



Polymethyl methacrylate based phase change microencapsulation for solar energy storage with silicon nitride

Yanyang Yang^{a,b}, Xiaoxin Ye^{a,b}, Jie Luo^a, Guolin Song^{a,*}, Yuan Liu^a, Guoyi Tang^{a,b,*}

^a Institute of Advanced Materials, Graduate School at Shenzhen, Tsinghua University, Shenzhen 518055, China

^b Key Laboratory of Advanced Materials, School of Materials Science and Engineering, Tsinghua University, Haidian District, Beijing 100084, China

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Abstract

Silicon nitride was applied to enhance the thermal performance and mechanical properties of phase change microencapsulation (PCM) based on polymethyl methacrylate (PMMA) shell and n-octadecane core. 'A molecular bridge' was constructed to modify the surface of silicon nitride and to eliminate boundary layer between inorganic silicon nitride particle and organic PMMA shell. Thus, an innovative modified silicon nitride phase change microencapsulation (MPCM) with which silicon nitride uniformly disperses in PMMA as shell and n-octadecane as core was successfully prepared. The microencapsulation was characterized using Fourier transformed infrared spectrophotometer (FTIR), field emission scanning electron microscope (FESEM), differential scanning calorimeter (DSC) and thermal gravimetric analyzer (TGA). A micro/nano-hardness tester was also employed, in order to investigate mechanical performance of shell. The result shows that the MPCM containing 66.4% n-octadecane has 121.11 J/g latent heats of melting and 122.01 J/g latent heats of crystallization with the modified silicon nitride percentage up to 10 wt.%. The modified silicon nitride could not only enhance thermal performance, but also improve mechanical strength up to 16.24 mN which is 4 times higher than that of PCM. Additionally, wrinkles on the surface of MPCM improved special surface area as well as adaptation of volume changes during phase change process. The prepared MPCM is expected to exhibit better performance in solar energy storage technology.

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1. Introduction

Phase change energy storage technology is one of the most efficient approaches to cover uneven energy distribution in space and time (Salunkhe and Shembekar, 2012), which has served to solar energy (Konuklu et al., 2014; Zhang et al., 2012), heat recovery (Wu et al., 2014; Park et al.,

2014), battery thermal management (Archibold et al., 2014; Duan and Naterer, 2010), building (Khudhair and Farid, 2004; Kuznik et al., 2011) and other fields (Zhou et al., 2014; Diaconu et al., 2010). In recent researches, one of the most efficient way of phase change materials is phase change microencapsulation (PCM) by which erosion and leakage were prevented (Oró et al., 2012; Hawlader et al., 2003). Shells of PCM made from macromolecular materials are extremely critical (Hänchen et al., 2011; Toppi and Mazzarella, 2013; Zhao et al., 2013) playing an important role in regulating the performance of PCM, such as preventing the interior phase change materials from leaking,

* Corresponding authors at: Institute of Advanced Materials, Graduate School at Shenzhen, Tsinghua University, Shenzhen 518055, China. Tel./fax: +86 75526036752.

E-mail addresses: song.guolin@sz.tsinghua.edu.cn (G. Song), tanggy@tsinghua.edu.cn (G. Tang).

enhancing mechanical strength and improving thermal conductivity (Rao et al., 2012; Yavari et al., 2011; Ma et al., 2013). As can be observed from the literature, some polymer shells are toxic or low thermal performances or uneconomicalness for industrialization (Khateeb et al., 2005; Shirin-Abadi et al., 2011; Zeng et al., 2014). Zhu et al. (2012) prepared microencapsulation with melamine–formaldehyde resin as shell and n-dodecane as core. Since virulence couldn't be eliminated coming from raw materials, the applications were strongly restriction. Chen et al. (2012) embedded alumina into melamine–formaldehyde shells so as to improve thermal conductivity. While the maximum additional quantity of alumina in the microencapsulation was up to 12.7%, it may lead alumina break from shell and even leakage during the phase change process for the huge different thermal expansion rate between of alumina and macromolecule material. Thus, a good thermal conductivity materials with low expansive coefficient was needed. Also, the mechanical performance need to improve to keep the microencapsulations from broking. Researches on mechanical strength which is a critical nature of PCM were rarely reported, because the microencapsulation is too small to measure.

Poly(methylmethacrylate) (PMMA) (Antonioli et al., 2012; Pan et al., 2013; Bendic and Amza, 2012) which is a commercially available acrylic resin on the merits of non-toxic, chemical resistance and high impact strength is an excellent shell material that had been reported in our group previous work (Qiu et al., 2012; Ma et al., 2010; Qiu et al., 2013). However, the improvement of mechanical toughness of PMMA shells and the shorten supercooling of PCM are inevitable to widen the application of PCM.

In previous researches, data for phase change materials based on blends of inorganic silicon nitride, with an improved thermal conductivity up to 35% and reduced supercooling of PCM are presented (Yang et al., 2014). Inorganic silicon nitride particles (Zhang et al., 2014; Hao et al., 2013; Deepthi et al., 2012) are economical, stable abrasability, non-oxidizability, thermal vibration resistance and high thermal conductivity ceramic powders. The thermal conductivity and linear expansion coefficient of Si₃N₄ are 16.7 W/(m K), and $2.75 \times 10^{-6}/^{\circ}\text{C}$ (20–1000 °C), respectively. In this paper, silicon nitride was employed to enhance organic polymer shell performance. However, there are very severely boundary problem between inorganic silicon nitride particles and organic shells which prevent silicon nitride binding to PMMA shells efficiently. In order to eliminate surface boundary effect, the surface of silicon nitride was pre-modified with silane coupling agent connecting silicon nitride. Comparing with unmodified silicon nitride, modified silicon nitride proved to be more compatibility with PMMA shells by our data. As a net construction, PMMA could fasten silicon nitride and form high performance shells of PCM. Thus, the mechanical strength of MPCM was substantially improved as well as the thermal performance of MPCM. Preparing PCM with inorganic materials was

rarely reported. Furthermore, enhanced performance of PCM by silicon nitride had never been noted in any article.

2. Materials and experimental

Silicon nitride (Tianjin Nitride Advanced Materials co., Ltd, China) was modified by silane coupling agent KH-570 (Sinopharm Chemical Reagent Co., Ltd.) at first. 95 wt.% ethyl alcohol and 5 wt.% water were join into a flask to form alcohol-water solution. A little acetic was drop in it adjusting the pH between 4.5 and 5.5. After that, 2 wt.% silane coupling agent KH-570 was added stirring for 5 min. Then silicon nitride were mix into the solution, and dispersed using ultrasound under 80 °C for 1 h. The modified silicon nitride were cleaning twice by ethyl alcohol. And then it was moved to an oven desiccating at 120 °C for 10 h.

Suspension-like polymerization method was employed and recipes for various types of polymerization monomers are given in Table 1. 7 g sodium salt of styrene-maleic anhydride copolymer (TA, Shanghai Leather Chemical Works, China) was dissolved in 100 g distilled water for 15 min under vigorous agitation (1000 rpm). At the same time, 10 g n-octadecane (99 wt.%, Alfa) and 3 g Pentaerythritol tetraacrylate (PETRA, 80 wt.%, Nanjing Shoulashou Co., Ltd.) 7 g Methylmethacrylate (MMA, A.R., Tianjin Damao Chemistry Reagent Co., Ltd.) and 3 g modified/unmodified silicon nitride (Tianjin Nitride Advanced Materials Co., Ltd, China) were ultrasonic dispersion separately for 15 min. And then, all the prepared materials were sequential gathering into a 250 mL three-neck round bottomed flask and maintained in a 35 °C water bath for 15 min under vigorous agitation (1000 rpm) to form a stable oil-in-water emulsion. After the addition of 2, 2-azobisisobutyronitrile (AIBN, 98 wt.%, Shanghai Jingchun Chemical Co., Ltd.), the polymerization process had been proceeded at 85 °C under vigorous agitation (540 rpm) for 5 h. The obtained microcapsules were purified by centrifugation and dried in an oven at 45 °C for 24 h.

The chemical structure was investigated using Fourier transformed infrared spectrophotometer (FTIR, VERTEX 70, BRUKER). The particle sizes was measured by laser scattering particle size distribution analyzer (LA-950V2, HORIBA). Covered with a layer of gold, the surface morphologies were observed under a field emission scanning electron microscope (FESEM, S4800, HITACHI). Thermal properties were recorded on differential scanning calorimeter (DSC, 823E METTLER TOLEDO) in the range of 0–50 °C in argon atmosphere with a heating/cooling rate of ± 5 °C/min and a flow rate of 60 mL/min. The thermal stability of microencapsulation was characterized by a thermal gravimetric analyzer (TGA, TGA/DSC1 METTLER TOLEDO) in the range of 0–500 °C in argon atmosphere with a heating rate of 10 °C/min.

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