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Fabrication and stability of form-stable diatomite/paraffin phase change material composites

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ABSTRACT

In this study, three grades of diatomite particles, DP1, DP2 and DP3, were used to produce phase change material composites, DP1P, DP2P and DP3P, by absorbing paraffin into the pores of diatomite particles. The absorption mechanism of the finest DP1 was found to be different with that of DP2 and DP3. In addition to absorption mechanism, stability of the composites was also investigated. However, a significant amount, up to 67%, of paraffin leaked from the composites when they were used in concrete mixture directly. In order to prevent the leakage, a surface-modification method was proposed and promising results were achieved. Thermal behaviors of concrete panels incorporating with DP1P and Micronal were then compared with a control one. The results indicate that addition of PCM composites improves the thermal inertia.

1. Introduction

Phase change materials (PCM) have the ability to store a large amount of latent heat within the small temperature range associated with a phase change; thus, incorporating phase change materials in building components may help reduce indoor temperature oscillations. Additional advantages of utilizing PCMs include improved occupant comfort, compatibility with traditional building enclosure technologies, and the potential for application in retrofitting projects. Extensive research on the application of PCMs in buildings to achieve latent heat storage has been conducted in the past several decades to make use of these advantages; examples of these works can be found in the literature [1–3].

Materials used as PCMs can be divided into different subcategories based on their chemical composition. Three groups are commonly made: organic compounds, inorganic compounds and inorganic eutectics or eutectic mixtures. Inorganic PCMs include salt hydrates, salts, metals and alloys, whereas organic PCMs consist of paraffin, fatty acids, fatty acid ester and polyalcohols. Eutectics may be divided in three groups according to the materials of which they consist: organic–organic, inorganic–inorganic and inorganic–organic eutectics. Applications of different PCMs were summarized in the review papers [4,5]. More recently, fatty acid ester [6,7] attracts the attention as a new subgroup among organic PCMs. However, among these materials, paraffin is believed to be the most promising phase change material because it has a large latent heat, low cost and is stable, nontoxic and not corrosive [1,8].

The latent heat, or thermal energy storage, of PCMs could be utilized in buildings in many different forms, including a PCM-enhanced building envelope, arrays of PCM containers, or thermal insulation materials impregnated with PCMs. In these applications, PCM-enhanced building envelopes, such as wallboards, floors and ceiling boards that have been impregnated with PCM, have been proven to be successful because of their ability to reduce energy consumption due to space conditioning and to reduce peak loads. Most PCMs, including paraffin, use chemical bonds to store and release the heat, and the thermal energy transfer occurs when a material changes from a solid to a liquid or a liquid to a solid.

However, one of the basic problems with the use of solid-to-liquid PCMs is the containment of the PCM in the building material. For solid-to-liquid PCMs, the PCM exists in the solid phase below the melting point and in the liquid phase above the melting point. Certain problems, such as leakage, are associated with the presence of a liquid-phase PCM. Another concern regarding the use of PCMs in building materials is that, if improper containment is used, additional thermal stresses and vapor pressures caused by the volume change during a solid-to-liquid phase change could occur.

PCMs could be impregnated in building envelopes in two different ways. In the first method, PCMs are directly incorporated into building materials by wet or dry mixing and immersion. U.S. patent by Salyer et al. [9] disclosed phase change compositions that embedded long-chain hydrocarbons into cementitious

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Fig. 1. DP1, DP2 and DP3 (from left to right).

compositions by either wet or dry mixing. Hawes et al. [10] investigated concrete impregnated with PCMs by immersion and determined the absorption constant of the concrete according to different processing conditions, including the immersion time, type of PCM, temperature, etc. Direct incorporation methods are simple and easy to use. However, the amount of PCM that they can incorporate is small, which results in a low thermal energy storage level.

For the second incorporation method, PCMs can also be indirectly incorporated into building materials by using microencapsulated PCM and granular PCM composites. Microencapsulated PCM is produced by enclosing PCM in a thin polymeric film that has a high molecular weight. Microencapsulated PCM can be in powder form or slurry, and its particle size is in the order of microns. Micronal® from BASF is a type of microencapsulated PCM that can be incorporated in mortars, concrete and plasterboard as a passive thermal energy storage system. It is also used as slurry in active systems [11]. Zhang et al. [12] developed a vacuum impregnation method to make granular phase change composites. In this investigation, expanded perlite, expanded clay and expanded fly ash granules were used, with mean particle sizes of 2 mm, 3 mm and 10 mm, respectively. The granular phase change material composites (expanded perlite/paraffin) were used to produce extruded cementitious materials [13]. It was found that the heat capacity and thermal inertia of the PCM-incorporated samples were significantly larger than the samples without PCM.

Although the commercial microencapsulated PCMs demonstrated a positive thermal impact on the thermal energy storage of the cement-based composites, there were some incompatibility problems with cement matrix. Microencapsulated PCMs with broken polymer shells were found in the cement matrix because of collisions and abrasion with other aggregates and because of the high pH environment of the cement hydration system [14].

Thus, there is still a need for the development of a PCM containment method that is both economical and stable during the incorporation into the application of building materials. Additionally, it is better to have the contained PCMs be provided in a loosely bound or powder-like form, which will minimize the risk of PCM leakage.

Accordingly, researchers have developed a series of powder-like, form-stable PCM composites. U.S. Patent titled “Dry powder mixes comprising phase change materials” awarded to Salyer [15] disclosed a production method of a free flowing, conformable, powder-like mix of silica particles and phase change materials. The silica particles have a critical size of approximately 0.005 to 0.025 μm, and the amount of PCM added to the silica must be 75% or less. Sari et al. [16–19] have developed many form-stable PCM composites by using a vacuum impregnation and blending method. In their investigations, building materials such as gypsum, cement, perlite, diatomite and vermiculite were used to produce form-stable, powder-like PCM composites. The particle size of these materials is below 150 μm. Xu et al. [20] developed diatomite/paraffin form-stable PCM composites by using the blending method. The mean particle size of diatomite is 26.12 μm, and melting temperature of paraffin is approximately 40 °C. Porous carbon materials including expanded graphite, active carbon and ordered mesoporous carbon were used to developed form-stable PCM composites by Wang et al. [21]. PX PCM powder from Rubitherm® is one of the commercially available, powder-like PCM composite in the market. The average particle size of PX powder is 150 μm. However, the significant leakage of paraffin was found when the authors attempted to mix the PX powder into a concrete mixture.

To the authors’ best knowledge, the influences of the particle size of the carrier material and the stability of the phase change material composites have not been discussed in the literature. In this study, form-stable phase change material composites were developed by blending three particle-size grades of diatomite particles and RT21 paraffin, which has a melting point of approximately 21 °C, from Rubitherm®. The composites were then incorporated into concrete to determine the amount of paraffin leakage. Diatomite originates from a siliceous, sedimentary rock that principally consists of the fossilized skeletal remains of diatom, a unicellular aquatic plant related to algae that existed during the tertiary and quaternary periods [22]. Because of its high porosity and thermal resistance, diatomite is an attractive material that offers many promising applications in a variety of industries. We did not use the vacuum impregnation method because we found that there was a negligible difference in the absorption amount of samples impregnated with or without the vacuum method. The behavior of the composites in high-temperature conditions and the stability of the composites in the concrete mixture were also investigated. To overcome the problem of the paraffin leaking during the incorporation of the composites into the concrete, a simple but effective surface-modification method was proposed. Preliminary results indicated that the leakage was eliminated.

2. Materials

2.1. Diatomite

Diatomite with three grades of particle size, shown in Fig. 1, was provided by Mount Sylvia Pty. Ltd. (Queensland, Australia). In the following analysis, the diatomite particles will be denoted as DP1, DP2 and DP3 according to increasing particle size. DP1 is produced by milling larger diatomite particles such as DP2 or DP3. Fig. 1 shows that angle of repose of DP1 is much larger than the angles

Table 1
Bulk density and particle density of diatomite particles.

	Bulk density (g/cm^3)	Apparent particle density (g/cm^3)	Ratio
DP1	0.291	0.855	0.340
DP2	0.484	1.261	0.38400
DP3	0.513	1.360	0.377