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Positive influence of expanded graphite on the physical behavior of phase change materials based on linear low-density polyethylene and paraffin wax

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ABSTRACT

The preparation of Phase Change Materials (PCM) with improved thermal conductivity, based on linear low-density polyethylene (LLDPE), paraffin wax with a melting point of approximately 42 °C and expanded graphite as well as the investigation of their physical behavior is reported in this paper.

The absorption of heat energy is realized by phase transitions in the wax structure, i.e., solid–solid transitions between different crystalline structures of wax and, primarily, by a solid–liquid transition. An estimate of the total heat energy that can be reversibly absorbed or released by the designed materials was determined using Differential Scanning Calorimetry (DSC).

The improvement in the thermal conductivity of the polymeric materials was obtained by incorporating expanded graphite into the blends. Expanded graphite has a reinforcing effect on the PCM, resulting in an increase in the toughness (Young's modulus) Significant amounts of wax are leached from the samples, and a higher wax content also results in more wax leakage. Graphite significantly reduces the wax loss from the samples.

1. Introduction

In hot countries, such as Qatar, the relatively harsh climate can produce temperatures up to 45 °C during the long summers and as low as 5 °C during the short winters, which leads to large energy consumption, particularly for air conditioning. In general, it has been estimated that buildings consume up to 40% of global energy use [1]. Building designs that consider the local climate and utilize energy from the environment are called bioclimatic buildings. The sun is the most common source of energy that can be used to fulfill the energetic requirements of buildings. For this reason, researchers from academia and industry focus their attention on developing materials that effectively absorb and release thermal energy from the sun to ensure thermal comfort with minimal use of electrical energy.

Materials with this set of properties are called heat absorbers or, more commonly, Phase Change Materials (PCMs). Their primary characteristics are the ability to undergo phase transition (usually from a solid to a liquid) at relatively low temperatures while absorbing or releasing a large amount of energy proportional to their specific enthalpy of melting. PCMs have received growing interest for use in many applications, particularly in the building industry.

Other important applications of PCMs include the photovoltaic industry. Silicon photovoltaics (PV) show a significant power drop when the panel temperature is above 25 °C depending on type of PV cell and the manufacturing technology [2]. The operating temperature reached by PV panels and associated power drop largely depends on the climate of the site. The advisable operating temperature limits for PV range from −40 to 85 °C. However, in hot and arid climates, PV temperatures frequently rise above this

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temperature range [3], which results in temperature-induced power failures. In an attempt to avoid temperature-dependent PV power loss, passive cooling of PVs with paraffin-wax-based solid–liquid PCMs is frequently used [4,5].

Various inorganic and organic compounds can be used as PCMs. The most common are inorganic salts (e.g., polyhydric alcohols), fatty acids, and paraffin waxes [6]. Paraffin waxes are the most promising PCMs due to their favorable characteristics, such as high latent heat of fusion, negligible super-cooling, stability, availability, and relatively low price [7]. The melting temperature of these paraffin waxes ranges from 30 to 90 °C, depending on the number of carbons in the wax chains. The specific melting enthalpies of these waxes are between 180 and 230 $kJ\,kg^{-1}$, resulting in an excellent energy storage density for paraffin waxes [8,9]. After melting, paraffin waxes have a tendency to leak from the system. To suppress this effect, paraffin waxes are enclosed in tanks or containers during heating to suppress leakage [6,7]. These waxes can also be fixed in stable forms via encapsulation in a polymeric shell (using emulsion polymerization, for example) or by blending with certain polymers [10–16]. After blending, the polymeric matrices fix the paraffin wax in a compact form to suppress leakage after the wax melts.

Polyethylene is the most frequently used polymer for blending with paraffin waxes due to their chemical and structural similarities [17–19], which leads to the incorporation of a large amount of wax in polyethylene matrix without significant wax leakage due to incompatibility between the components.

Another important feature of PCMs involves an improvement in heat transport within the materials, which is associated with low thermal conductivity of the polymers and paraffin waxes. These values range from 0.15 $W\,m^{-1}\,K^{-1}$ for amorphous polymers, such as polystyrene or polymethylmethacrylate (and also paraffin waxes), to 0.5 $W\,m^{-1}\,K^{-1}$ for highly crystalline polymers, such as high-density polyethylene [20].

Heat transport is usually enhanced in PCMs by incorporating thermally conductive fillers. However, problems arise when attempting to minimize the filler content and maximize the paraffin content to optimize the functionality and maintain a compact form, even after melting, which prevents the wax from leakage. Thus, these three components must be optimized. Exfoliated graphite is prospective filler for improvement of the thermal conductivity of polymeric composites [21,22]. Recently, Kalaitzidou et al. [22] reported a significant increase in the thermal conductivity of polypropylene-based composites, when filled with a small amount of exfoliated graphite. Xie et al. [23] demonstrated that graphene nanosheets are more effective conductivity enhancers than carbon nanotubes.

Expanded graphite (EG) has been extensively used over the past few years to improve thermal conductivity of PCMs, particularly paraffin-based PCMs. Zhao et al. [24], Zhong et al. [25], Sari and Karaipekli [26] and Xia et al. [27] prepared EG/paraffin composites with mass fractions of EG varying from 0 to 10 wt.%, which resulted in improvements in the thermal conductivities of the materials. Similarly, Zhang et al. [28] studied paraffin with 8 wt.% EG and found a significant improvement in the thermal conductivity.

A composite composed of paraffin, styrene-butadiene-styrene (SBS) triblock copolymer and exfoliated graphite was prepared by Xiao et al. [29]. The composite exhibited high thermal conductivity and nearly 80% of the latent heat of fusion per unit mass of paraffin. Composites composed of paraffin mixed with graphite nanoparticles were prepared, and the microstructures and thermal properties of the materials were examined by Li [30]. The thermal conductivity of the material containing 10% graphite nanoparticles was found to be 0.9362 W/m K. The cellular graphite/paraffin composites prepared by Vitorino et al. [31] experienced remarkable increases in their thermal conductivities in the range from 5 to 8 W/m K.

The melting point of paraffin wax used in this study is around 42 °C. For instance, it is appropriate in outdoor building applications in the regions where heat accumulates at a higher temperature [32] (Middle East countries, Africa). Furthermore, it is highly useful for heat protection of solar collectors [33].

2. Experimental

Linear low-density polyethylene powder (MFI = 1 g/10 min, QAPCO, Qatar) was mixed with paraffin wax (grade RT42, Rubitherm, Germany) in selected ratios.

The LLDPE/wax ratios were = 70/30, 60/40, and 50/50 w/w. Wax contents greater than 50 wt.% led to significant phase separation of the blends and loss of their mechanical integrities. To improve the thermal conductivities of the materials, expanded graphite (GFG200, SGL Carbon, Germany) was used. The average size of the graphite platelets is approximately 200 μm.

Dog-bone specimens with working areas of 30 × 4 × 1 mm were cut from the slabs. The mechanical properties were measured at

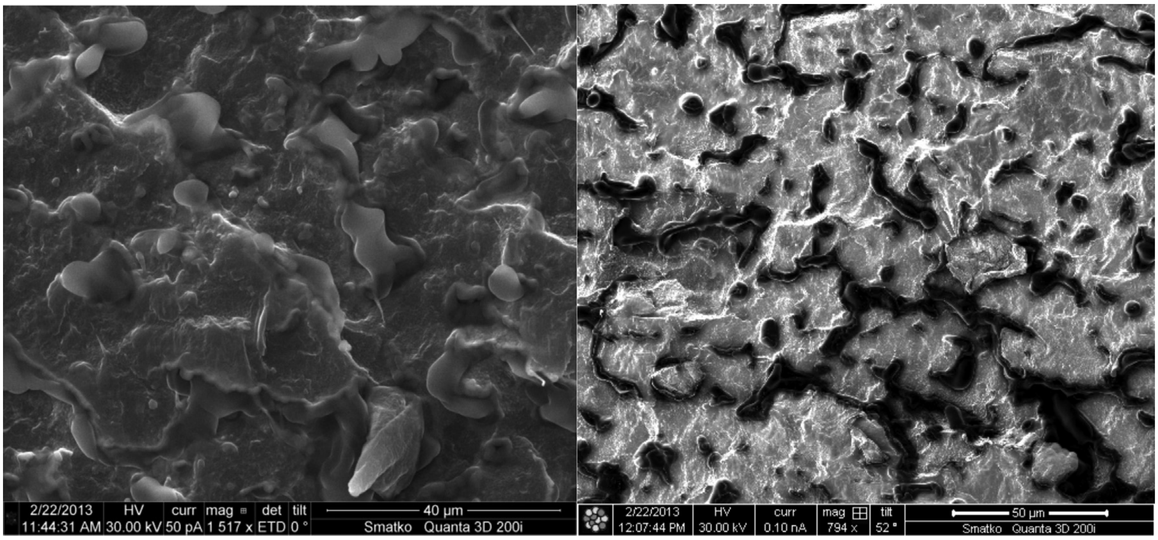

Fig. 1. SEM micrograph of the LLDPE/wax = 50/50 w/w blend in electron beam (left) and ion beam mode (right).