



Preparation and Characterization of Paraffin / Graphite Composite as Thermal Energy Storage Materials

Dihia Djefel ^{#1}, Said Makhlouf ^{#2}, Souad Khedache ^{#3}, Gilles Lefebvre ^{*1}, Laurent Royon ^{§1}

[#], *L.M.S.E. Laboratory, Mouloud Mammeri University,
Po Box 17 RP 15000, Tizi Ouzou, Algeria.*

¹ dihia.dj@gmail.com

² saidmakhlouf@yahoo.com

³ souadkhedache01@gmail.com

^{*}*CERTES-IUT Laboratory, Paris Est Créteil University,
61 Av. Général de Gaulle, 94010 Paris Créteil, France.*

^{*} glt.lefebvre@laposte.net

^{*}*Matière Système Complexe Laboratory, Paris Denis Diderot University,
UMR 7057 CNRS 75013 Paris, France*

[§]laurent.royon@univ-mlv.fr

Abstract— Paraffin/Graphite composite as thermal energy storage material was prepared using impregnation methods. In the composite, graphite acted as the supporting material, and paraffin is selected as the phase change material (PCM) due to the desirable thermal properties to the thermal storage by latent heat. Environmental scanning electronic microscope (ESEM) was used to determine microstructure of graphite (Gr) and Paraffin/Graphite composite (P/Gr composite). The thermal properties, such as phase change temperature and latent heat, and thermal stability were investigated by a differential scanning calorimeter (DSC) and thermogravimetric analyzer (TGA). Thermal conductivity was determined by a Hot disk Method. The SEM results showed that paraffin was well dispersed in the porous network of graphite. The DSC results indicated that the composites solidify at 55.16 °C with a latent heat of 110.83 J/g and melt at 50.97 °C with a latent heat of 111.31 J/g when the mass percentage of paraffin in the composites was 55%. TG analysis revealed that the P/Gr composite PCMs had good thermal stability above their working temperature ranges. The thermal conductivity results showed that addition of graphite improved thermal conductivity. The thermal conductivity of the Paraffin/Graphite composite was 7.5 higher than that of paraffin alone.

Keywords— PCM composite, thermal properties, paraffin, graphite, thermal energy storage

I. INTRODUCTION

The thermal energy storage by latent heat using phase change materials (MCPs), there has been growing interest in research in energy, due to the high density of thermal storage in almost constant conditions and of the role important in

achieving energy savings in a variety of applications [1-5]. Among the large number of MCPs studied [6], paraffins are considered promising for the latent heat storage due to their appropriate thermal properties, their chemical and thermal stability, lack of supercooling, noncorrosive their behavior, and their non-toxicity [7-8].

Nevertheless, the problem remains of MCPs finding appropriate methods for their incorporation into functional structures to avoid the leakage problem at the solid-liquid transition phase.

Therefore, special items or spherical heat exchangers containing MCPs are required to encapsulate the MCP operation that cannot be achieved without additional cost. Until recently, this problem has been solved by using porous materials which can be prepared by impregnation of the porous structure MCP in various support materials. The graphite was utilized as additive or matrix for improved thermal conductivity of composite PCM [9-10].

In the present study, a PCM composite based graphite (as matrix) and paraffin (as PCM) was developed. The preparation technique used is the dispersion of the graphite powder in melted paraffin, followed by uniaxial cold pressing.

II. EXPERIMENTAL

A. Materials

Paraffin with melting temperature of 53.37 °C and latent heat of 190 J/g was used as thermal energy storage material.



Physical property parameters of paraffin are showing in Fig. 1. The graphite which is not subjected to any chemical or thermal treatment was used as adsorbent in this study. Graphite size distribution curve is shown in Fig. 2 and its size distribution parameter is listed in Table 1.

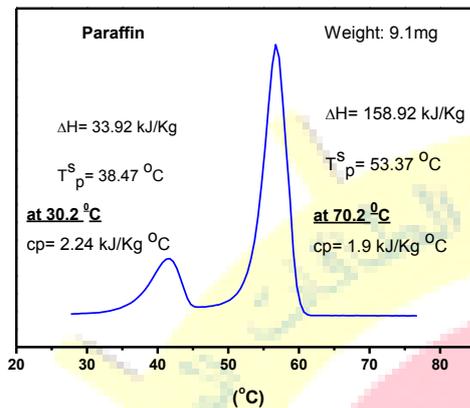


Fig. 1 DSC analysis data for investigated paraffin

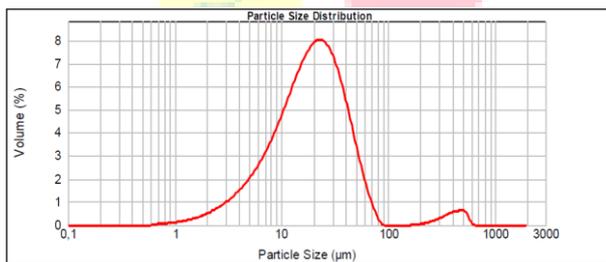


Fig. 2 Size distribution curve of graphite

TABLE I
SIZE PARAMETERS OF GRAPHITE

Sample	d _(0.1) (μm)	d _(0.5) (μm)	d _(0.9) (μm)	d _{av} (μm)	S/g (m ² /g)
Graphite	5,647	19,100	46,128	32,073	0,519

Annotation: D50 means the corresponding particle size when the cumulative distribution percentage reaches 50%; D10 means the corresponding particle size when the cumulative distribution percentage reaches 10%; D90 means the corresponding particle size when the cumulative distribution percentage reaches 90%; Dav means the average particle size in the cumulative distribution of size; S/g means specific surface on weight basis.

A. Preparation of P/Gr composite

P/Gr composite was prepared by adding graphite to melted paraffin. Then, the temperature was adjusted to 70 °C using a constant temperature bath. In order to determine the highest fraction without leakage, the composite PCMs were prepared at different mass fraction of paraffin (the fraction of 55 % is returned). Fig. 3 represented the final composite after compression.



Fig. 3 Sample of P/Gr composite

B. Characterisation

The morphology and microstructure of Gr and P–Gr composite were observed on a field emission scanning electron Microscopy environmental (ESEM, PHILLIPS ESEM XL 30), with 20 kV of tension acceleration. Phase change properties, melting and freezing temperatures, and latent heats of the paraffin and P/Gr composite were determined by differential scanning calorimeter DSC applied with Mettler Toledo Co instrument during a thermal cycle of heating to cooling, at a heating and freezing rate of 5 °C per min and temperature range from room temperature to 100 °C in air atmosphere. Approximately 9 mg of each sample was used for the DSC study to avoid the possible thermal lag. The extrapolated onset temperature, peak temperature and extrapolated end temperature of the paraffin and P/Gr composite were obtained from the special software of the DSC. The phase change latent heats were determined by numerical integration of the area under the peaks. Temperatures have been registered in general with a minor error of 0.2 °C and latent heats within 10%. The thermal stability of P/Gr composite was investigated by a thermogravimetric analyzer (TGA) from room temperature to 600 °C with a heating rate of 10 °C/min. Thermal conductivity values of paraffin and P/Gr composite were measured at room temperature by using TPS 500 hot disk thermal constants analyzer technique, which was developed by Gustafsson [9]. The main advantages of the hot disk technique include: wide thermal conductivity range, from 0.005 W/(m K) to 500 W/(m K); wide range of materials types, from liquid, gel to solid; easy sample preparation; non-destructive; and more importantly, high accuracy [10].

III. RESULTS AND DISCUSSION

A. Morphology of graphite and P/Gr composite



Fig. 4 (a and b) illustrated ESEM photographs of graphite and P/Gr composite. As can be seen in Fig. 4. a, the graphite has a microstructure with multiple pores. As shown in Fig. 4. b, the paraffin was embedded and dispersed in the porous network of the graphite. The ESEM images of composite PCM show that P/Gr composite was well retained into the pores of graphite. The porous structures of the graphite provided the mechanical strength for the whole composite and prevented the seepage of the melted paraffin.

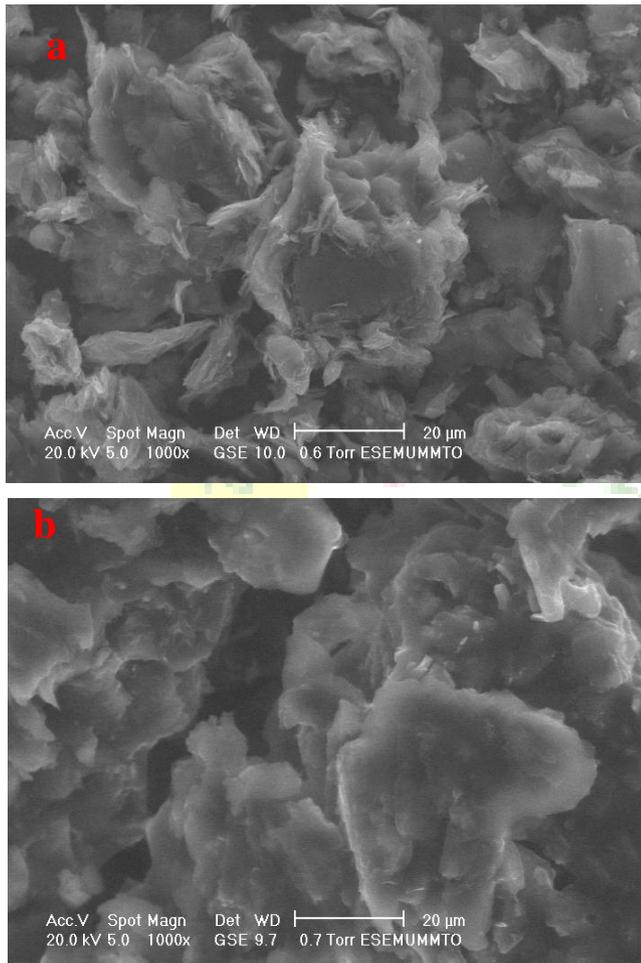


Fig. 4 ESEM images of a) graphite and b) P/Gr composite

B. The thermal properties of the paraffin and P/Gr composite

1) DSC analysis of P and P/Gr composite

The thermal properties of composite MCPs, such as the transition temperature, the melting temperature and the latent heat, are determined by DSC analysis. The experiments were repeated for three times on each sample and the average values were reported recorded in table .II and table. III. Fig. 5 shows the melting-freezing DSC curves of paraffin and P/Gr

composite. The curve of paraffin is used as a reference to assess changes in the thermal properties of the composite PCM. The main peak presents the solid-liquid phase change, and the minor peak corresponds to the solid-solid phase change.

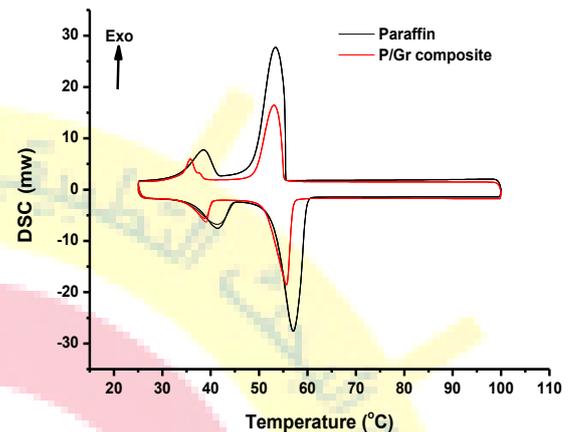


Fig. 5 Curves of paraffin and P/Gr composite

The melting onset temperature extrapolation of the paraffin is 52.59 °C, the peak is 57.03 °C, and the end temperature extrapolation is 59.69 °C. In the P/Gr composite, the onset temperature extrapolated, peak, and end temperature extrapolated during melting process decreased with 1.62 °C, 1.4 °C, and 2.67 °C compared with paraffin alone. Therefore the melting temperatures range of PCM composite decrease with 1.17 °C.

TABLE III
CHARACTERISTICS TEMPERATURES AND ENTHALPIES DURING MELTING PROCESS.

Samples	Melting process characteristic temperature (°C)				ΔH_{pc}^m (kJ/k)
	Onset T_o^m	Peak T_p^m	End T_e^m	Range $T_e^m - T_o^m$	
Paraffin	52.59	57.03	59.69	7.1	213.56
P/Gr composite	50.97	55.63	56.90	5.93	111.31

During freezing process the characteristic temperatures are approximate to that of paraffin.

The total latent heat (for both of solid-liquid and solid-solid phase change) of the P/Gr composite is 110.83 J/g for solidification process and 111.31 J/g for melting process. The latent heat values of the composite PCM are slightly lower than the values of paraffin alone (213.09 J/g, 213.56 J/g). This



can be attributed to the mass of paraffin in the PCM composite. It can remarkably be noted that the prepared P/Gr composite PCM has high latent heat and appropriate phase change temperature. It shows that the prepared composite PCM has good thermal energy storage.

TABLE IIIII
CHARACTERISTICS TEMPERATURES AND ENTHALPIES DURING FREEZING
PROCESS

Samples	Freezing process characteristic temperature (°C)				ΔH_{pc}^f (kJ/kg)
	Onset T_o^f	Peak T_p^f	End T_e^f	Range $T_e^f - T_o^f$	
Paraffin	55.52	53.37	48.97	6.55	213.09
P/Gr composite	55.16	53.05	49.23	5.93	110.83

C. Thermal conductivity measurement

Thermal conductivity is an important property [10] in energy storage applications. In this study, the thermal conductivity of the composite PCM was determined by the hot disk method at room temperature (25 °C) using a conductivity meter (500 Hot Disk TPS). The analysis method is based on Hot Disk theory Transient plane source (TPS) [11]. A probe consisting of a double spiral in nickel serigraphed on an insulating support kapton was positioned between two samples of the same dimensions. A level of power was applied to the probe, and the elevation of temperature was determined from the variation of the resistance of the nickel. The cubic test pieces of the sample were formed by compaction, in order to minimize the porosity of the system. The diameter of the probe used for the tests was 20 ° C was 3.200 mm (C5465 Reference).

Table. IV summarized the experimental results of conductivity for paraffin and P/Gr composite. Thermal conductivity of paraffin at the room temperature was measured as $0.1716 \text{ Wm}^{-1} \text{ K}^{-1}$. In order to improve thermal conductivity of paraffin, the graphite was added to the melting paraffin in mass fraction of % and the thermal conductivity was measured to be $0.472 \text{ Wm}^{-1} \text{ K}^{-1}$. It can be seen that the thermal conductivity of paraffin was ameliorated significantly with 7.5 higher than that of paraffin alone.

TABLE IVV

Experimental thermal conductivities of paraffin and P/Gr composite

Sample	Conductivity (W / m K)
Paraffin	0.1716
P/Gr composite	1,3039

A. Thermal stability of paraffin and P/Gr composite

The TGA and DTG curves of the paraffin and P/Gr composite are shown in Figs. 6. Table IV presented the charred residue amount at 550 °C and the temperatures (T1) and (T2) of onset and maximum weight loss, respectively. The TG curves of the paraffin and P/Gr composite include one step occurs at the temperature between 150 and 300 °C which corresponding to the thermal degradation of the paraffin molecular chains. In addition, as can be seen from TG curves, the P/Gr composite was not degraded or showed almost no weight loss at lower temperature than 100 °C. This result means that the composite PCMs have good thermal stability in their working temperature range.

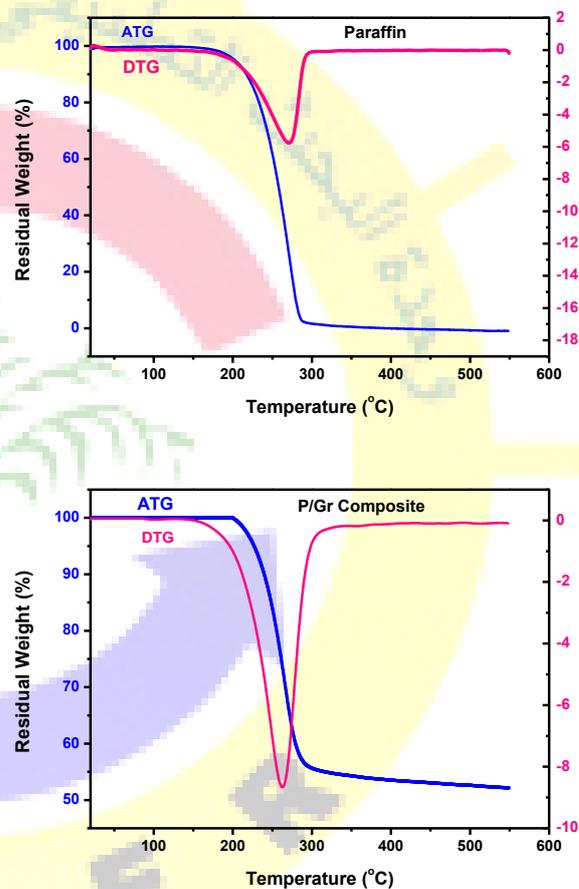


Fig. 6 Curves ATG and DTG of paraffin and P/Gr composite

IV. CONCLUSIONS

A composite PCM was prepared within the graphite (Gr) as the support for PCM. The optimized percentage paraffin confined in the Gr was found as 55 wt%. The P/Gr composite PCM was characterized by ESEM. The microstructure analysis proved that the paraffin was dispersed in the porous network of Gr. It was concluded that the melting and freezing temperatures and latent heats of composites PCM were



acceptable determined using DSC analysis. Thermal conductivity of the composite PCM was increased by addition for graphite. TG analysis revealed that the P/Gr composite PCMs had good thermal stability above their working temperature ranges. Based on all results, it was concluded that the P/Gr has demonstrated the ability to tailor PCMs with acceptable thermal properties.

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