



Preparation, stability and mechanical property of shape-stabilized phase change materials



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ARTICLE INFO

Article history:

Received 31 December 2013

Received in revised form 16 March 2014

Accepted 19 March 2014

Keywords:

Shape-stabilized phase change materials (SSPCM)

Cyclic stability

Mechanical property

ABSTRACT

A series of shape-stabilized phase change materials (SSPCMs) were prepared by melt blending with paraffin and polymer alloys. The polymer alloys used in this work are high-density polyethylene (HDPE) with various elastomers including ethylene-propylene-diene copolymer (EPDM), three styrene-butadiene-styrene block copolymers (SBS) with different S/B ratios and configurations and styrene-isoprene-styrene block copolymer (SIS). These alloys were used as matrices to encapsulate and support paraffin. Their thermal properties, cyclic stability and mechanical properties were investigated by differential scanning calorimetry (DSC), cyclic stability test and tensile test respectively. The results indicate that these alloys are able to encapsulate paraffin well with latent heats of 80–100 J/g and mass loss of 8–10% after 100 cycles. Crosslinking of the matrices was carried out and its effects on tensile property and cyclic stability were explored. Proper crosslinking can increase the tensile strength without sacrificing the cyclic stability.

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1. Introduction

Phase change materials (PCMs) can absorb heat when they change from solid to liquid and vice versa. With high energy density and little temperature fluctuation, PCMs are very useful in thermal management applications [1]. The energy storage ability of PCM can increase the energy utilization efficiency and smooth the gap between the energy supply and demand. Therefore, PCMs have been applied in many fields such as energy-saving buildings, waste heat recovery, storage and reuse, and thermal management of electronic devices [2–5]. There are organic and inorganic PCMs, and organic PCMs are widely used for its stability, non-corrosiveness, long lifetime and small super cooling.

Paraffin, an organic PCM, is the most widely studied because of its high density of thermal storage, wide phase transition temperature range as well as low cost. In order to prevent the flow and leakage of liquid paraffin, it is usually encapsulated in applications. One of the effective ways is to encapsulate paraffin in a polymer matrix to form a shape-stabilized phase change material (SSPCM). Since Inaba and Tu [6] first utilized high-density polyethylene (HDPE) as a matrix, more and more matrices have been reported such as polyolefins [6–24], elastomers [13,24–28] and polymer alloys [29–31]. Various capsulation abilities of these

SSPCMs have been reported with the phase change temperatures in a range of 313–333 K and the latent heat of 100–200 J/g. In addition, thermal conductivity enhancement and flame retardance of SSPCMs have been investigated.

The applications of SSPCM in energy-saving buildings have been reported in numerous literatures. Zhang et al. [13,29,32] and Zhou et al. [33–36] argued that the appropriate phase change temperatures of SSPCMs should be limited at ~293 K. However, most of the paraffins used today have the phase change temperatures in a range of 313–333 K, which does not work for energy-saving buildings. In addition, most studies have been focused on thermo-physical properties (phase change temperature and enthalpy), while much less reports have concerned thermal conductivity, flame retardance, especially cyclic stability and mechanical property of SSPCMs although they are of crucial importance for practical applications. For example, Alken et al. [23] demonstrated that the melting and crystallization latent heats of the paraffin/PP composites declined significantly after 3000 thermal cycles.

In this work, SSPCMs for energy-saving buildings were prepared with the paraffin (melting temperature of 293 K) encapsulated with HDPE and elastomers (EPDM, SBS and SIS). HDPE served as a strong supporting material; while elastomers exhibit good absorption and encapsulation to paraffin. In addition to general thermo-physical properties, the mechanical property and cyclic stability were investigated. SSPCMs prepared in this work show high latent heat, excellent strength and high stability.

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Table 1
Compositions of SSPCMs.

Paraffin (wt%)		60	60	60	60	60
Matrix (wt%)	HDPE	0	8	10	15	20
	Elastomers (EPDM/SBS/SIS)	40	32	30	25	20

2. Materials and methods

2.1. Materials

Paraffin, with a phase change temperature of 293 K and latent heat of 183 J/g, was from Nanyang Wax Fine Chemical Plant (Henan, China). High-density polyethylene (HDPE) was from Beijing Eastern petrochemical Co. Ltd. (Beijing, China). Ethylene-propylene-diene copolymer (EPDM, 3745P) was from DOW Chemical. Styrene-butadiene-styrene block copolymer (SBS) #1 (linear copolymer, S/B=4/6) and styrene-isoprene-styrene block copolymer (SIS) were from Baling Petroleum Chemical Industry Co. Ltd. SBS #2 (linear copolymer, S/B=3/7) and #3 (star copolymer, S/B=3:7) were from Dushanzi Petrochemical Co. Ltd. Dicumyl peroxide (DCP), with melting point of 41 °C and density of 1.082 kg/m³, was from Sinopharm Chemical Reagent Co. Ltd.

2.2. Preparation of SSPCMs

Paraffin was mixed with one of the elastomers (EPDM, SBS or SIS) and kept at room temperature for 12 h, and then HDPE was added and mixed. The mixtures were melt-blended in a torque rheometer (HAAKE PolyLab QC) at different temperatures and screw rotating speeds for specific times. The compositions of the SSPCMs are listed in Table 1. The SSPCM samples were then hot pressed to plates for further characterization. In order to improve the mechanical property of SSPCMs, various amounts of DCP were added and the compositions are listed in Table 2.

2.3. Characterization

The phase change temperature and enthalpy of SSPCMs were measured by differential scanning calorimeter (DSC) (Q100, TA Instrument, USA). About 5 mg samples were tested at temperatures ranging from 233 to 333 K with the heating/cooling rate of 10 K/min in a nitrogen atmosphere.

The maximal absorption amount of different elastomers to paraffin was tested at room temperature. One gram elastomer pellets were immersed in liquid paraffin for different times. The surface was dried by filter paper. The mass was then weighed and the absorption ratio was calculated as follows:

$$\text{Absorption ratio} = \frac{\text{mass after absorption} - 1.00}{\text{mass after absorption}}$$

To test the cyclic stability of the SSPCMs, SSPCM plates of 50 mm × 30 mm × 3 mm were weighed first, heated at 333 K in an oven for 1 h and then cooled down in a refrigerator of 273 K for 1 h to finish a cycle. The plates were weighed every 5 cycles to calculate the paraffin loss. Each sample was tested twice and the mean value was reported.

Table 2
Compositions of crosslinked SSPCMs.

SSPCM	DCP (phr)
Paraffin/EPDM/HDPE = 60/30/10	0.2–3.2
Paraffin/#1SBS/HDPE = 60/30/10	0.4–0.8
Paraffin/#2SBS/HDPE = 60/30/10	0.2–0.4
Paraffin/#3SBS/HDPE = 60/30/10	0.2–0.6

The samples were prepared in HAAKE at 413 K and 60 r/min.

The mechanical properties of SSPCMs and crosslinked SSPCMs (63 mm × 13 mm × 4 mm) were tested by an universal testing machine (Gotech, Taiwan). Each test was repeated at least three times with different samples and the mean value was reported.

3. Results and discussion

3.1. Absorption ability of elastomers

Fig. 1 shows paraffin absorption ratio and rate of the elastomers (EPDM, SBS and SIS). All of the five elastomers have good absorption ability, although they have different absorption rates and final absorption ratios. SBS and SIS absorb paraffin much faster than EPDM. All three SBSs showed similar absorption rates and reached the maximal paraffin absorption ratio in two hours. SIS and EPDM reached the maximal in 8 h or longer. The maximal absorption ratios of these five elastomers showed an order of SIS > #2SBS ≈ #3SBS > EPDM > #1SBS. SIS has a highest saturated absorption ratio of 96%, but it is paste-like finally and cannot maintain its shape. The maximal absorption ratio for EPDM is 86%. The saturated absorption ratio of #1SBS is 72%. SBS#2 and SBS#3 are with slightly higher ratios of 80%. #1SBS and #2SBS are linear copolymers with different S/B levels. #2SBS and #3SBS have same S/B levels but the later is a star copolymer. This result indicates that the maximal absorption ratio is related most likely to the content of butadiene units other than to the chain configuration. Unlike SIS, EPDM and the SBSs are able to keep the shape well with the maximal absorption of paraffin.

3.2. Preparation and crosslinking

Elastomer/HDPE alloys were used in this paper as the matrices, in which elastomers can absorb large amount of paraffin and HDPE maintains a good processing performance and mechanical property. SSPCMs, with various mass ratios of elastomers to HDPE, were prepared by HAAKE. EPDM/HDPE and SBS/HDPE alloys, with the mass ratio of elastomer to HDPE 3:1, exhibited good encapsulation as well as excellent mechanical strength. SSPCMs with higher mass ratio, i.e., with more elastomers, were too soft to keep the shape at the room temperature. Less elastomer caused the migration of paraffin from SSPCM. SSPCMs made with SIS/HDPE alloy with different mass ratios were either soft or with a poor encapsulation of paraffin. Paraffin diffused outwards when it is poorly encapsulated, which caused an oily surface. Therefore, EPDM/HDPE and SBS/HDPE with a mass ratio of 3:1 were used in the following experiments.

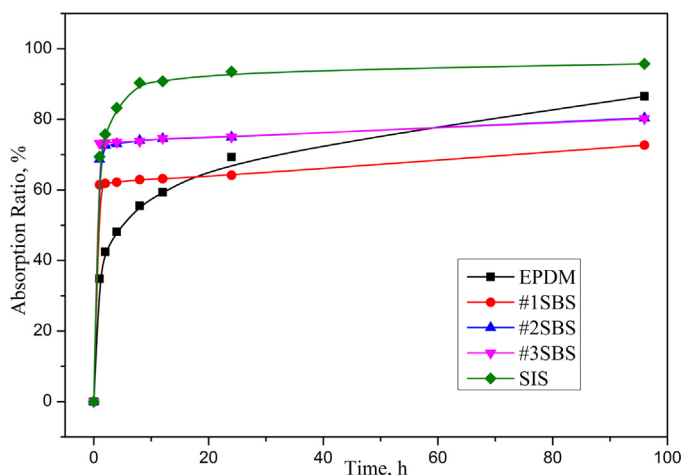


Fig. 1. The absorption ratios of SIS, SBSs and EPDM.

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