



Preparation and properties of fatty acids based thermal energy storage aggregate concrete

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HIGHLIGHTS

- Thermal storage aggregates were prepared using fatty acids, diatomite and ceramsite.
- Thermal storage aggregates reduced early age temperature rise rate and the peak value.
- The room temperature fluctuation was reduced by as high as 5.94 °C.

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ABSTRACT

This paper aims to study the influence of macro-encapsulated phase change material (PCM) aggregate on the workability, mechanical strength, early age hydration temperature rise and thermal properties of cement concretes. A ternary fatty acid eutectic composed of laurie acid, myristic acid and palmitic acid was used as PCM, and diatomite and ceramsite based thermal energy storage aggregates were manufactured by using a direct impregnation treatment. The results show that the PCM has a melting temperature of 31.1 °C and latent heat of 166.6 J/g. The PCM adsorption volume in pore system reaches 28.1% and 89.8% of the total porosity for ceramsite and diatomite respectively. The addition of thermal energy storage aggregates decreases the slump at fresh status, compressive strength and thermal conductivity of concrete. Concrete incorporating 80% of thermal energy storage aggregate by volume has a compressive strength of higher than 18 MPa. Diatomite based thermal energy storage aggregate presents a better effect on controlling the early age hydration temperature rise than ceramsite based one. The incorporation of thermal energy storage aggregate significantly improves the temperature fluctuation in the testing room when exposed to a heating-cooling environment. Therefore, this developed thermal energy storage aggregate concrete has a great potential for improving the thermal comfort of buildings in severe climates.

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1. Introduction

Due to the global warming and shortage of energy resources, much attention has been paid to the application of phase change materials (PCMs) to thermal energy storage (TES) systems in recent years. PCMs can effectively store or release a great amount of latent heat to reduce energy consumption during the process of melting or solidification. It was reported that about 65% of energy consumption in buildings is attributed to space heating and cooling [1]. The integration of PCMs in buildings presented promising

results to improve indoor air temperature fluctuations and thermal comfort of buildings in different climates [2–5]. Meanwhile, PCMs used in residential buildings could contribute to mitigate greenhouse gas (CO₂) emission over the life span of building and effectively reduce the disadvantages of the urban heat island (UHI) phenomenon [6–8]. On the other hand, with human settlements expanding, more and more desert and plateau regions become potential living places. In these regions, thermal comfort of buildings is severe due to the significant temperature variation between daytime and nighttime. Therefore, developing a structural-functional building material by incorporating PCMs is an optional method, which stores energy during daytime and releases it during nighttime [9,10].

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Based on phase change state, PCMs can be divided into four categories including solid-solid PCMs, solid-liquid PCMs, solid-gas PCMs and liquid-gas PCMs [11]. Among them the solid-liquid PCMs is regarded as the most suitable material for thermal energy storage of buildings. According to the chemical composition, the solid-liquid PCMs comprises organic PCMs, inorganic PCMs and eutectics [12]. Organic PCMs have several advantages including availability in a large temperature range, no supercooling phenomenon, chemical stability, recyclability and good compatibility with other materials [12,13]. As typical organic PCMs, fatty acids have a high latent heat and adjustable melting temperature. Furthermore, they are chemically stable, nontoxic and non-corrosive [14]. It was reported that capric acid ($C_{10}H_{20}O_2$), lauric acid ($C_{12}H_{24}O_2$), myristic acid ($C_{14}H_{28}O_2$), palmitic acid ($C_{16}H_{32}O_2$) and stearic acid ($C_{18}H_{36}O_2$) have suitable melting temperatures of around 30–70 °C and superior latent heat of higher than 145 kJ/kg [15]. Therefore, fatty acids attract a lot of attentions for reliable thermal energy storage of buildings.

The simplest method of applying PCMs into building structures is the direct impregnation of PCMs into gypsum, concrete or other porous materials [16]. However, severe leakage of PCM was observed after cycles of heating and cooling and an interaction between PCM and porous matrix possibly occurs [17], and finally the long-term effectiveness is significantly hindered. To overcome these shortages, different encapsulation methods were developed to prepare shape-stabilized PCMs which can be then incorporated in building materials. The micro-encapsulation consists in enclosing PCM substance in microscopic polymer capsules in the form of powder, which is then incorporated in gypsum, mortar and concrete [18,19]. The macro-encapsulation consists in incorporating PCM in porous granules (expanded clay ceramsite, normal clay, expanded shale and diatomite) to prepare thermal energy storage aggregates [20,21]. These porous granules are usually applied as lightweight aggregates in concretes, having a good compatibility with cement matrix. Expanded clay ceramsite has a honeycomb structure which is formed by the release of thousands of small bubbles when heating clay to around 1200 °C in a rotary kiln. Diatomite is a naturally occurring siliceous sedimentary rock that has a low density and strength due to its high porosity. Both expanded clay ceramsite and diatomite have high porosities for absorbing PCM and are easily available in China. Test results showed that the maximum absorption of PCM by porous aggregates reached as high as 68% by weight, having a large thermal energy storage density and being easy for large scale processing [22,23]. However, different PCM-porous aggregate systems have variable influences on concrete properties and further studies should be carried out on the thermal performance of the manufactured concrete exposed to different climates.

In this paper, a mixture of lauric acid, myristic acid and palmitic acid was prepared with melting temperature of 31.1 °C and ceramsite and diatomite were used as containers to manufacture two types of thermal energy storage aggregates. The workability, compressive strength, thermal conductivity and early age hydration temperature rise were carried out on concretes with different content of thermal energy storage aggregates replacing normal aggregates. Finally, the thermal performance was determined on concretes containing thermal energy storage aggregates when being exposed to a severely variable temperature environment by using a self-designed setup.

2. Materials and experimental details

2.1. PCM preparation

Three fatty acids with analytical purities were used to prepare a composite PCM. They are lauric acid ($C_{12}H_{24}O_2$), myristic acid ($C_{14}H_{28}O_2$) and palmitic acid ($C_{16}H_{32}O_2$) with melting temperature of around 26 °C, 50 °C and 64 °C and latent

heat storage capacity of approximately 184 kJ/kg, 204 kJ/kg and 185.4 kJ/kg respectively [24]. These ingredients were weighted at different ratios by mass and then mixed for 10 min after all materials were completely molten at 80 °C. Finally, a homogenous mixture was obtained as the ternary composite PCM. The thermal properties of prepared PCMs were determined using a TG/DTA instrument under nitrogen atmosphere in the temperature range of 0–40 °C and at a heating and flow rate of 5 °C/min and 50 mL/min.

Due to the low thermal conductivity of 0.14–0.17 W/(m·K) for fatty acids, a graphite powder with a thermal conductivity of 50.63 W/(m·K) was added by 1%, 3%, 5%, 7% and 9% of PCM weight respectively. The graphite added PCM was poured into two $150 \times 150 \times 50 \text{ mm}^3$ specimens for thermal conductivity measurement and the detailed method was given in Part. 2.3.

2.2. Macro encapsulated PCM-lightweight aggregate

Two types of lightweight aggregates with size of 5–20 mm, ceramsite (expanded clay) and diatomite particles as shown in Fig. 1, were used as containers for PCM. The pore structure of them were characterized by scanning electron microscope (SEM) and mercury intrusion porosimetry (MIP). A simple direct impregnation method was used in this study to prepare the macro encapsulated PCM-lightweight aggregate. First, lightweight aggregate particles were dried at 105 °C for 24 h. The solid PCM was melted into a homogeneous liquid at temperature of 80 °C and then added lightweight aggregate particles. The mixture was frequently mixed for 3 h to accelerate the dispersion of PCM into the pore system of aggregate. Finally, the PCM adsorbed aggregate was obtained after cooling at room temperature. To form a protective shell for lightweight aggregate particles containing PCM, a spraying glue Guerqi 901 (made in China) was sprayed twice with interval of 30 min at room temperature. This glue is widely used in building decoration, having a strong adhesion, flexible operation and good resistance to high temperature up to 75 °C and low temperature down to –35 °C. After complete hardening of the glue, the aggregate particles were wetted by mist spray and covered by a thin layer of cement for 7 days of moist curing. After these treatment, the macro encapsulated PCM-lightweight aggregates were prepared as shown in Fig. 2. A strong protective layer formed on aggregate particle surface for avoiding PCM leakage and preventing particles from being broken during concrete manufacture.

The PCM encapsulation amount in aggregate pores was evaluated by a simple weighting method. And the leakage of PCM from macro encapsulated aggregate was determined by the mass loss after cycles between 10 °C and 60 °C. Chemical structure of the form stable composite PCM was analyzed by using FT-IR spectrometer. The spectra were recorded on a Nicolet 60 SXB FTIR Spectrophotometer as KBr pellets with resolution of 4 cm^{-1} in the range of $400\text{--}4000 \text{ cm}^{-1}$.

2.3. Concrete mixture and basic properties measurement

One normal gravel aggregate concrete was designed as the control mixture in Table 1. The raw materials include: ordinary Portland cement with strength grade of 42.5 complying with the Chinese Standard GB175 (corresponding to ASTM C150 [25]), river sand with fineness of 2.65, crushed gravel with particle sizes ranging from 5 to 20 mm, and a commercially available naphthalene-based superplasticizer (SP) with a recommended usage of 0.6–1.0% and water reduction ratio of 25%. Based on the gravel concrete, two types of thermal energy storage aggregates (ceramsite and diatomite) were incorporated by 20%, 40%, 60%, 80% and 100% in volume to replace gravel respectively. Therefore, 11 concrete mixtures in total were prepared in this study.

For every concrete mixture, the slump test was carried out to evaluate the workability of fresh concrete. $150 \times 150 \times 150 \text{ mm}^3$ cubic specimens were cast in room temperature, demold at 1-day age and then stored in standard curing condition (>RH 90%, $20 \pm 2^\circ\text{C}$) until 3 days or 28 days age for compressive strength test according to Chinese standard GB/T 50081 (corresponding to EN 12390-3 [26]). At the same time, specimens with size of $150 \times 150 \times 75 \text{ mm}^3$ were prepared with the same procedure. After 28 days age, the specimens were then oven-dried at 40 °C for 7 days to remove free water in capillary system of concrete. The thermal conductivity for every mix was conducted at room temperature by using TC3000E thermal conductivity measurement device (made from Xi'an Xiotech Electronics Co., Ltd, China). This testing operation was performed according to Chinese standard GB/T 10297 (corresponding to ASTM D5470 [27]).

2.4. Early age hydration temperature rise of concrete

To evaluate the influence of PCM-lightweight aggregate on the cement hydration induced temperature rise of concrete at early age, which potentially leads to cracking of massive concrete structures [28], a semi-adiabatic experimental setup was developed as shown in Fig. 3(a). Fresh concrete (1 L in volume) after mixing procedure was immediately injected into a vacuum thermos and then sealed by a polystyrene board plug. A T-type thermocouple was embedded in concrete sample and the data acquiring line passed through a small hole at the center of the polystyrene board plug. To avoid the heat exchange between concrete sample and outer environment air, a layer of polyurethane foam was sprayed around the top part of vacuum thermos as shown in Fig. 3(b). The sealed vacuum thermos was finally

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