The effect of expanded graphite on the performance of paraffin phase change materials used in thermal energy storage

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Abstract. Paraffin is the most commonly used phase change type of material for thermal energy storage but the main drawback is it has very low thermal conductivity. This study aims to enhance the thermal conductivity of the base PCM through the use of expanded graphite as supporting material. The expanded graphite at different percentage 3wt%, 6wt%, 9wt%, 12wt%, 15wt% is mixed with paraffin and five samples are prepared. Experimental results obtained on the samples clearly show that the thermal properties of samples are improved when compare to that of pure paraffin. It is found that both thermal conductivity and thermal diffusivity are increasing rapidly. Melting temperature is standard there is no change for all the compositions. Specific heat gradually started increasing during the initial time and then it started decreasing as the expanded graphite percentage increased. The latent heat value of this composition has decreased a lot when compared to pure paraffin.

Keywords: Phase change material; Paraffin; Expanded graphite; Thermal Properties; Thermal energy storage

1. Introduction
In recent years, due to the problems of reduction of quantity conventional energy sources which are a good source of energy, the need is to develop new modern techniques to provide the means to employ good sources of energy that arose [1-3]. Thermal energy storage (TES) plays an important role in protecting of available energy and its reasonable use, as it allows to dissociate between energy production and demand. Nowadays the efficiency of thermal energy storage is increasing and there is the main problem in this era. The use of thermal energy storage is very important in many fields, so by using the proper materials, it could be achieved very effectively. Many research proved that phase change materials (PCMs) are the most facilitative for storing a large amount of heat energy because of their natural high latent heat values. Most of the PCMs have got unique qualities to store thermal energy [4,5]. Phase change materials have been classified into three types mainly they are organic, inorganic and eutectics. Paraffin falls under the category of organic PCM. Many varieties of organic PCMs are available but paraffin has got unique characteristics such as they are non-corrosive, stable and inert, less expensive, non-toxic, it has come up with properties like high latent heat. During the process of melting it has properties like lowering volumetric change and low vapour pressure [6-9] (Gulfam Raza et al. 2016; Patrik Sobolclia et al. 2016; Shalabya et al. 2014; Adil et al. 2018. PCMs have received greater interest in the field of thermal energy storage. PCMs have application in a large number of fields, such as the use of solar energy, active and passive cooling of electronic devices and recovery of waste heat from industries, etc. Thermal energy can be stored in the internal energy of a material in the form of sensible heat, latent heat and chemical energy. Of these, latent heat storage is
the most efficient energy storage technique. For example, it takes eight times more energy to melt water than it does to raise water temperature. PCM has come into demand because to make use of energy when energy not available as we know the solar water heater works during all the day time when natural renewable source sunlight is available but the problem is the working of the same solar heater during night time when sunlight is not available. So to make it work during energy is not available PCM is used for storing thermal energy. Phase change material is used mostly because it is capable of storing a large amount of energy and releasing energy when it is required. PCM is a substance with a high fusion of heat which melts and solidifies at a certain temperature. When the material changes from solid to liquid, the energy is stored and when the material changes from liquid to solid, the energy is liberated [10]. Thermo-physical properties of PCM at melting point are important in determining the material's suitability. The PCMs should have the properties such as high specific heat, high thermal conductivity, stable composition, chemically inert and nontoxic. On the other hand, there are certain reasons which making its efficiency less, like high initial cost, less space availability and lesser sunshine [11,12]. Phase change materials undergo phase change during heating, either solid-liquid, solid-solid or liquid-gas. In thermal energy storage applications, PCM take an advantage because during phase change lot of latent heat is required and during phase change lot of energy is required to break down the bonds also temperature remains constant throughout the period because the system needs to run for longer periods [13,14]. Phase change materials (PCMs) with high thermal conductivity are used to fill the gap between a heat source transfer surfaces is essential to meet the increasing demands of heat dissipation for higher power device. Paraffin-based thermally conductive insulating phase change materials have always been studied as thermal interface materials because they have attracted particular attention with regard to their capacity to store and release thermal energy with a capacity high storage, going through liquidation and solidification in a small temperature range [15-17]. PCM has applications in different fields such as air-condition, telecom shelters, transportation, automobiles, electronics and catering.

Thermal energy storage (TES) is an innovation that stores thermal energy by heating or cooling a storage medium so that the stored energy can be used later for heating and cooling. TES systems store energy in the form of heat for later use. Extensively, TES systems include three main stages: thermal loading, thermal storage and thermal discharge. Materials can store heat in three essential ways, involving sensible heat, latent heat, and chemical reactions [18]. By mixing expanded graphite with paraffin phase change material thermal diffusivity and thermal conductivity are increased. As they are initially at a very low level. In this work paraffin is used as PCM because of all its unique qualities and expanded graphite is used as supporting material for increasing the thermal properties because it is very good example of material having high thermal conductivity [19]. PCMs have a significant feature of high heat storage because of this reason they are chosen as sensible heat storage material. A large variety of PCMs is available for different temperature ranges. Differential scanning calorimetry (DSC) is used to find the thermal properties of PCMs of basic properties such as melting and crystallization temperatures, the specific heat of melting and crystallization or the specific heat capacity. However, DSC gives information about samples that are very small in size because it has its capacity. In case if want to find out for a larger sample it need to make an experimental set up. A PCM integrated into photovoltaic panels absorbs the excess heat generated from solar radiation and reduces the temperature of the cells, therefore providing an effective passive thermal buffering method and improving their efficiency. In continuous demand and supply of solar energy, the thermal energy storage has been developed for storing the thermal energy in the form of PCM. The role of natural convection has shown the melting process of paraffin in thermal storage [20].

2. Experimental method
In this work, the performance of paraffin with extended graphite as a phase change material is studied. The PCMs have different materials but paraffin is chosen because of its unique qualities. The drawback is it has a very low thermal conductivity which is making it weaker as thermal storage
property. To find a solution to this problem, the expanded graphite is mixed with paraffin because of its high thermal conductivity which is going to improve the performance of paraffin after mixing. The samples are mixed at different ratios and the weight of the calculated amount is chosen for preparing the samples. After the preparation of samples, the kd2 pro analyzer which is shown in figure 1 is used for finding thermal conductivity and diffusivity. The melting temperature and specific heat are found using differential scanning calorimeter (DSC). The schematic diagram of DSC is shown in figure 2. The test has been conducted in a laboratory and the obtained results show that the good performance can be obtained in PCM using paraffin with expanded graphite material as thermal storage because both thermal conductivity and diffusivity are increased.

![Figure 1. KD2 PRO analyzer](image1)

![Figure 2. Schematic diagram of differential scanning calorimeter](image2)

2.1. Materials

The required materials for this project are pure paraffin (P) and expanded graphite (EG). Paraffin is chosen as PCM because of its properties and from the past few years, expanded graphite has been adversely used for improving the thermal conductivity of paraffin because expanded graphite is a good example of high thermal conductivity. Each material weighing about one kilogram is purchased from a chemical store. The pure paraffin and expanded graphite are shown in figure 3 and 4. The properties of paraffin are shown in table 1.
### Figure 3. Pure paraffin

### Figure 4. Expanded graphite

### Table 1 Properties of paraffin

| Material | Mass (Grams) | Melting Temperature (°C) | Thermal Conductivity (W/mK) | Thermal Diffusivity (m²/s) | Latent Heat (kJ/kg) | Heat (kJ/kg K) | Specific Heat (kJ/kg K) |
|----------|--------------|--------------------------|-----------------------------|----------------------------|---------------------|-----------------|------------------------|
| Paraffin Wax | 100 | 68.2 | 0.3712 | 1.42x10⁻⁷ | 190 | 2.14 |

#### 2.2. Sample preparation

The paraffin PCM has thermal conductivity of 0.3712 (W/mK). The five different samples are prepared for conducting the tests. Sample preparation includes the following steps as follows. Paraffin wax about the calculated amount is taken for conducting tests for all different samples. The solidly structured paraffin is placed in oven then paraffin is heated at 100°C and after some time it starts melting, the volume of paraffin increases while it changes from solid to liquid state and due to continuous heating paraffin turns into liquid state now calculated amount of expanded graphite is added to the liquid state paraffin at 40°C then start mixing both the materials sample using magnetic stirrer because expanded graphite has to be distributed uniformly in paraffin and it should go into the porous of paraffin and form paraffin-expanded graphite structure. In this same process, five different samples are prepared according to the calculated ratio of paraffin and expanded graphite for conducting tests. The prepared samples are gone under the curing process for twenty-five days at room conditions.
temperature this process is undergone to make composite tough and harder to bring back to its original properties. One sample of paraffin with expanded graphite is shown in figure 5.

![Figure 5. Paraffin with expanded graphite (sample)](image)

### 3. Results and Discussion

#### 3.1. Thermal conductivity

In this experiment kd2 pro analyzer is used for finding the thermal conductivity of five different samples. There are different types of sensors available in kd2 pro but in this experiment TR-1, a single needle sensor is used because it is the best sensor for finding thermal conductivity. To find thermal conductivity, single needle sensor is placed inside the sample and here is how it works: a heater and temperature sensor are housed inside needle then current is passed through the heater and temperature of probe is monitored over time, an analysis of probe temperature is used to calculate thermal conductivity that deep and complicated math amend is used to deal with problems, all of these are accomplished with push of button; power button is turned and by activating in the menu it displays the value. During the addition of expanded graphite to paraffin thermal conductivity started increasing gradually as the percentage of expanded graphite is increased. The rate of heat transfer is found to be increasing by mixing paraffin with expanded graphite. It seems thermal conductivity is increasing constantly when the ratio of expanded graphite is more with paraffin.

#### 3.2. Thermal diffusivity

It describes how quickly a material can change its temperature. In this test to find the thermal diffusivity using a kd2 pro analyzer but the sensor used in this test is different in this test SH-1 sensor is used for finding thermal diffusivity because it is the best sensor to find thermal diffusivity for solid samples. The double-needle sensor is used for finding out thermal diffusivity by placing the needle in all different samples here is how it works: heater is housed in one needle and temperature is housed in another separate needle then both the needle are placed separately inside in the composition and accomplished by push of button and values are displayed on the screen note it down. These values show an increase in thermal diffusivity of paraffin by mixing expanded graphite. Thermal diffusivity of paraffin can be increased by mixing expanded graphite as supporting material.

#### 3.3. Melting temperature

There is a thermo analytical technique called DSC in which the difference in the amount of heat required to increase the temperature of a sample and used for finding out the melting temperature of this composition during starting of the experiment the DSC equipment is brought to initial temperature to obtain good results. The change in substance from one state to another state is defined as melting temperature. Generally material around 52-58°C produce best results. In these experiments, C is used
for finding the melting temperature of this composition. To receive accurate results, a DSC has to be regularly calibrated if necessary should be adjusted also both the sample and reference side should not differ more than 0.2 milligrams. The samples of calculated weighted are placed in the crucibles by calculating the weight on the weighing box and lid of the crucible is closed by applying force with crucible press and crucible is sealed.

3.4. Specific heat

The store and release amount of thermal energy increase with the addition of expanded graphite as there is a slight decrease in CP value while there is an increase in expanded graphite. The amount of heat per unit mass required to raise the temperature by 1°C is defined as specific heat. The specific heat is calculated for all the samples by using differential scanning calorimeter. It is necessary to determine the accurate weight of the sample because melting enthalpy taken is a value normalized by weight. Place the crucible support near the DSC and open the sample change cover then using tweezers place the crucible containing sample on the position of the sample changer tray. Make sure that an empty reference crucible is already on the reference side of the sensor and now start the experiment.

3.5. Latent heat

DSC is the most frequently used technique in a thermal analysis it is used to study the behaviour of materials as a function of temperature or time. In this experiment, a sample is placed in a crucible or tray, which is located directly above the sensor, both the sample and the reference crucibles are surrounded by a heated chamber or oven. The sensor is the heart of DSC and detects heat flow. The cooling option is often necessary to perform the experiment below room temperature. Cooling is accomplished using an air flow around the oven or using a cryostat or intercooler. Samples can simply be loaded into the oven when using the dry gas inlet.

The obtained results for five different samples 3%, 6%, 9%, 12%, and 15% of expanded graphite mixed with pure paraffin show that thermal conductivity is found to be increasing by mixing paraffin with expanded graphite it clearly shows that thermal conductivity is about 5 times higher than pure paraffin. The rate of heat transfer for paraffin will be higher after mixing with expanded graphite and better performance is resulted by PCMs by using this composition. In the case of thermal conductivity, it is kept on increasing so that the change in temperature of the material can be gained so fast. Melting temperature is standard there are no changes but the drawback is value is around 68.2°C in which the better results cannot be obtained. Specific heat is found increasing at a lower percentage later it decreased up to a certain extent at last at a higher percentage again it started increasing the overall performance is found increasing and decreasing. Properties of samples are shown in table 2.

| Table 2 Properties of paraffin and expanded graphite |
|-----------------------------------------------------|
| Material   | Mass (Grams) | Melting Temperature (°C) | Thermal Conductivity (W/mK) | Thermal Diffusivity (m²/s) | Latent Heat (kJ/kg) | Specific Heat (kJ/kg K) |
|------------|--------------|--------------------------|----------------------------|---------------------------|-------------------|------------------------|
| Sample 1   | 97P+3EG      | 68.3                     | 0.892                      | 5.48x10⁻⁷                 | 140.33            | 2.08                   |
| Sample 2   | 94P+6EG      | 68.3                     | 1.61                       | 9.16x10⁻⁷                 | 126.87            | 1.92                   |
| Sample 3   | 91P+9EG      | 68.4                     | 1.992                      | 1.02x10⁻⁶                 | 118.44            | 1.88                   |
| Sample 4   | 88P+12EG     | 68.3                     | 2.283                      | 1.22x10⁻⁶                 | 112.56            | 2.1                    |
| Sample 5   | 85P+15EG     | 68.4                     | 2.418                      | 1.32x10⁻⁶                 | 108              | 2.26                   |
4. Conclusion

The main aim of this work is by using EG as supporting material to increase the thermal conductivity of paraffin which is the most commonly used material in PCM for the storage of thermal energy. Using five different samples tests are conducted and the obtained results clearly show a significant change in thermal conductivity and thermal diffusivity of paraffin by the addition of expanded graphite about 3-15%. When compared to pure paraffin the paraffin mixed with expanded graphite gives better performance and thermal properties are improved a lot. The obtained results show that the performance of PCMs can be increased by using this combined mixture of paraffin and EG. The increase in thermal conductivity results shows that the rate of heat transfer will be more in PCMs and can store in a larger amount. Thermal diffusivity is also increased so that the material used in thermal energy can acquire a fast change in temperature as compared to pure paraffin.

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