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Review

Synthesis and thermal properties of cross-linked poly(acrylonitrile-co-itaconate)/polyethylene glycol as novel form-stable change material

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ABSTRACT

In this paper, a novel form-stable phase change materials (FSPCMs) of cross-linked poly(acrylonitrile-co-itaconate) (P(AN-co-IA))/polyethylene glycol (PEG) blend was prepared successfully. And it was systematically characterized by Fourier transform infrared (FT-IR), Raman, differential scanning calorimetry (DSC), X-ray diffraction (XRD), polarized optical microscopy (POM), Scanning electron microscope (SEM), Thermogravimetric (TG) and thermal conductivity measurements. FTIR analysis indicated that the cross-linked network had been synthesized. The results of DSC analysis showed that the cross-linked FSPCMs had good thermal reliability and heat storage durability after 1000 thermal cycles. The phase change temperature of the cross-linked FSPCMs was in the range of 23–53 °C and the phase change enthalpy was around 118.5 J/g, respectively. The XRD results indicated that PEG units in the cross-linked FSPCMs had the same crystal structure as the pure PEG. And it was observed that the PEG was dispersed into the network of the solid cross-linked P(AN-co-IA) by investigation of the structure of the FSPCMs using a SEM. TGA results confirmed that the initial degradation temperature of the cross-linked FSPCMs was 249 °C, which proves that cross-linked FSPCMs appeared good thermal stability. These results indicated that cross-linked FSPCMs could be considered as latent heat storage material for thermal energy storage applications.

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1. Introduction

The energy storage and reuse have attracted considerable attention in recent years because of the energy crisis, the high cost of fossil fuels, the environmental impacts as well as the low price [1,2]. Owing to its ability to provide a high storage density at nearly constant temperature, latent heat thermal energy storage (LHTES) using phase change materials (PCMs) is considered to be the most effective and promising technique for solar energy heating and cooling applications [3]. In recent years, PCMs have been widely employed in various fields including solar energy system, building energy conservation, thereto-regulated fibers and so on [4–8].

Among the PCMs investigated, PEG is taken as a promising PCM because of its nontoxicity, relatively large fusion heat, congruent melting behavior, resistance to corrosion and ease of chemical modification [9–11]. However, the melted PEG has to be kept in a closed tank or container to prevent leaching during the phase transition, which not only increases production costs, but also reduceses the heat transfer efficiency. Physical entanglement or chemically cross-linking of PEG with other substances, it may maintain the original solid state during phase transition of PEG, namely forms the so-called FSPCMs, which can effectively solve the above problems.

FSPCMs can be prepared by encapsulation of PCM into inorganic materials with porous lamellar structure or polymers with high melting point and so on [12–14]. Although inorganic materials can ensure the same macroscopic shape before and after the phase transition, the PCM is easy to loss by migrating to the surface after repeated use. Therefore, a considerable amount of research has been carried out on the preparation of suitable polymer-based FSPCMs with crosslinking structure. There are two basic approaches: chemical and physical approach. In the chemical approach, the PCM is bound onto supporting polymeric materials by chemical grafting and cross-linking copolymerization [9,15]. Li et al. [16] synthesized a novel solid–solid phase change material (SSPCM) via the two-step condensation reaction of polyethylene glycol (PEG10000) with pentaerythritol (PE) and 4,4′-diphenylmethane diisocyanate (MDI). The results showed that the novel polymeric SSPCMs possesses excellent crystal properties and high phase change enthalpy. Xiang et al. [17] synthesized novel environmentally friendly poly(hydroxybutyrate-co-hydroxyvalerate) and poly(ethylene glycol) (PHBV/PEG) copolymer networks as a PCM with PHBV diacrylate (PHBVDA) and polyethylene glycol diacrylate (PEGDA) as macromonomers. PHBV/PEG copolymer network had a higher latent heat enthalpy, which didn't reduce with the components of PHBV increased. Moreover, PHBV/PEG copolymer network still had good thermal stability even at 300 °C. Li et al. [18] synthesized a novel melamine/formaldehyde/polyethylene glycols (MFPEG) SSPCMs with crosslinking structure composed of PEG as energy-storage ingredient and melamine as crosslinking functional reactant. The SSPCMs were synthesized via two steps of amine-aldehyde condensation reaction and aldolization. The maximal latent heat enthalpies of heating and cooling cycle were 109.4 and 103.9 J/g, respectively. In another study, PEG10000/polyglycidyl methacrylate cross-linked copolymer as a novel SSPCM was successfully synthesized [19]. The DSC results indicated that the copolymer imparted balanced and reversible phase change behaviors at the temperature range of 25–60 °C, and it had high latent heat storage capacity of more than 70 J/g. However, there was a remarkable obstacle in the practical application for energy storage with chemical methods of synthesizing SSPCMs due to its complexity. Moreover, owing to the chemical linkage function of the skeleton polymer to the phase change substance, the phase change enthalpy and the phase change temperature of these phase changed substance composites obviously reduces.

The other approach is the physical approach in which many researchers aim to construct cross-linked microstructure in the matrix of polymer-based FSPCMs [16,18]. Here, the cross-linked microstructure plays an important role in overcoming the leakage of phase change materials. It is mainly because molecular chain of phase change working substance is confined in the micro-deformation of cross-linked microstructure, so it migrate into surface of matrix hardly. Chen et al. [20] prepared a series of cross-linked high-density polyethylene (HDPE) FSPCM with different gel contents. The result showed that cross-linking can improve the stability of FSPCM with the formation of the network, which would prevent the paraffin from diffusing to the outside. Moreover, Chen et al. [21] evaluated the miscibility of the paraffin in the three types of polyethylene. The atomic force microscopy (AFM) results verified the partial miscibility of paraffin in polyethylene, and concluded that paraffin is less miscible in HDPE than in LDPE or LLDPE. In the next paper, to evaluate the leakage behavior of paraffin in PE for future FSPCMs, Chen et al. [22] explored morphological structures with paraffin leakage behavior in polyethylene/paraffin binary composites. The paraffin leakage rate from HDPE was considerably slower than blends produced with LDPE or LLDPE for leakage test.

As one kind of important engineering polymer, Polyacrylonitrile (PAN) possess solvent resistance, chemical stability, thermal and mechanical stability, high tensile strength and good insect resistance [23,24]. It has attracted more and more researchers to explore the PAN as PCM packaging material in recent years [25–28]. Here, we prepare a novel cross-linked network structure of acrylonitrile copolymer P(AN-co-IA) with itaconic acid (IA) as comonomer. When compared this method with the others in literature, it is simple, easy and does not need long time, high temperature and special catalyst. Thereafter, a novel cross-linked FSPCMs with PEG as work materials and cross-linked P(AN-co-IA) as cladding layer and supporting material were prepared. The cross-linked P(AN-co-IA) not only provided a good supporting skeleton to the whole composites, but also prevented the leakage of solid–liquid PCM during phase change process. The structure, thermal properties and thermal reliability of cross-linked P(AN-co-IA)/PEG were investigated by FTIR, Raman, DSC, XRD, POM, SEM, TGA and thermal conductivity testing instrument. The results showed that the cross-linked FSPCMs possess excellent phase change properties. It is easy to achieve desired shapes and likely to be produced in large scale.

2. Experimental

2.1. Experimental materials

Acrylonitrile (AR) was purchased from Tianjin Fuchen chemical reagents factory. Itaconate (AR) was purchased from aladdin reagent Co., Ltd. P(AN-co-IA) (33.4 wt% IA) was synthesized by laboratory. MDI (AR) was purchased from Wanhua Chemical Group Co., Ltd. N,N-dimethylformamide (DMF) (AR) and PEG (Mn = 4000) were purchased from Tianjin Kermel chemical reagent Co., Ltd.

2.2. Preparation of cross-linked P(AN-co-IA)/PEG FSPCMs

Firstly, 1.0 g P(AN-co-IA) (0.00513 mol —COOH), 0.65 g (0.0026 mol) MDI (molar ratio of —COOH to MDI = 2:1) and 70.0 g DMF were dissolved in a three-neck round-bottom flask with an overhead stirrer. The reaction mixture was stirred at 65 °C for 4 h. Then, 0.25 g (0.001 mol) MDI was dissolved in the reaction mixture, and slowly heated to 90 °C and left to stir for

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