



Fabrication and characterization of stearic acid/polyaniline composite with electrical conductivity as phase change materials for thermal energy storage



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ABSTRACT

This paper presents the experimental investigation on the thermal properties and electrical conductivity of the new microencapsulated phase change material by entrapping of stearic acid (SA) into PANI (polyaniline) shell through self-assembly method. Experimental results reveal that PANI nuclei grew on the surface of SA, and then copied its original morphological structure and finally exhibited peony flower-like morphology. The two components have good compatibility and have no chemical reaction both in the process of fabrication and subsequent use, while hydrogen bondings between the imino groups and carboxyl groups exist. The maximum mass fraction of stearic acid loaded in SA/PANI is determined as high as 62.1 wt% without seepage of melted SA from capsules. Due to the secondary doping with carboxyl group, the composite phase change material embedded with SA exhibits improved electrical conductivity from 0.3968 S cm⁻¹ to 0.7042 S cm⁻¹ when compared to PANI. The phase change temperatures and latent heats of SA/PANI are measured to be 55.6 °C and 113.02 J/g for melting and, 50.8 °C and 112.58 J/g for freezing, respectively. TG analysis test revealed that the prepared SA/PANI composite PCM has high thermal durability in working temperature range. Moreover, the results of DSC, FT-IR, TG, conductivity investigation and thermal cycling test are all show that the thermal reliability and electrical conductivity of the SA/PANI have imperceptible changes. In total, the additional electrical conductivity, high heat storage potential and good thermal reliability and stability facilitated SA/PANI to be considered as a viable candidate for thermal energy storage, antistatic thermo-regulating textiles and other applications.

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1. Introduction

Thermal energy storage is an essential method to match the thermal energy supply and demand in time or space, improve energy utilization efficiency, reduce environmental impact related to fossil energy use and overcome the intermittence and instability of the renewable energy sources [1,2]. Among various technologies for thermal energy storage and retrieval, latent heat thermal energy storage (LHTES) embedded with phase change materials (PCMs) is considered to be the most effective and promising way. Phase change materials are latent heat energy storage media, work as “latent heat storage unit” in LHTES system, which can absorb or

release high amount of latent heat when they change phase from solid state to liquid state (endothermic process) or vice versa (exothermic process) with small temperature variations. This main feature makes PCMs under the spotlight in a wide range of applications including solar energy storage and transfer [3,4], smart air-conditioning buildings [5], thermo-regulating or intelligent textiles [6], industrial waste heat recovery [7,8] and so on.

Different inorganic compounds including hydrated salts, nitrates and hydroxides, as well as organic substances, typically paraffin waxes, fatty acids and high-chain fatty acid esters of lower alcohols, have already defined as PCMs [9,10]. It is well known that inorganic PCMs have some attractive properties such as higher energy storage density, larger thermal conductivity, being non-flammable, being inexpensive and readily available. However, they also suffer principally from the strong corrosion, large super-cooling degree and segregation during phase transition under thermal cycling which requires the use of some nucleating and thickening agents in their applications. Therefore, organic solid-liquid PCMs

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have undergone rapid development and will play a prominent role in the future due to the avoidance of the problems inherent with inorganic PCMs [11]. Among various investigated organic PCMs, fatty acids were considered as potential PCMs due to their desirable features including high energy storage density and heat capacity, excellent thermal and chemical stability and appropriate thermal characteristics, such as negligible supercooling, smaller volume change during the phase transition, low vapor pressure, nontoxic and self-nucleating behavior [12–16]. Most of all, the good compatibility with construction materials and the adjustable phase change temperatures formed by binary and ternary eutectic mixtures expanded the applications of fatty acids [16,17]. Despite many desirable properties of fatty acids, they could also incur several bottleneck issues, leakage during the solid–liquid phase change, lower thermal conductivity and limited cycles of melting and solidification. To overcome the problems, alternative methods have been introduced. The main methods are concentrated in preparing microencapsulated PCMs (MEPCMs) [1,3,7,10,12], grafting PCMs with polymers [18], impregnating PCMs into porous substances (shape-stabilized PCMs, SSPCMs) [2,5,13] and inserting fins or particles with high thermal conductivity [19]. The logical solution, introduction of thermal conductivity promoter, not only improves the thermal conductivity of the PCMs but also increases the cost of the system. In particular, determining proper configurations of high conductivity inserts/structures and their interactions with conduction, convection and phase change heat transport mechanisms poses challenging issues [19]. The other feasible and cost-efficiency method, preparing MEPCMs or SSPCMs, has also been developed by scientists' significant efforts due to the shape can be maintained by the supporting structure. Due to the PCMs constrained by capillary forces in SSPCMs tends to diffuse to surface and loose gradually [7], MEPCMs received more comprehensive attention.

Microencapsulated phase change materials are very small bicomponent particles composed of the core of PCMs and the outer shell formed by polymeric or inorganic compounds. By microencapsulating PCMs into capsule wall, MEPCMs shield the PCMs from influences of outer environment and enable the core to withstand the volume change of the PCM as the phase change happens. The large surface-to-volume ratio not only improves the heat transfer rate greatly [7,10,20] but also enhances the possibility of application in the small and portable LHTES system [19]. Meanwhile, the outer shell maintains the fixed shape and resists the leakage of PCMs when phase change occurs which makes it can be used directly without extra tanks or containers in thermal applications. Thus, choice of shell materials for microencapsulation plays an very important role in regulating the properties of the microcapsules, such as morphologies, heat capacities and thermal stabilities [7,21]. Previous researches are commonly using silica or organic polymers as shell materials. Extensive efforts have been reported in improving the performance of MEPCMs, selected polystyrene (PS), melamine formaldehyde resin (MF), polymethyl methacrylate (PMMA), urea formaldehyde resin (UF), arabic gum, gelatin formaldehyde resin, polycarbonate (PC), styrene-methyl methacrylate copolymer and rigid polyurethane (PU) as shell material [3,7,14,15,19,22–24]. Unfortunately, to the best of our knowledge, the study focused on the conductive polymers microencapsulated PCMs is rare. In fact, using conductive polymers as supporting or encapsulating material to prepare MEPCMs or SSPCMs is not only feasible in theory but also reinvest the composite PCMs with the antistatic function which will expand the application area of the PCMs [16,25].

One of the promising conductive polymers is polyaniline (PANI) which possesses the merits of easy polymerization, chemical stability, environmental friendliness, relatively high electrical conductivity and can be prepared in a variety of morphologies such as

nano-fiber/-wire/-rod/-tube [26,27]. Furthermore, PANI has been applied in smart windows for dynamic daylight and solar energy control in buildings [16,28]. From this point of view, it is worthwhile to investigate the possibility of applying PANI as supporting material to prepare microencapsulated PCMs. In the previous works, Zeng JF synthesized PANI coated form-stable composite PCMs using tetradecanol as solid–liquid PCMs [29]. However, the reaction mixture is very difficult to filter. They also prepared series of myristic acid (MA)/PANI [16] and palmitic acid (PA)/PANI [27,30] form-stable PCMs by means of surface polymerization method, and then, exfoliated graphite nanoplatelets (xGnP) [30] and copper nanowires (Cu NWs) [27] were introduced to enhance the thermal conductivity of the composite PCMs. The results indicated that the PANI based form-stable PCMs exhibited satisfactory thermal performance and the thermal conductivity could be greatly improved by dopants. However, the thermal energy storage capacity was maintained for xGnP loaded products while it decreased linearly for the composite containing Cu NWs with the increasing of insert. But beyond these, studies about PANI based composite PCMs are still rare as well as no research focused on doped-PANI shell phase change material. On the other hand, the morphology, structure and properties of the composite depend on many parameters such as fabrication recipes, molecular weight of shell, additives and type of used PCMs. As a result, it is worthwhile to investigate the morphology, electrical conductivity and thermal properties of the doped-PANI microencapsulated PCMs.

Based on the greater importance of thermal energy storage, together with the studies of MEPCMs incorporated with doped-PANI, the stearic acid/polyaniline (SA/PANI) composite was fabricated via self-assembly method using SA as organic PCM and PANI as shell material aiming to develop a high-performance PANI-based composite PCMs with electrical conductivity. The microstructure, properties (thermal performance and electrical conductivity), reliability and stability of SA/PANI MEPCMs were characterized by scanning electron microscope (SEM), Fourier transformation infrared spectroscopy (FT-IR), thermogravimetric analysis (TGA), differential scanning calorimetry (DSC), accelerated thermal cycling test (ATC) and standard four probe method, together with those of SA and/or PANI for comparison purpose. Afterward, fabrication mechanism of the as-prepared samples was also explained.

2. Experimental

2.1. Materials

Stearic acid (SA, commercial grade) was obtained from Beijing Chem. Co. Ltd. and used as phase change material without further purification. Aniline (ANI, analytical reagent) was purchased from Tianjin Beichen Chemical Co. Ltd. and employed as shell monomer which double distilled before polymerization. The other chemicals, ammonium persulfate (APS, $(\text{NH}_4)_2\text{S}_2\text{O}_8$, as oxidant) was supplied by Tianjin Yongmao Chemical Reagent Factory, benzoperoxide (BPO, as oxidant) was procured from Shanghai Zhongli Chemical Co., Ltd, polyethylene glycol octyl phenylether (TX-100, as surfactant) was received from Shanghai Chemical Reagent Co. Ltd., sulfamic acid (SFA, as dopant) was sourced from TCI, Shanghai Chemical Co. Ltd., were designated to be of analytical grade and they were used without further purification. Deionized water consumed in all polymerization recipes was self-made by our laboratory.

2.2. Preparation of SA/PANI capsules

The SA/PANI capsules were chemically synthesized via a self-assembly process. Typical synthetic process is as follows: 4 mL aniline monomer, containing 12.8 g sulfamic acid was dissolved in

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