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## Preparation and properties of polyethylene glycol based semi-interpenetrating polymer network as novel form-stable phase change materials for thermal energy storage



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#### ABSTRACT

A semi-interpenetrating polymer network (semi-IPN) of polyethylene glycol (PEG)/poly(polyethylene glycol diacrylate) (PPGD) composites as novel form-stable PCMs was successfully prepared via in situ polymerization. The mass percentage of PEG reached 70 wt% without any leakage above the melting point of PEG. The prepared form-stable PCMs were investigated by scanning election microscopy (SEM), Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), polarization optical microscopy (POM), differential scanning calorimetry (DSC) and thermogravimetric analysis (TG), respectively. SEM and FTIR results show that the PEG is uniformly dispersed into the PPGD network and there are only physical interactions between PEG and PPGD. POM images and XRD patterns reveal that the crystalline structure of PEG is not affected by PPGD network and the crystal size of PEG in form-stable PCMs decreases due to the restriction of PPGD network. DSC analysis results present that the melting and freezing temperatures and the latent heats of form-stable PCM were measured as 58.62 and 37.45 °C and 117.41 and 115.17 J/g, respectively. Thermal cycling test and TG analysis confirm that the prepared form-stable PCMs exhibit good thermal reliability and stability. The prepared form-stable PCMs have great potential application in the area of solar energy storage.

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#### 1. Introduction

In recent years, latent heat thermal energy storage (LHTES) using phase change materials (PCMs) has been drawing a great attention with the shortage in traditional fossil energy and the increase in greenhouse gas emissions [1,2]. PCMs are commonly acknowledged as the efficient thermal energy storage materials, owing to the advantages of high density of energy storage and small temperature fluctuation during the phase change process [3,4]. Therefore, PCMs can be broadly applied in the fields of solar energy utilization [5], heat transfer fluid [6], temperature regulating greenhouses [6], building materials and temperature-regulating textiles [7,8].

PCMs commonly fall into two major categories: solid-solid PCMs and solid-liquid PCMs. The merit of solid-solid PCMs is that no container is needed to seal them, due to that there is no gas or liq-

uid generate during the phase transition. Nevertheless, small latent heat restrict their applications [9,10]. A great variety of solid-liquid PCMs including paraffin wax [11], fatty acids [12], fatty alcohol [13] and polyethylene glycol (PEG) [14] have been developed and applied extensively in the field of LHTES, due to their excellent properties such as high latent heat storage capacity, appropriate phase change temperature ranges, little or no super-cooling, non-toxicity, less or no volume change during phase transition, good thermal stability and so on [15]. Nevertheless, a special container/device is necessary to encapsulate and prevent the leakage of solid-liquid PCMs during phase change process. To address these problems, form-stable PCMs have drawing extensively attention from researchers in recent years [16–18].

In general, form-stable PCMs are mainly composed of solid-liquid PCMs and supporting materials. The biggest advantage of form-stable PCMs is that the materials shape can be kept well when the ambient temperature is higher than the phase change temperature of used solid-liquid PCMs [19]. Therefore, the major research direction of form-stable PCMs is concentrating on finding

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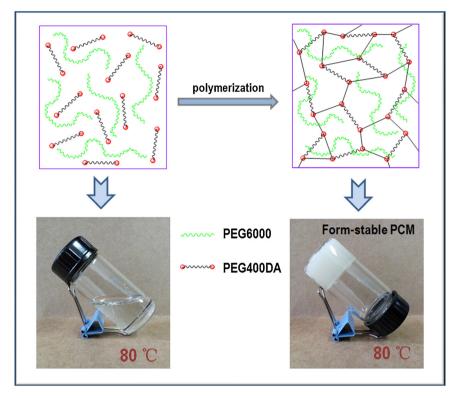


Fig. 1. Schematic route for the preparation of PEG/PPGD composite form-stable PCMs.

efficient supporting materials to remain the shape stabilization of PCMs during the phase change process. Many materials have been developed to serve as supporting materials, such as expanded perlite [20], diatomite [21], bentonite [22], vermiculite [23], expand graphite [24], polymethyl methacrylate (PMMA) [25], high-density polyethylene (HDPE) [26], poly(melamine-formaldehyde) [27] and so on.

As typical solid-liquid PCMs, PEG with excellent properties of non-toxicity, good thermal stability, relative high latent heat and wide melting point range has been extensively studied for formstable PCMs. Wang et al. [28] studied the PEG/silicon dioxide composite as form-stable PCMs. The highest weight percentage of PEG was as high as 85 wt% without leakage during the phase transition and the maximum latent heat reached 162.9 J/g. Karaman et al. [29] developed PEG/diatomite composite as form-stable PCMs. 50 wt% of PEG was retained into the porous diatomite and the phase change temperature and latent heat of PEG/diatomite composite were 27.7 °C and 87.09 J/g, respectively. Chen et al. [30] successfully prepared ultrafine phase change fibers via electrospinning with PEG as thermal storage materials and cellulose acetate as supporting substance. The maximum PEG content in the fibers was reached up to 70 wt%, resulting in a latent heat storage capacity as high as 120.18 J/g. Fang et al. [31] reported the PEG/epoxy resin composite as form-stable PCMs for thermal energy storage and its latent heat reaches 132.4 J/g.

In this study, semi-interpenetrating polymer network (semi-IPN) of PEG/poly(polyethylene glycol diacrylate) (PPGD) composite as novel form-stable PCMs was successfully prepared via in situ polymerization. The PPGD was used as supporting materials in form-stable PCMs preparation for the first time and the IPN structure can well immobilize the liquid PEG and keep good shape stability of form-stable PCMs during the phase change. The prepared form-stable PCMs were investigated with scanning election microscopy (SEM), Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), polarization optical microscopy (POM),

differential scanning calorimetry (DSC) and thermogravimetric analysis (TG), respectively.

#### 2. Experimental part

#### 2.1. Materials

Polyethylene glycol (PEG, analytical grade,  $M_n$  = 6000 g/mol) was obtained from Chengdu Kelong Chemical Reagent Co. Ltd. (China) and served as PCM. Polyethylene glycol 400 diacrylate (PEG400DA, 99.9% pure) was purchased from Guangzhou Lihou Trade Co. Ltd. (China) and used to synthesize PPGD network, Benzoyl peroxide (BPO, analytical grade) was kindly supplied from Chengdu Kelong Chemical Reagent Co. Ltd. (China) and used as initiator.

#### 2.2. Preparation of form-stable PCMs

In preparation of PEG/PPGD composite form-stable PCMs, the in situ polymerization method was conducted. Fig. 1 illustrates the schematic process for the PEG/PPGD composite form-stable PCMs. The quantified amount of PEG was added in a 100 ml round bottom flask, and placed in a water bath at 70 °C until melting completely. Then, predetermined mass of the PEG400DA with 1% monomer quality of BPO were added into the flask, and the mixture was quickly stirred for homogenization. The mixture was putted into the prefabricated mould and placed in a vacuum oven. The thermal curing was conducted in the nitrogen atmosphere at 60 °C for 3 h and then at 90 °C for 2 h to obtained PEG/PPDG composites. To verify the maximum encapsulation ratio, the composites were prepared at different mass fraction of PEG (0, 50, 60, 70, 75 and 80 wt%). All prepared composites were performed to leakage test by heating the samples to 80 °C (above melting temperature of PEG) for 30 min. The composites that do not observe leakage of melted PEG from the composites were recognized as form-stable PCMs.

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