



Hierarchical nano-activated silica nanosheets for thermal energy storage



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ABSTRACT

This paper reported on the synthesis of silica nanosheets (SNSs) by structurally modifying natural coal-series kaolinite mineral (Kc). Ag nanoparticles (AgNPs) of about 5 nm were uniformly attached on the surface of Sn^{2+} -activated SNSs to form an emerging hierarchical porous nanostructure, which was further hybridized with polyethylene glycol (PEG) to produce PEG@Ag/Sn²⁺-SNSs. The maximum loading capacity and melting enthalpy of PEG@Ag/Sn²⁺-SNSs could reach 66.1% and 113.9 J/g, respectively, and its thermal conductivity showed up to 0.84 W/(m K). The results demonstrated that well dispersion of AgNPs and hierarchical porous structure of Ag/Sn²⁺-SNSs could synergistically enhance the thermal conductivity. Intriguingly, the introduction of AgNPs could lead to the obvious decrease of the melting and solidifying period, and simultaneously promoted the heat transfer of the composite phase change material (PCM). Furthermore, the composite PCM could retain good thermal reliability after 200 cycles, indicating its potential application in the thermal energy storage system. Atomic-level mechanism for the enhanced thermal conductivity of the composite PCM was also discussed.

1. Introduction

The demand and consumption of energy is increasing significantly along with the blooming of the industry and agriculture. As a consequence, it is important to use existing energy resources effectively. Both sensible heat and latent heat thermal energy storage (LHTES) have been confirmed as efficient techniques for energy conservation. The phase change material (PCM) used in the LHTES possesses the advantages of high energy density and chemical stability compared with the sensible heat thermal energy storage. Therefore, application of PCM in LHTES has become a common choice [1–6]. PCM is defined as material that is able to store and release thermal energy from the melting to the solidifying process. When the outer circumstance temperature exceeds melting temperature of PCM, it will absorb the heat, otherwise the chemical bonds of PCM would be destroyed. When the circumstance temperature becomes lower than solidifying temperature of PCM, the material will release heat and return to solid state [7]. Until now, three major categories of PCMs have been studied: organic, inorganic and eutectic PCMs [8–11]. Organic PCMs have been used extensively because of the appropriate phase change temperature, small volume change in the phase change process and commercial availability at a reasonable cost [12,13]. For instance, PEG is a typical organic PCM

that has been well studied [14–16]. However, two disadvantages have been identified for PEG and PEG-based PCMs. One is the leakage of liquid phase over the melting temperature, and the other is the relatively low thermal conductivity. In order to solve these issues, many clay minerals have been extensively studied as supports to stabilize organic PCM and prepare composite PCM due to their natural porosity, thermal stability, and low price [17–21]. Popular examples are halloysite [22–28], attapulgite [29–31], montmorillonite (bentonite) [32–36], kaolinite [37–39], diatomite [40–42] and expanded perlite [43–45].

Although the thermal conductivity of clay-based PCMs has been increased remarkably, the highly efficient heat transfer technology is still in demand [15,46,47]. Sari impregnated PEG into the bentonite to prepare a new composite PCM [48], the thermal conductivity of composite PCM was improved from 0.39 to 0.69 W/(m K) by addition of graphite (5 wt%). Similarly, coal-series kaolinite mineral (Kc), as a natural mineral, has unique characteristics for technological applications. Kc ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$) is a 1:1 type clay mineral formed by SiO_4 tetrahedral sheets and $\text{AlO}_2(\text{OH})_4$ octahedral sheets. In order to change its poor textural properties, Kc could be pretreated to prepare the SNSs, and Ag nanoparticles (AgNPs) can be added into this composite PCM to further improve the thermal conductivity.

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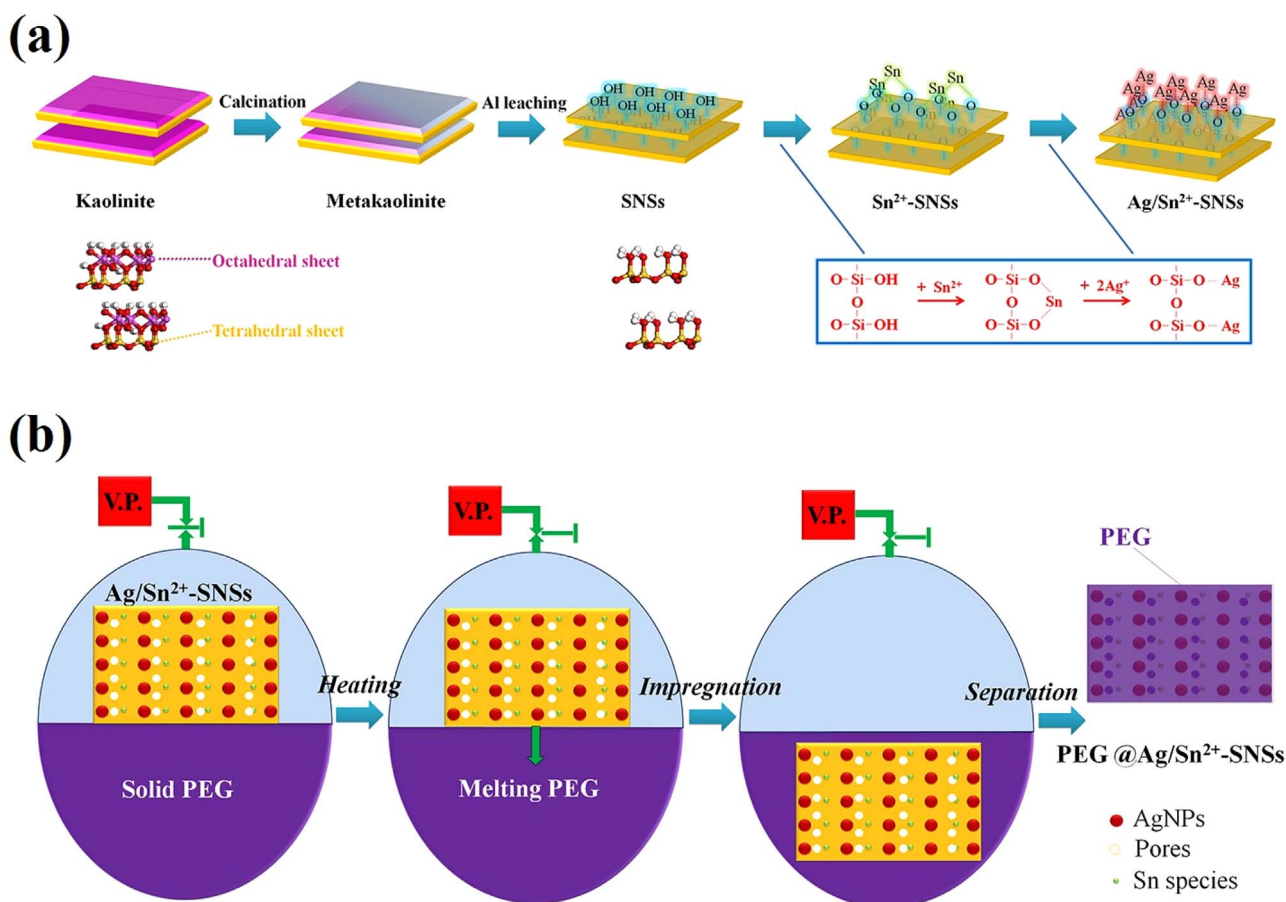


Fig. 1. (a) Schematic routes for preparation of Ag/Sn²⁺-SNSs powder; (b) images of vacuum impregnation treatment for preparing PEG@Ag/Sn²⁺-SNSs composite (V.P.: vacuum pump).

This paper aims to prepare an emerging nanocomposite with high thermal conductivity for thermal energy storage. Natural Kc was structurally modified to prepare silica nanosheets (SNSs), which was later activated by adding Sn²⁺ and loaded with AgNPs. Further hybridizing PEG resulted in the formation of composite PCM. The microstructure, phase change performance, thermal conductivity, thermal stability and reliability of the composite PCM were systematically studied.

2. Experimental

2.1. Preparation of silica nanosheets (SNSs)

Pristine Kc used in our experiments was obtained from Shanxi, China. Fig. 1a shows the schematic presentation of the preparation of Ag/Sn²⁺-SNSs. In a typical preparation of silica nanosheets, natural Kc was first transformed into the anhydrous phase metakaolinite by calcination at 700 °C for 5 h. Then, 1 g of metakaolinite was treated with 50 mL of 2 M HCl solution at 80 °C for 8 h. The resultant precipitate was filtrated, washed and dried at 110 °C to obtain the resulting product silica nanosheets (SNSs).

2.2. In-situ reduction of Ag nanoparticles (AgNPs) on Sn²⁺-silica nanosheets (Sn²⁺-SNSs)

1 g of SNSs was added to 100 mL distilled water. 0.25 g of SnCl₂·2H₂O was added into 50 mL of 0.01 M HCl solution, and mixed with the above solution for 8 h at room temperature. The precipitate was then recovered by sedimentation, followed by washing with distilled water three times. Therefore, the Sn²⁺ activated SNSs (Sn²⁺-

SNSs) were obtained. Afterward, 1 g of Sn²⁺-SNSs was added into 100 mL 1 mmol of AgNO₃ aqueous solution under vigorous stirring for 5 min at room temperature. Then 11 mL of ammonia solution was added and stirred for 3 h.

2.3. Preparation of PEG@Ag/Sn²⁺-SNSs composite PCM

Chemically pure PEG (*M_w* 6000) was supplied by Sinopharm Chemical Reagent Co., Ltd. PEG@Ag/Sn²⁺-SNSs was prepared by vacuum impregnating method (Fig. 1b): 3 g Ag/Sn²⁺-SNSs and 7 g PEG were added into a conical flask by attaching a vacuum pump to prevent backflow. Firstly, vacuum the system to 0.1 MPa for 10 min, and subsequently put the conical flask into thermostatical water bath at 85 °C for 30 min. After cooling, the reactants were placed at 80 °C to remove the excessive PEG to finally obtain PEG@Ag/Sn²⁺-SNSs composite.

2.4. Characterization

X-ray diffraction (XRD) of the samples was carried out using a Rigaku D/max-2550VBR + 18 kW powder diffractometer using Cu Kα radiation ($\lambda=1.5418 \text{ \AA}$) from 5° to 80° of 2θ. Fourier transform infrared spectroscopy (FTIR) spectra of the samples (KBr pellets) were recorded on Nicolet 5700 spectrophotometer. Scanning electron microscopy (SEM) was performed with a field-emission scanning electron microscopy (TESCAN, Model MIRA3 LMU) system to observe the surface morphology of the samples, and the samples should be coated with conductive gold before test. Transmission electron microscopy (TEM) images were collected with a JEOL JEM-2100F electron microscope, and the morphology of Ag/Sn²⁺-SNSs was observed by a high-

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