

# THE EFFECT OF EXPANDED GRAPHITE ON THE CHARGING TIME OF STEARIC ACID PHASE CHANGE MATERIAL IN A HOT WATER TANK

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## Abstract

The present work is to investigate the thermal conductivity of the phase change material (PCM) of stearic acid (SA) by using the supporting material of expanded graphite (EG). The EG is used with the mass ratio of 10%, 15% and 20%. SA is commercial grade and the EG is formed by thermal method using natural graphite powder and crystalline zinc nitrate. The composite materials of (SA/10% EG, SA/15% EG and SA/20% EG) are prepared by direct impregnation method. Thermal conductivity of pure SA and composite PCMs are investigated by laboratory apparatus of heat transfer base unit TD1002. The thermal conductivity of the SA/10%EG composite PCM has been improved to 0.64 W/mK from that of SA (0.28 W/mK) with the percentage enhancement of 129%. To verify the morphology of the materials, scanning electron microscopy (SEM) analysis is utilized. The results show that the EG is formed in multilayers with pores in which stearic acid is well absorbed. The charging characteristics of pure SA, SA/10% EG and SA/15% EG for low temperature latent heat storage (LHS) system are studied experimentally. The system is applied for domestic water heating. The PCM is filled in a 33 mm diameter cylindrical copper tube and placed in hot water storage tank. The water is filled in tank from the top section through the distributor to uniform entry. The phase change phenomena of the PCMs are measured by inserting thermocouples at two layers of PCM tube. The result shows that the melting time decreases with the increasing mass ratio of expanded graphite in composite PCM. Although the melting time of both layers are nearly the same at the experiment of composite PCMs, the lower layer takes longer time than upper layer for pure PCM. The charging time is decreased by 67% for SA/10%EG and 79% for SA/15% EG composite PCM from that value of pure SA.

**Keywords:** Domestic water heating, Expanded graphite, Experimental, Latent heat storage system, Stearic acid, Thermal conductivity

## Introduction

Nowadays, renewable energies are mostly interested in worldwide due to greenhouse effects. Among these energies, solar energy is popular one. But, the solar energy can't get every times. Therefore, energy storage system is required to hold the solar energy for whenever it is necessary to use. There are three types of thermal (heat) storage systems: 1) Sensible heat storage SHS can store the energy for a given range of temperature without phase change. 2) LHS can store large amount of energy within small range of temperature change. 3) Thermochemical energy is the interaction of chemical substances. Out of three types, latent heat storage materials or PCMs are great interested to store and reuse the more energy with change in phase.

PCMs are organic and inorganic types. Organic PCM has the desired properties which are suitable for low temperature LHS system. Stearic acid or fatty acid also has the commonly advantages of low melting temperature (55°C) and high latent heat fusion. It can

also get more easily in affordable price. Meanwhile, the SA has a lack of low thermal conductivity. K. Raznjevic et al., D. Buddhi et al. and N.K. Bansal et al. [1-3] expressed the thermo physical properties of PCMs in which thermal conductivity of stearic acid is 0.29 W/mK. To overcome this effect, the objects having high thermal conductivity values are used to add into PCM such as copper, aluminium and carbon.

Currently, researchers are interested for the improvement of the thermal conductivity of PCM, especially with the additive of EG. Y. Li et al. [4] used the EG to enhance the thermal conductivity of binary nitrates. The result showed that thermal conductivity of nitrate was increased to 8 times by adding 10% EG and recommended that EG is a good one to enhance thermal conductivity for PCM. Y. Li et al. [5] fabricated the composite PCM of Decanoic acid/EG by considering for high stability as well as for excellent thermal conductivity. The result shows that it was achieved the major aims with thermal efficiencies improvement. Bahraseman et al. [6] analysed the efficiency of EG for paraffin type PCM. The penalty results showed that by using 80% EG was 7 times faster the charging time than 10% EG with a small reduction in energy storage.

Moreover, EG is mostly used in composite PCM by mixing with stearic acid. Fang et al. [7] prepared the SA/EG composite PCM with different mass ratios to analyse the characterization of thermal energy storage. Fang et al. found that the phase change conditions and latent heat values of composite PCMs were the same with the positions of pure SA. Choi et al [8] prepared composite PCMs by mixing the EG for three different PCMs. The author summarized that the EG fillers are more significant for thermal conductivity improvement however storage capacity is slightly decreased. Cheng et al. [9] studied and compared the thermal conductivity and thermal performance of SA/EG and SA/CNTs (carbon nanotubes) in low temperature storage system. The result indicated that thermal conductivity of SA/EG was greatly higher than SA/CNTs and EG was more improvable for thermal performance of S.A. Dinker et al. [10] performed the thermal conductivity of SA by mixing the different mass ratios of expanded graphite for low temperature TES. It is noted that the melting time is 21% decreased by adding 20% EG as compared with pure SA. From the above overview, the expanded graphite has been made by acid oxidation method using the hazardous chemicals that cause the environmental pollution and it takes long processing time. Therefore, Nyssanbayeva Gulnura et al [11] introduced the thermal method to form the porous structures of EG which is used for adsorption property. This method is simple and less-polluted approach to prepare the EG.

From above researches, stearic acid is a suitable PCM for low temperature thermal energy storage system. Expanded graphite is an effective supporting material to fill the lack of low thermal conductivity of different PCMs. SA/EG composite PCM increase the conduction heat flow which reduce the melting time of energy storage system. The major aim of this work is the experimental investigation on the thermal conductivity improvement of SA by adding the different mass ratios of EG. To prepare the expanded graphite, thermal method is used. Moreover, the phase change characteristics of PCMs are studied experimentally.

## **Experiment**

### **Material**

SA (commercial grade) imported from Malaysia is used as PCM. Sample of stearic acid is shown in figure 1. Natural graphite powder (HORKIN&WILLIAMS Ltd, England) and zinc nitrate ( $Zn(NO_3)_2 \cdot 6H_2O$ ) (BDH Chemical Ltd., Poole, England) are used to prepared the expanded graphite.



Figure 1. Stearic acid sample

### Preparation of Expanded Graphite

Natural graphite (NG) powder is mixed with the crystalline hydrates of zinc by mass ratio of 1:4 to form the porous structure of expanded graphite. NG 5g and crystalline zinc nitrate 20g are mixed and stirred for 1 minute at room temperature. After that, this mixture is placed in a furnace for 10 minutes which has been setting the temperature of 500°C. Finally, it is cooled in the air and expanded graphite is obtained. The evident expanded volume of EG is compared by using a graduated cylinder as shown in Figure 2. It is observed that the volume of expanded graphite is 43 ml from the original volume of 14 ml with the volume expansion factor of 3.07.

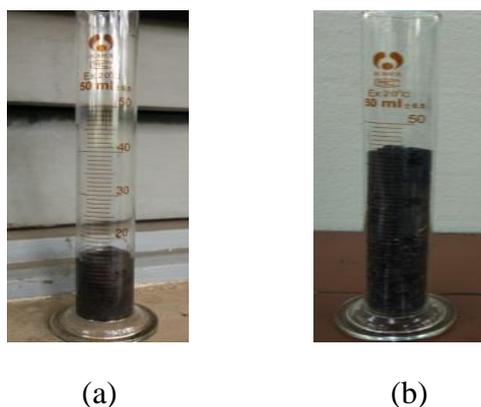


Figure 2. (a) Mixture before heat treatment and (b) Expanded volume after heat treatment

### Preparation of SA/EG composite PCM

The SA is heated with the hot plate until it melted completely. The EG is added to the liquid PCM. This mixture is then stirred for 1 hour with a magnetic stirrer to obtain homogeneous mixture. The percentages of EG in composite PCMs are 10%, 15% and 20%. The composite PCMs are dried in an oven at 60°C for 3 hours. The composite SA/EG PCMs are obtained after cooling naturally. The microstructure of PCMs is investigated by Auriga Cross Beam Workstation FE-SEM (S-4507, Auriga, Germany) with a high-resolution image quality.

### Thermal Conductivity Measurement

Zhao et al. [12] reported the several measurement techniques of thermal conductivity and thermal conductance for the solid state materials. There are four measuring methods, absolute technique, comparative cut bar, which is based on one-dimensional steady state linear heat flow and used the Fourier's Law Equation 1. The heat rate is employed the actual

amount of heat flowing through the sample and equal with those of standard materials (upper rod and lower rod). The authors recommended that the comparative cut bar technique is widely used with the best accuracy.

N. Thet Swe Aye [13] analyzed the heat transfer characteristics of SA (melting temperature  $55^{\circ}\text{C}$ , latent heat  $256\text{ kJ/kg}$ , thermal conductivity  $0.36\text{ W/mK}$  and density for solid state  $828.57\text{ kg/m}^3$ ) used in LHS unit. Thermal properties of SA were measured and thermal conductivity value by the help of professor from Kumamoto University situated in Japan. Thermal conductivity of sample was calculated by Fourier's law equation in which the mean heat flux was used. The heat flux values of upper rod and lower rod were evaluated by temperature gradient and thermal conductivity of rods.

For the present study, laboratory apparatus of basic Heat Transfer Unit TD1002 shown in Figure 3 is used to measure the thermal conductivity of PCMs. It is constructed with a fixed bench frame including test bar, digital display, a suitable electrical supply and cold water input and output are controlled by manual check valve.

The apparatus consists two solid brass bars and measured sample which is sandwiched and changeable. The upstream (upper) rod includes the electric heater, and downstream (lower) rod also includes water-cooled chamber. The two rods are attached with two thermocouples respectively.



Figure 3. Photo of heat transfer base unit TD1002

The amount of power from the heat source is supplied  $5\text{ W}$  due to allowable melting temperature of PCM and cold water temperature is kept at approximately  $10^{\circ}\text{C}$ . Thermocouples are fitted with  $20\text{ mm}$  apart respectively as shown in Figure 4.

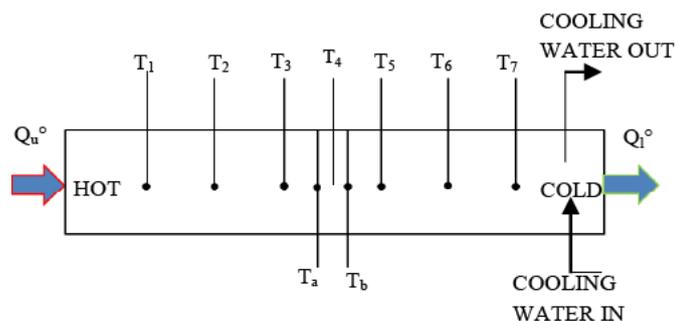


Figure 4. Schematic of thermocouples arrangement

Firstly, pure SA and composite PCMs of SA/10%EG, SA/15% and SA/20%EG are fabricated into the circular discs with the dimensions of 30 mm diameter and 8.5 mm thickness, as shown in Figure 5. The samples are perfectly insulated by the foam material and complete spread of the thermal conductive paste over these surfaces. When the temperature distribution of sample is equilibrium, the measured temperatures are carried out. The experiment is performed by three times for each sample. The uncertainty of experimental results for PCMs are found within  $\pm 0.02$  W/mK.



Figure 5. PCM sample of (a) Pure SA and (b) SA/EG composite

Thermal conductivity of sample is then calculated by Fourier's Law Equation 1.

$$Q^{\circ} = -kA \frac{dT}{dx} \quad (1)$$

Where,  $Q^{\circ}$  is the heat transfer rate,  $k$  is the thermal conductivity,  $A$  is the surface area of rod,  $dT$  is the temperature difference and  $dx$  is the respective distance of  $dT$ . The mean value of heat transfer rate applied to the sample,  $Q_m^{\circ}$  is calculated by equation 2 [13].

$$Q_m^{\circ} = \frac{Q_u^{\circ} + Q_l^{\circ}}{2} \quad (2)$$

Where,  $Q_u^{\circ}$  is the heat transfer rate of upper rod and  $Q_l^{\circ}$  is the heat transfer rate of lower rod.

### Thermal Energy Storage Measurement

The phase change characteristics of PCMs during melting process are investigated experimentally. The encapsulated PCM is used in the main storage unit. The temperature of hot water is set to  $65^{\circ}\text{C}$ . The flow rate is constant at 6 lit/min. The hot water enters the top position of storage tank through the flow distributor.

**Table 1: Dimension of Main Storage Tank and Heat Source**

	Main Storage Tank	Heat Source Tank
Outside Diameter, mm	470	530
Inside Diameter, mm	320	365
Height, mm	600	1060
Height with Base, mm	650	-

The tanks are made of steel 304 and insulated with polystyrene foam. The water capacity is 120 liters for heat source and 48 liters for main storage tank. A heater is used to obtain the constant heat source for instead of solar energy. The dimensions of main storage tank and constant heat source tank are shown in Table 1 and the experiment set up is shown in Figure 6.

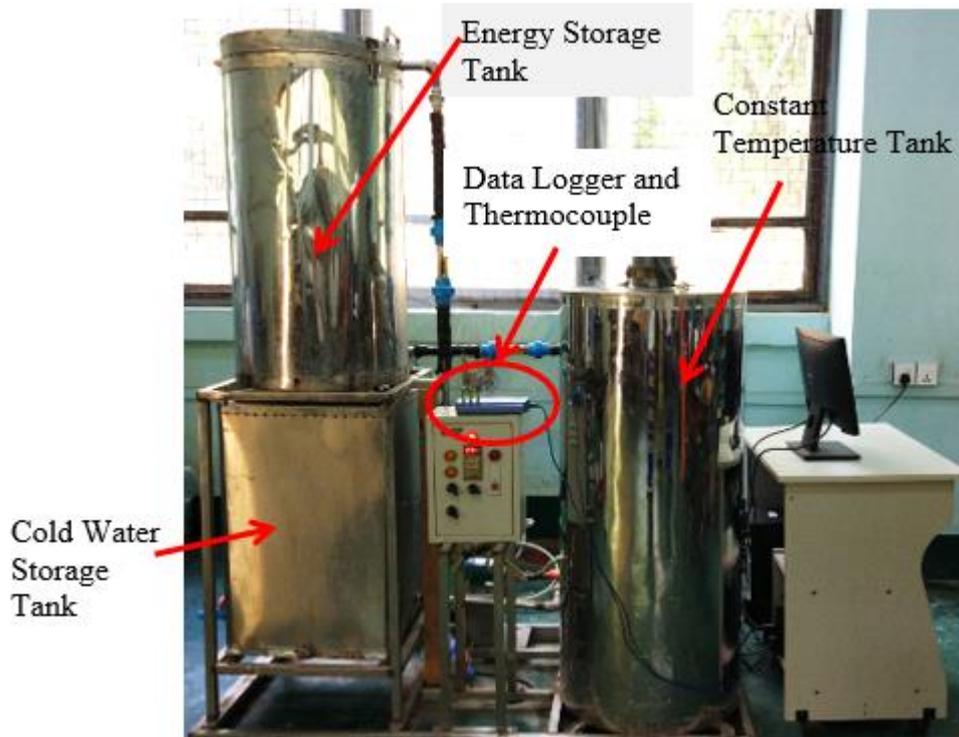


Figure 6. Photo of the experimental setup

To store for the latent heat, four PCM tubes are used in the storage tank. PCM is filled with 70% of tube volume which outside diameter is 35 mm and length is 550 mm. The tube arrangement and exact spacing of thermocouple attachments are shown in Figure 7.

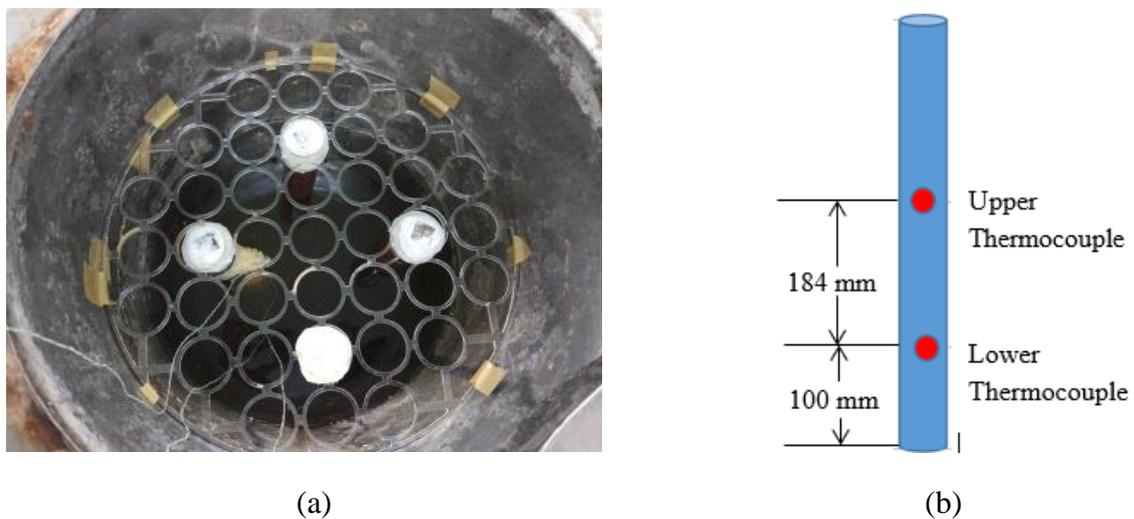


Figure 7. (a) PCM tube arrangement and (b) Spacing of thermocouple in PCM tube

## Results and Discussion

### Microstructure of the Samples

After preparing the composite phase change material, SEM analysis is performed to observe the topographic of material. The SEM images of natural graphite, EG, and SA/EG composite PCM with 10wt% of EG are shown below (Figure 8).

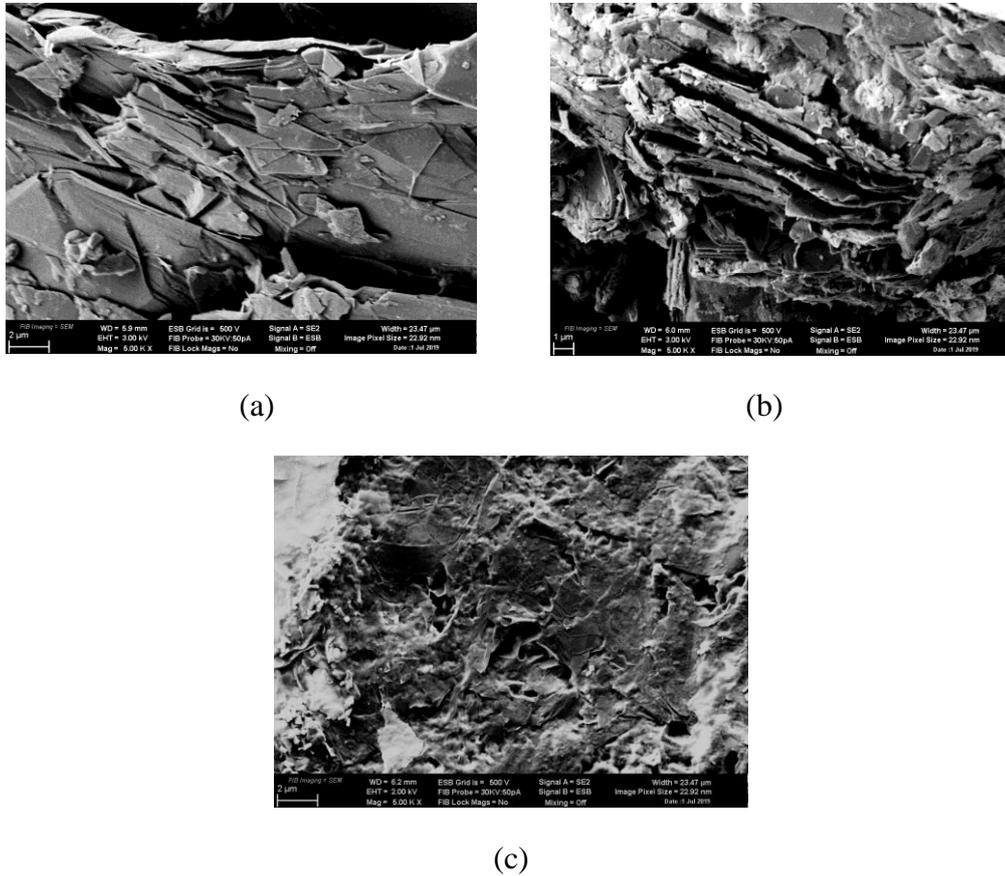


Figure 8. SEM images of (a) Natural graphite (b) Expanded Graphite (c) SA/EG composite PCM

Figure 8(a) is the image of natural graphite powder which has the flattened structure with no pores. The structure of the EG is not the same as that of natural graphite as it can be seen in Figure 8(b). The structure of the graphite is totally changed to another shape and the pores have appeared. The graphite is said to be expanded after heat treatment. This expanded surface area is more effective for conduction heat flow. Figure 8(c) displays the image of the composite PCM in which SA is entered inside the separated layers of the EG. It shows that the physical absorption of stearic acid is completed in the layers of EG as the mixture is homogeneous.

Figure 9 shows the thermal conductivity of pure stearic acid and composite PCMs. The thermal conductivity of the PCM is significantly improved with the amount of EG content. The thermal conductivities of the composite PCMs of SA/10%EG, SA/15%EG and SA/20%EG are 0.64, 0.745 and 0.93, which are higher than that of pure SA. It is found that the EG achieves the heat transfer improvement of the composite materials more effectively. Therefore, the thermal conductivity values of supporting materials became higher, these values of composite materials will be increased.

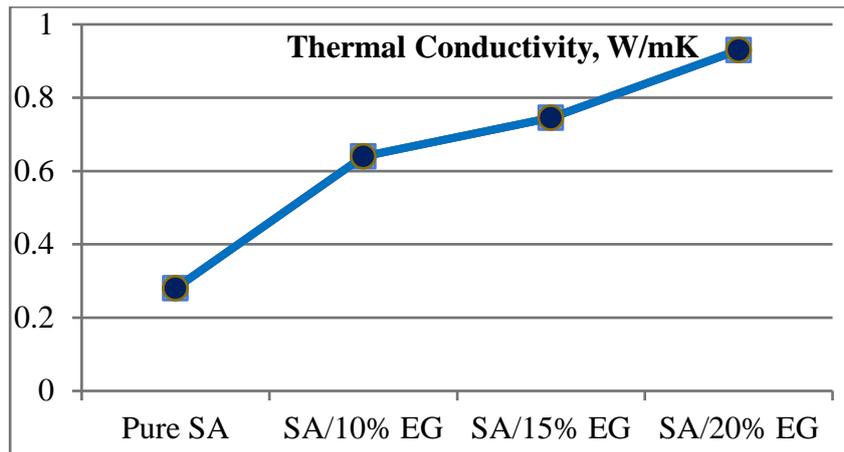


Figure 9. Thermal conductivity value of pure SA and SA/EG composite PCMs

The phase change phenomena of PCMs in vertical cylindrical tube are investigated. The temperature variations of the PCMs at two layers of 70% volume filled PCM tube are shown below. There are four PCM tubes in the storage tank where the initial temperature of PCMs is approximately 30°C with the volume flow rate of 6 litres/min. Then, the inlet HTF is maintained to constant temperature of 65°C.

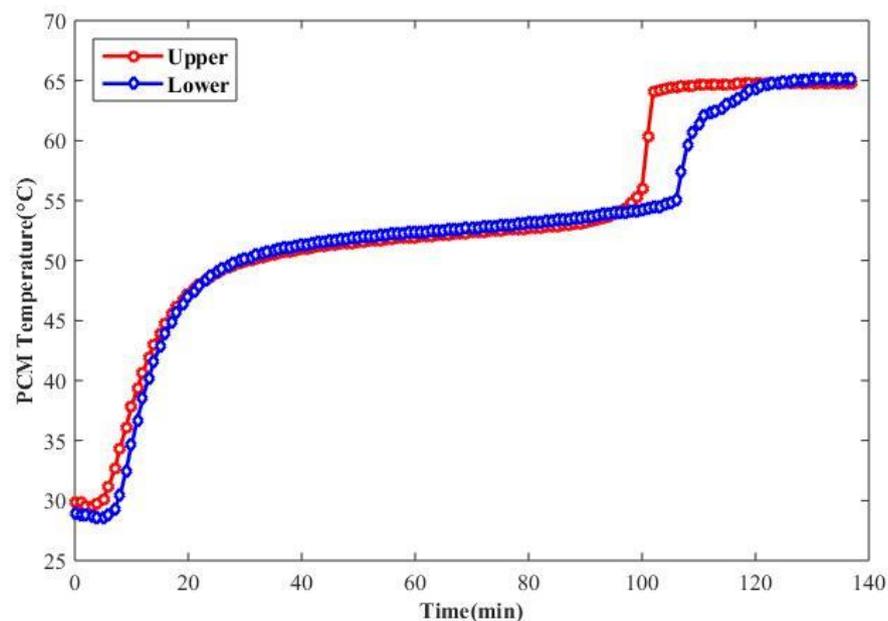


Figure 10. Temperature profile of pure stearic acid PCM

The temperature profile of pure stearic acid at two layers of vertical PCM tube is shown in Figure 10. It is found that the temperature of both layers at the beginning of the process rises rapidly until they reach the melting temperature of 51°C. It takes about 30 minutes for both layers to reach the melting temperature. After that point, the temperature of both layers rises very slowly until the PCM melted completely at 55°C. And then, the temperature of the PCM rises rapidly during the heating of the liquid PCM. The time taken to complete the melting process (55°C) is 99 minutes for the upper layer and 106 minutes for the bottom layer. Hence, the total time to complete the charging process of both layers is 127 minutes.

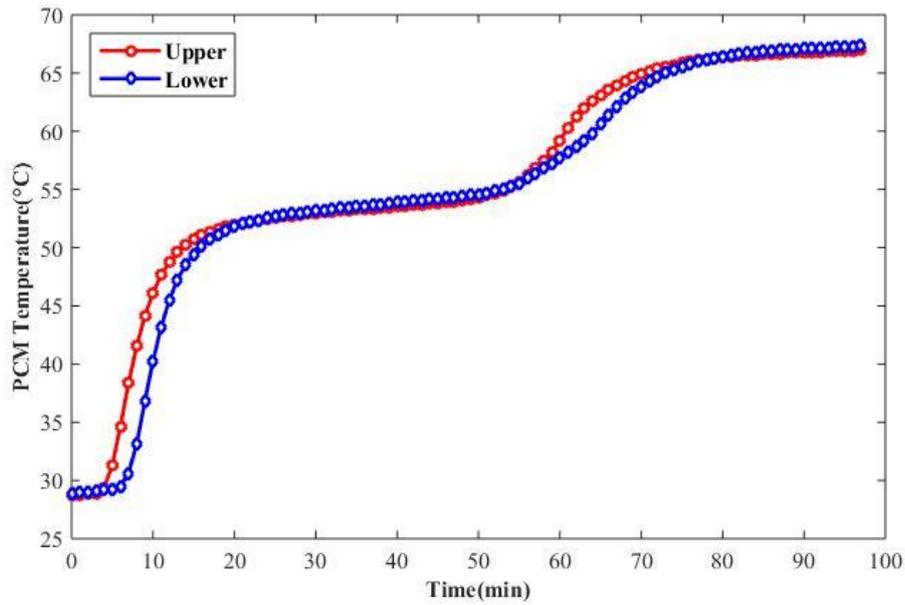


Figure 11. Temperature profile of SA/10%EG composite PCM

Figure 11 illustrates the temperature profile of SA/10% EG composite PCM at two layers of PCM tube. The temperature of PCM increases to reach the melting point (51°C) in 16 minutes for the top layer and 18 minutes for the bottom layer. The time required to complete melting of both layers is 53 minutes which is faster 49 minutes for top layer and 56 minutes for lower layer than those of pure PCM. It is observed that the slope of composite PCM is steadily increased while that of pure PCM is more steeply. The time taken to complete the charging process for both layers is 76 minutes which means it decreases 67% more than pure PCM.

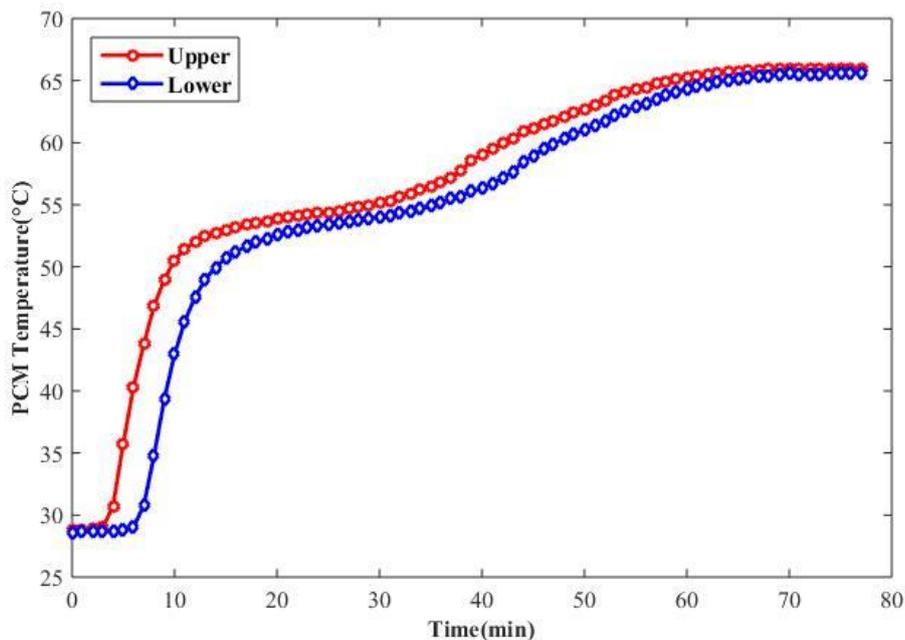


Figure 12. Temperature profile of SA/15%EG composite PCM

Figure 12 shows the temperature variation of SA/15% EG composite PCM at two layers of PCM tube. The temperature of PCM increases to reach the melting point (51°C) in 12 minutes for the top layer and 15 minutes for the bottom layer. The time required to complete the melting of top layer is 37 minutes (168% decreased than pure PCM) and 40 minutes for the lower layer (165% decreased than pure PCM). It is found that the slope of composite PCM is more uniform than the above two cases. The time taken to complete the charging process for both layers is 71 minutes (79% decreased than pure PCM).

## Conclusions

In the present work, the preparation, thermal conductivity measurement and phase change characteristics of PCMs have been studied experimentally. Thermal conductivity value of pure stearic acid and composite PCMs of SA/EG with the mass percentages of 10%, 15% and 20% are prepared for the application of domestic water heating. The SEM analysis is made to verify the microstructure of materials. Thermal conductivity values are measured by using the laboratory apparatus of heat transfer base unit TD1002. The melting process of LHTES system is studied experimentally. PCM is packed in 33 mm cylindrical copper tube and placed in vertical direction inside the LHS tank. The water is used as heat transfer fluid which filled surrounding the PCM tubes.

The findings are summarized as;

1. Thermal conductivity value is increased by 123% for 10% EG, 168% for 15% EG and 229% for 20% EG as compared with the pure stearic acid of 0.28 W/mK.
2. The charging time is decreased by 67% for 10% EG and 79% for 15% EG using in composite PCM as compared with that of pure stearic acid.

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