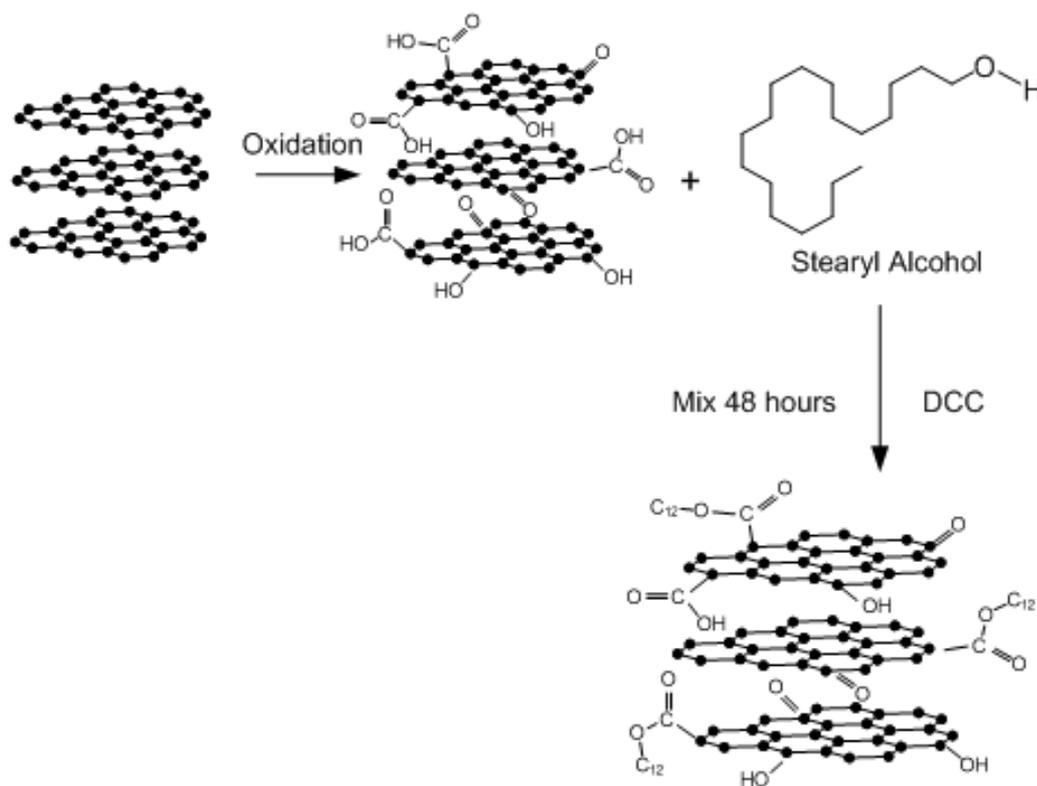
2.3.2. Thermal analysis

Thermal conductivities for all samples were measured at room temperature using a thermal conductivity analyzer (Hot Disk, TPS-500).

2.3.3. Measurement of the heat storage and release

After being heated to the molten state, same volumes of paraffin and phase-change composites were placed into an aluminum tube with inner diameter of 20 mm. A temperature probe was inserted into the center of the sample. After the temperature of the material inside the test tube

maintained constant at 30 °C, the test tube was placed in a water bath tank, in which the temperature was kept constant at 80 °C. Then the heat storage curve was measured. During the experimental process, the temperatures were recorded every 30 s. The heat release curve was obtained by first, the test tube with the phase-change material was heated in a water bath with constant temperature of 100°C. Then, the tube was quickly moved to a water bath with constant temperature of 22°C. The heat curve was completed once the temperature variation was recorded.



Scheme 1. Organo-modified reaction of EG

3. Results and discussion

3.1. Morphology of the PCM composites

Figure 1 shows the SEM photographs of the EG/paraffin composites. Many inter-lamellar microhoneycomb pores in the structure and rough surface were found for the un-modified EG/paraffin composite in Figure 1(a). On the other hand, smoother surface was found for the organically modified EG/paraffin composite in Figure 1(b). It demonstrates that the pore structure of EG is filled with much modified paraffin than un-modified one. Because the organo-modified process of EG generates a large number of alkyl groups on the surface of EG. These alkyl groups are similar to the functional groups in the structure of the paraffin, hence the interaction is chemistry in nature. The structural characteristics and surface properties of the modified EG make it a very strong interaction with paraffin-typed large organic molecules. Therefore, the phase-change material has a greater affinity to the surface of the modified EG.

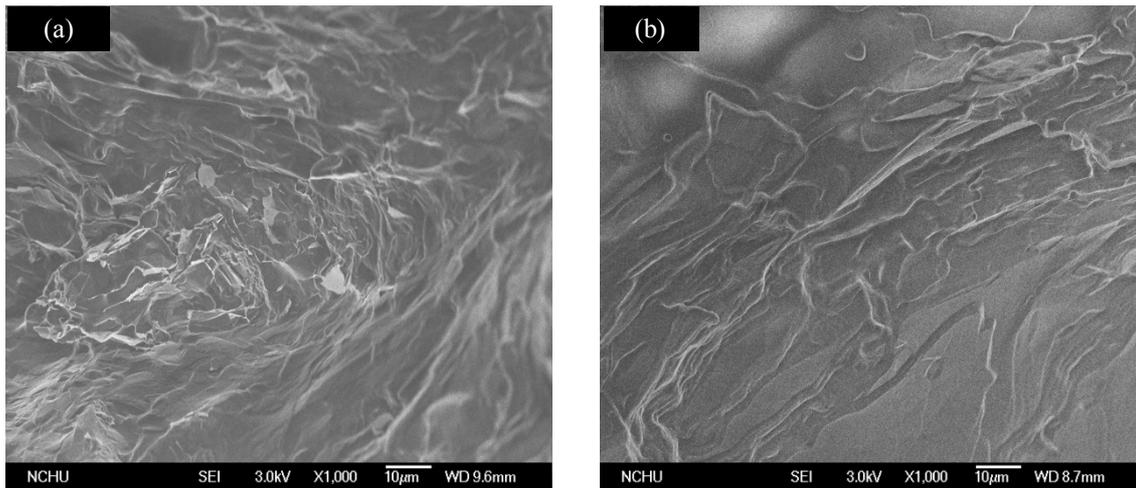


Figure 1. SEM photographs of the (a) un-modified EG/paraffin composite (b) modified EG/paraffin composite

3.2. Thermal conductivity of the PCMs

Figure 2 shows the influence of the EG content on the thermal conductivity of the phase-change composites. With the reduction in the paraffin content and the increase in the modified EG content, the thermal conductivity of the composites increased, especially when the EG content was larger than 20%. The thermal conductivity of paraffin (0.30 W/mK) was increased to 8.377 W/mK by adding 30% mass fraction of modified EG. However, the thermal conductivity of the un-modified EG/paraffin composites was lower than that of modified ones. Moreover, the un-modified EG/paraffin composites cannot be mixed homogeneously as the content of EG larger than 10%. It indicated that the compatibility of EG and paraffin was improved after the organo-modification process.

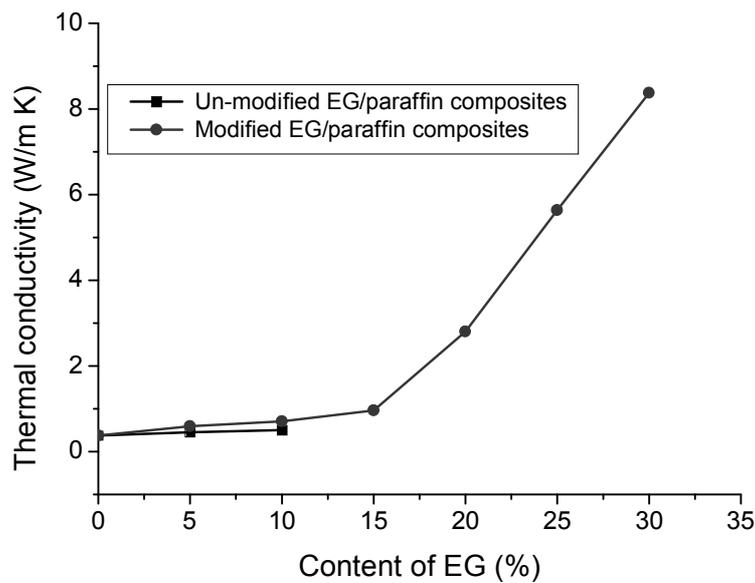


Figure 2. Thermal conductivity of the phase-change composites

3.3. The heat storage and release of PCMs

Figure 3 indicates the heat storage curves of the paraffin and the EG/paraffin phase-change composites. As can be seen from the curves, when the center temperature increased from 30 to 80°C, it took 500 s for paraffin and only 60-200 s for the EG/paraffin phase-change composites to reach the equilibrium temperature of the system. The time required for such temperature raise was reduced by 2.5-8 times. Figure 4 gives the heat release curves of paraffin and the EG/paraffin phase-change composites. The curves shows that after the heat storage process was completed, it took 1600 s for paraffin whereas only 400-800 s for the EG/paraffin phase-change composites for the temperatures to drop from 80°C to around 30°C (exothermic process). The time required than paraffin was decreased by 2-4 times. Whether it is heat storage or release process, the storage (release) time of the phase-change composites was greatly reduced compared with paraffin. According to the results of literature [6, 7], it was found that the solidification and melting rates of the prepared EG/paraffin PCM were just about two times of those of paraffin. The reason is that the modified EG in this study has better compatibility with paraffin, which can greatly improve the dispersion of EG in the paraffin and the heat transfer rate of the EG/paraffin phase-change composites in the heat storage (release) process. Thus, the heat storage and release time was significantly reduced.

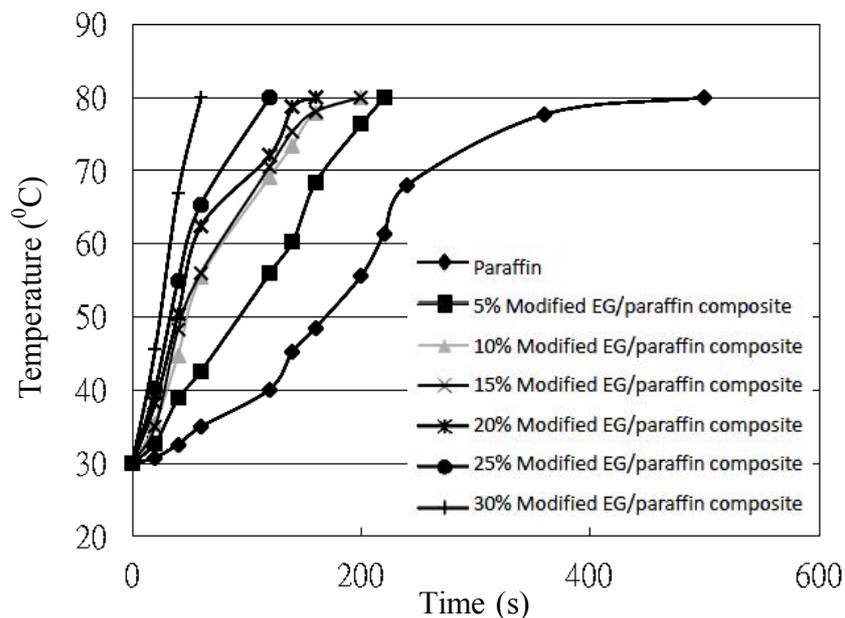


Figure 3. Heat storage curves of phase-change composites at different paraffin contents

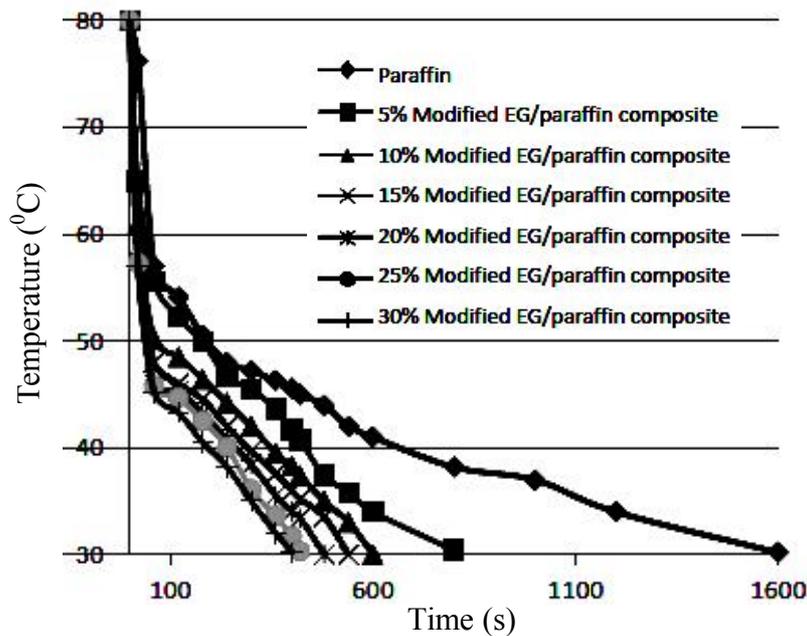


Figure 4. Heat release curves of phase-change composites at different paraffin contents

4. Conclusion

An organo-modified EG/paraffin phase change composite was successfully prepared. The stearyl alcohol modified EG was more compatible with paraffin than the un-modified one. The thermal conductivity of paraffin (0.30 W/mK) was increased to 8.377 W/mK by adding 30% mass fraction of modified EG. Moreover, the heating and cooling times of modified EG/paraffin composites were increased significantly with respect to those of pure paraffin. This stearyl alcohol modified EG /paraffin phase change composite can effectively improve the thermal conduction of paraffin.

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