



# Thermal characterization of phase change materials based on linear low-density polyethylene, paraffin wax and expanded graphite



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## ABSTRACT

Thermal characterization of Phase Change Materials (PCMs) based on linear low-density polyethylene (LLDPE), paraffin wax (W) and expanded graphite (EG) is reported in this paper. Investigated PCMs showed high potential for application in energy storage systems.

The latent heat,  $L_m$ , sensible heat  $Q_{sens}$ , and the ability of the prepared PCMs to store and release thermal energy were investigated using specific home-made equipment based on the transient guarded hot plane method (TGHPT). The sensible heat of PCM containing 40 wt.% of paraffin wax was investigated in the temperature range 25–35 °C, they exhibited a drop in  $Q_{sens}$  from 31 to 24 J/g depending on the concentration of EG. A similar decrease in sensible heat with increased loading of EG was observed for PCMs containing 50 wt.% of EG.

The storage and release of thermal energy during phase change which is associated with the latent heat of the materials were investigated within the temperature range 20–50 °C. PCMs containing 40 wt.% of paraffin wax exhibited latent heat of 36 J/g, whereas the latent heat of PCMs containing 50 wt.% of paraffin wax was 49 J/g. The addition of EG decreased the time needed to melt and solidify PCMs due to increase in thermal conductivity of PCMs with increase in EG content. This behavior was confirmed by the thermal conductivity measurements, where thermal conductivity increased from 0.252 for sample without EG to 1.329 W/m × °C for PCM containing 15 wt.% of EG.

The reproducibility of storage and release of thermal energy by PCMs was demonstrated by subjecting them to repeated heating and cooling cycles (over 150 cycles).

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## 1. Introduction

Thermal-energy storage systems are crucial for reducing dependency on fossil fuels and minimizing CO<sub>2</sub> emissions. The Kyoto Summit secured a commitment from most countries to establish a global program for carbon dioxide (CO<sub>2</sub>) emissions reduction. According to the World Business Council for Sustainable Development, buildings account for up to 40% of global energy use [1]. Therefore, there is a critical need to reduce CO<sub>2</sub> emissions from buildings. A portion of this reduction can be achieved by using building materials that are energy efficient.

Thermal energy storage can be accomplished either by using sensible heat storage or latent heat storage components. Sensible heat storage components have been used by builders for centuries to store and release thermal energy passively, but a much larger volume of material is required to store the same amount of energy in comparison to latent heat storage systems [2]. Latent heat storage is more attractive than sensible heat storage because of its high storage density with smaller temperature fluctuations [3,4].

PCMs, can undergo phase changes (usually solid to liquid changes) at relatively low temperatures while absorbing or releasing high amounts of energy [5]. During the last four decades, many PCMs, with different phase transitions (e.g., solid–liquid, solid–solid) and a wide range of transition temperatures, have been designed and studied extensively [6,7]. A suitable phase change temperature and a large melting enthalpy are two crucial requirements of PCMs.

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Various inorganic and organic substances have been used in the production of PCMs, the most common of which include various inorganic salts (e.g., polyhydric alcohols) and their eutectics, fatty acids, and *n*-alkanes. Inorganic salts (salts hydrates) were used as PCMs in the past, but they faced issues such as, such as super-cooling, corrosion, relatively high volume and chemical instability, hence making them an unfavorable choice. Whereas, metallic compounds have limited use as PCMs for commercial purpose due to their weight and cost [8].

The most promising materials used as PCMs for low-temperature applications (below 100 °C) are paraffin waxes due to their high latent heat of fusion, negligible super-cooling, low vapor pressure in the melt and chemical inertness [9]. As for practical applications, another important issue has to be taken into account, namely a suppression of flow (leaching), an issue faced by PCMs that undergo solid–liquid transition. There are a many strategies for avoiding this problem. First, paraffin waxes are kept in tanks of various shapes and volume that are incorporated into the building according to the specific needs. Second, porous materials can be impregnated by paraffin waxes; in this case capillary forces suppress leaching. Third, paraffin waxes can be encapsulated within a polymeric shell to form microcapsules. These microcapsules are frequently used in the textile industry (an impregnation of the surface of fabrics or direct incorporation of microcapsules into fibers during spinning) and in buildings. The most widely known example of such materials is MICROCONAL from BASF, which is frequently blended with plaster or concrete for designing of heat protective blocks. Lastly, paraffin waxes can be directly blended with polymeric matrices to avoid leaching and to retain a compact shape even after the paraffin wax has melted. Presently, paraffin waxes blended with appropriate polymers appear to be the best candidates for preparation of smart PCMs for various applications, such as thermal storage of solar energy, thermal protection of electronic devices, thermal protection of food and medical goods, passive storage in bioclimatic buildings, use of off-peak rates and reduction of installed power and thermal comfort in vehicles [10–12].

Polyethylenes appear to be the most suitable polymer for blending with paraffin waxes due to their chemical and structural similarities with paraffin waxes. Krupa et al. [13] investigated low-density polyethylene (LDPE) blends with soft and hard paraffin waxes. The blends were able to absorb large amounts of heat energy due to melting of paraffin wax, whereas the LDPE matrix kept the material in a compact shape on the macroscopic level. The importance of the structural similarity of was reported in a study that dealt with PCMs based on isotactic polypropylene (PP) blended with soft and hard Fischer-Tropsch paraffin [14]. The PP matrix held the material in a compact shape during the transition from solid to liquid. However, a much lower content of paraffin wax was incorporated due to strong phase separation of components, which is caused by the differing structure of crystallites. In the case of polyethylene, the zigzag structure of crystallites is much more favorable for mutual compatibility with paraffin waxes than the helical crystalline structure of polypropylene despite the fact that paraffin waxes, polyethylene and polypropylene are practically chemically identical. In addition, paraffin waxes can co-crystallize with polyethylene as discussed in a prior publication by the authors. The results obtained from solution crystallization confirmed a strong indication of co-crystallization in the case of LLDPE on one hand, and practically no miscibility in the crystalline regions of LDPE and oxidized Fischer-Tropsch paraffin wax on the other [15].

The fact that chemical and structural differences lead to strong phase separation of components was demonstrated in the work focused on PCM created from paraffin wax and polyamide [16].

Other polymer matrices, such as styrene–ethylene–butylene–

styrene (SEBS), styrene–isoprene–styrene (SIS), and styrene–ethylene–propylene–styrene (SEPS), have been examined [17,18]. Mentioned studies described a shape-stabilized PCM with a melting temperature of 56–58 °C. The results showed that the composites can retain their shape even when paraffin wax is in the liquid state, and no paraffin wax leakage was observed during thermal performance testing. Thermosetting resins (epoxy resin cured with amino-based hardener) were also tested for applications where blending with paraffin waxes at room temperature is required, so that low transition-temperature PCMs can remain in the solid phase during composite processing [19].

A Major drawback of polymer/paraffin wax PCM's is their low thermal conductivity. The thermal conductivity of paraffin waxes is approximately 0.2 W/m × °C, and the thermal conductivity of polymers varies from 0.15 W/m × °C for amorphous polymers such as polystyrene or polymethylmethacrylate, to the 0.5 W/m × °C for highly crystalline polymers such as high density polyethylene [20]. Thermal conductivity of polymers can be improved by blending them with inorganic materials [21,22], such as graphite and exfoliated graphite [23–25], metals [26–28], boron nitride [29], various metal-coated fillers [30] as well as carbon nanotubes [31].

EG is particularly useful for tailoring the thermal conductivity of materials (including PCMs) due to its inherently high thermal conductivity, favorable shape and high aspect ratio [30–34]. Recently, Trigui et al. [34] developed a new method and constructed a reliable device for testing the long-term performance of PCMs in large sized samples. They described the first store built with the composite material developed to improve different properties of PCM such as latent heat, storage and release energy, heat capacity and thermal conductivity. This unique method for thermal investigation of PCMs was used for resin epoxy/spherical paraffin wax [35] as well as for LDPE/paraffin wax composites at different concentrations of paraffin wax [36]. The objective of these investigations was to study the thermal conductivity of these PCMs and the amount of energy exchanged during the variation of thermodynamic state of the samples when the boundary temperatures vary [37]. Thermo-physical properties obtained through this method for PCMs are crucial in studying and simulating their behavior.

In this study, the thermal properties and the storage and release of energy in PCMs based on LLDPE, W (melting point approximately 42 °C) and EG are reported. The storage and the release of heat energy were determined using the Transient Guarded Hot Plate Technique (TGHT), which allows investigation of the thermal properties of large samples [36]. In comparison with results obtained from Differential Scanning Calorimetry (DSC) good reproducibility of the measurements was observed. .

Since prepared PCMs have a melting point of around 42 °C, they can be feasible for outdoor use in regions where heat accumulates at a higher temperature [38]. Furthermore, it can be useful for heat protection of solar collectors, batteries etc ... [39].

## 2. Experimental

### 2.1. Materials

Linear low density of polyethylene (MFI = 1 g/10 min, QAPCO, Qatar), paraffin wax (Grade RT42, Rubitherm Technologies, Germany) and expanded graphite (GFG200, SGL Carbon, Germany) having an average size of 200 μm were used for the PCMs preparation.

### 2.2. Composite preparation

LLDPE powder was mixed with paraffin wax using a Brabender instrument (Plasticorder PLE 331, Germany) at 140 °C.

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