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A quick-fix design of phase change material by particle blending and spherical agglomeration

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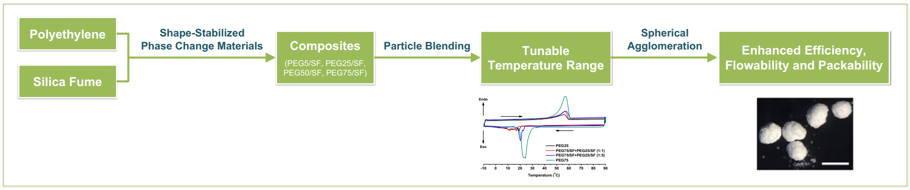
HIGHLIGHTS

- Low-cost polyethylene glycol/silica fume composite as phase change material.
- Composite blending for a tunable temperature range of phase transitions.
- Phase transition range of polyethylene glycol can be broadened to up to 40 °C.
- Spherical agglomerates can enhance mechanical properties and thermal behaviors.
- Thermal stability and reproducibility even undergoing 100 temperature cycles.

GRAPHICAL ABSTRACT



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ABSTRACT

The aims of this study were to impregnate polyethylene glycol (PEG) 4000 in low-cost silica fume (SF) to form phase change material (PCM) composites with cementitious value, and to provide a quick-fix design for PCM (1) with tailor-made thermal properties and behaviors by particle blending of two types of polyethylene (PEG)/silica fume (SF) composites having different PEG wt% loading, and (2) with enhanced physical properties by turning the powdery PEG/SF composites into round granules through spherical agglomeration. The simple composite blending method was used to broaden and tune the application temperatures in response to variable conditions and environments without the need of searching for new materials to mitigate global warming. Spherical agglomerates of PEG/SF composite exhibited a good homogeneity in thermal properties and low Carr's indices indicating of excellent flowability, packability and compactibility, and offering an enhanced contact area for heat transfer and uniform mixing with other building materials. Noticeably, the agglomerates displayed higher heat capacity values of solid phase, C_{ps}, and liquid phase, C_{pl}, than those of the composite determined by temperature-history method. The thermal stability of PEG75/SF composites was also attested by the small enthalpy loss, and the highly reproducible melting and solidification behaviors after more than 100 temperature cycles.

1. Introduction

Given with the increase in demand, thermal energy storage (TES) is one of the critical issues to be addressed for utilizing intermittent renewable energy sources in a thermal form. The energy can be reserved and stored in a well-insulated system while the

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internal energy of material changes in terms of sensible heat, latent heat and/or reversible chemical reaction [1,2]. Phase change material (PCM), also called latent heat energy storage material, possesses the characteristic of absorbing or releasing thermal energy as the material undergoes a phase change at a nearly constant temperature. Nowadays, many applications of PCM have been studied in building energy conservation [2–4], underfloor heating system [5,6], thermal protection of food and electronic devices [7,8], textiles [9,10], air-conditioning [11], and waste heat recovery [12,13] since the energy crisis during late 1970s [14].

PCMs are classified as solid-solid, solid-liquid, solid-gas and liquid-gas types. The former two types (especially solid-liquid type) of PCM have received much attention due to their thermal properties and practical considerations. Besides, solid-gas and liquid-gas PCMs are associated with higher latent heat of fusion but larger changes in volume/mass making the system more complex and impractical. Solid-solid PCMs have shown the advantages of non-toxicity, non-corrosiveness, high heat efficiency and stability. However, their drawbacks are poor thermal conductivity, relatively expensive and easy to turn into volatile plastic crystal during phase transition. Low-cost solid-liquid PCMs have a higher energy storage capacity and smaller volume/mass change during phase transformation compared to the other three types, but still suffer from phase separation, supercooling, easy leakage, and chemical instability [6,12]. Some special devices, such as shells and tubes, exchangers or cans, are required to prevent the leakage of PCM, and that however, will increase the heat resistance and the cost [15].

One of the applications, for example, is to mitigate global warming by reducing the heating and cooling load of buildings. Cementitious materials are widely used as building materials, and their mixing with PCMs physically can enhance the thermal energy storage capacity and improve the energy efficiency in buildings [16]. Although the direct incorporation by mixing was simple and handy, a low amount of PCM incorporated resulted in lower energy storage. Even worse, such method was vulnerable to leakage of liquid PCM and the leaked PCM could adversely affect construction system. For instance, a serious leakage of paraffin of 57.3% was reported while paraffin/diatomite composite was incorporated into cementitious mixture upon mixing with water [17]. Also, PCM is possible to leak from a porous material if ambient temperature is higher than melting point of the PCM, and its energy storage capacity will drop significantly.

A large number of studies focused on the exploration of the diversity of PCMs with desirable properties and utilizations. An encapsulation method has been used to prepare "shape-stabilized PCMs" or called "encapsulated PCMs" for resolving some of the above-mentioned issues. Additionally, this technique has also been developed by using a porous material as a carrier to adsorb very poorly soluble drug for drug release [18]. Shape-stabilized PCM usually includes a working substance and a supporting material. The functions of the supporting material are: (1) to confine the melted working substance, (2) to prevent from leaking and isolate it from harmful environmental factors, (3) to improve thermal conductivity, and (4) to keep the whole system in solid state even after phase transformation of the working substance by blending, adsorption and impregnation, or chemical methods such as graft copolymerization and sol-gel methods. Unlike common PCMs, no extra space or container is needed for encapsulation of PCM into a porous supporting material [19].

This kind of shape-stabilized PCM composite can be applied in the exterior wall of building to absorb heat from the surroundings and solar radiation during daytime, and then to release heat to the surroundings at night for control of the indoor temperature. Consequently, the improvement in the drastic change of global climate could be accomplished by extending the use of PCM composite instead of the use of air-conditioning and heating systems [1].

For building materials, low-cost, safety and ease of construction are the major concerns. Polyethylene glycol (PEG) was chosen as the working material in this study because of its inexpensive nature, non-volatility, high phase change enthalpy, chemical inertness and wide range of melting temperatures from 3.2° to 68.7 °C with a tunable molecular weight from 2000 to 12,000 [20]. The melting and solidification temperatures and the heat of fusion of PEG increased with the increase in the molecular weight [20]. As the molecular weight of PEG became larger, there was a tendency that PEG would form the crystalline phase more easily because of their extended segmental mobility and more convenient geometric alignment [21].

However, the leakage of PEG is a main drawback as a PCM. Since silica fume (SF) is a low-cost and common mesoporous material having a high surface area and could be used to enhance mechanical characteristics of concrete [22], it was chosen as the supporting material for salvage to encapsulate PEG and confine its crystallization. SF was a solid waste from ferrosilicon industry [23] and also one of the feasible candidates being an economical and lightweight building material [24]. Some other examples of shaped-stabilized PCM of utilizing PEG with more expensive substrates, such as PEG/active carbon [15,25], PEG/graphene oxide [20], PEG/expanded graphite [26], PEG/SiO_2/β-aluminum nitride [27], PEG/diatomite [1], PEG/silica [28–30], PEG/agarose, PEG/cellulose, and PEG/chitosan [31], have also been reported to prevent PEG from leaking by the capillary force.

In general, there are four methods to improve thermal properties and behaviors of shape-stabilized PCMs by varying (1) molecular weights [20], (2) porous supporting materials [20,26–31], (3) pore sizes of a porous material [15] and (4) mass fractions [31]. However, if a particular shape-stabilized PCM product prepared by those methods could not meet up with the desirable properties, conventionally, some of the listed factors would have to be redesigned. Different from those methods, our investigations were to impregnate the SF with different loading wt% of PEG to form different types of composite. The solidification temperature range and the thermal behaviors of the PCM materials would then be tailor made by simply blending the composites at different ratios for the first time as evidenced by differential scanning calorimetry and temperature-history method. This technology enabling platform would path the way for future study of building materials mixed with PCM products in response to variable conditions, environments and climates.

Furthermore, flowability, packability and compactability of powders can sometimes have major effects on downstream processes such as mixing and blending, filling, packaging and end-use [32]. For instance, the formation of ammonium nitrate agglomerates with a higher density and closer packing, which was utilized as an energetic material, was proven to overcome caking problem and avoid undesirable voids and cracks in propellant for a better burning characteristic [33]. As for the potential application of PCMs in building energy conservation, the enhancement in mechanical property is one of the important things to strengthen building structures, and therefore, to increase practicality and energy efficiency of PCMs/composites [17]. Yang et al. had filled three kinds of PCM into spherical capsules in a packed bed for more efficient thermal energy transfer [34]. Xu and Li reported that the compressive strength of paraffin/diatomite cement composite was reduced with the increase of paraffin in diatomite [35]. Therefore, in our present study, spherical agglomeration [36] has been extended to turning fine PCM composite powders into round granules for the very first time, and in consequence, leveling up their flowability, packability, mechanical strength and thermal behaviors.