Development of building materials by using micro-encapsulated phase change material

See Hoon Lee, Sang Jun Yoon, Yong Gu Kim, Young Chan Choi, Jae Ho Kim and Jae Goo Lee[†]

Korea Institute of Energy Research, Daejeon 305-343, Korea (Received 26 August 2006 • accepted 21 October 2006)

Abstract–Micro-encapsulated phase change material (Micro-PCM) could be used for high density thermal energy storage and also for PCM-building materials. Micro-PCM was prepared by *in-situ* polymerization at 60 °C after the emulsifying step by using 5 wt% Styrene Maleic Anhydride (SMA) solution as an emulsifier. The particle size of Micro-PCM decreased and the size uniformity was improved with an increase of the mixing intensity during emulsification. The average diameter of Micro-PCM was 5-20 µm. The thermal fluctuation of PCM-building materials such as gypsum and olefin film mixed with Micro-PCM was smoother and smaller than typical building materials without PCM.

Key words: PCM, Thermal Energy Storage, Building Material

INTRODUCTION

Latent heat storage material that is called as phase change materials (PCMs), has been a main topic in thermal energy storage (TES) for the last 20 years. With no agglomeration during PCM solidification, microcapsules can be used to store the latent heat storage material [1-4]. The thermal energy storage and heat exchange systems are mainly made up by using sensible heat materials such as water. In recent years, however, new materials for transportation as well as storage of energy by latent heat are under development such as ice slurries, micro-encapsulated phase change materials and micro-emulsion [5]. The new technologies are more effective than conventional systems that are only using sensible heat. Especially due to PCM segregation even during the phase change stage in microcapsule, the newly developed Micro-PCM have higher capacity of heat storage and transportation than the conventional ones [5-8]. Therefore, the system operation cost by using Micro-PCM might be lower than for a conventional system. And with the advent of PCM implemented in gypsum board, plaster, concrete or other wallboard material, thermal storage can be a part of construction technology for light weight buildings [9].

In the micro-encapsulation process, the emulsifier as a modifier used for forming shell structure was induced to be separated from a new, viscous, polymer-rich phase by adding a nonsolvent or a second polymer, lowering the encapsulating temperature, controlling pH, and other operating conditions. It is essential to control conditions that would cause the polymer to come out of the solution and to aggregate around the core droplet to form the microcapsule shell [5].

In the present study we used hexadecane or octadecane as core materials and melamine-formaldehyde as a shell material for the production of Micro-PCM for PCM-building material. And the characteristics of Micro-PCM and the heat storage capacity of PCMbuilding materials were determined.

[†]To whom correspondence should be addressed.

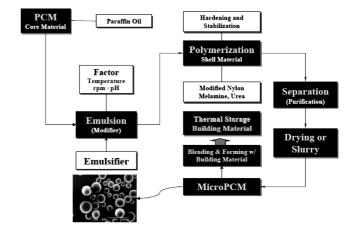


Fig. 1. Processes of Micro-PCM production.

EXPERIMENTAL

To prepare the Micro-PCM, in-situ micro-encapsulation process was used as shown in Fig. 1. Hexadecane ($C_{14}H_{36}$) or octadecane $(C_{18}H_{38})$ was used as a core material having melting temperature of 23 °C and 26 °C, respectively, and melamine-resin was used as a shell material. At the first process core material was emulsified in the solution which was prepared by homomixer (ME100LC, ROSS). Also melamine-formaldehyde (MF) prepolymer was made by heating an aqueous MF mixture at pH of 8.0-8.5 and 60 °C for 1 h. Melamine reacts with up to six molecules of formaldehyde under slightly alkaline conditions to melamine methylol derivatives that contain six methylol groups per melamine molecules. When heated, the methylol-melamine is condensed to form a crosslinked structure [5]. A known amount of SMA was dissolved at 60 °C to make 5 wt% solution, and then cooled down to room temperature. SMA solution was mixed with a mixture of hexadecane, octadecane and water for 10 minutes by homomixer. To control the capsule size and to make stabilized droplets, the speed of homomixer was regulated from 3,000 to 10,000 rpm. Finally, emulsified core material was mixed into the acidified aqueous melamine-formalin (MF) prepolymer solution. The polymerization was carried out at 60 °C over 3 h to

E-mail: jaegoo@kier.re.kr

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form a highly crosslinked membrane of the each droplet of the core material.

To evaluate the heat storage characteristics of the building materials, gypsum wallboard and the olefin film containing Micro-PCM were produced as shown in Fig. 2. The PCM gypsum wallboards were made in a mold $(0.3 \text{ m} \times 0.3 \text{ m} \times 0.015 \text{ m})$ by mixing gypsum

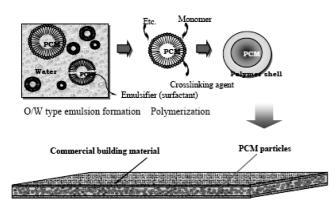


Fig. 2. Concept diagram of building materials containing Micro-PCM.

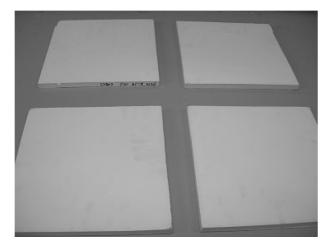


Fig. 3. Photograph of PCM gypsum wallboards.

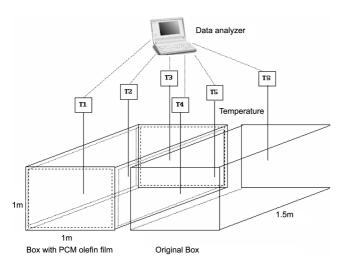


Fig. 4. The experimental chambers with PCM olefin films.

with water and PCM particles, of which weight fractions were 10, and 20 wt%. A photograph of the PCM gypsum wallboards was shown in Fig. 3 and the thermal conductivity of various gypsum wallboards was determined. To compare heat storage capacity, olefin films with PCM particles and test chambers were used and shown in Fig. 4. The olefin film with PCM was attached on the simulation chamber wall and its temperature profiles were T-1 to T-3. In a reference chamber with no PCM film,, the temperature profiles were T-4 to T-6. The experiment was carried out by heating the inside box 30 °C with 200 W heating elements for 30 minutes and cooling down.

RESULTS AND DISCUSSIONS

A highly effective MicroPCM slurry system was designed and constructed by *in-situ* polymerization process. For optimizing the process conditions, the *in-situ* polymerization were conducted with varying operation parameters such as emulsification speed, mixing ratio, surfactants, reactant ratio (M/F), polymerization time, temperature in react FT-IR analyzer. The optimum reactant ratio (M/F) was found to be 1 : 3 [4]. The particle size distributions of Micro-PCM with various conditions are shown in Fig. 5. As can be seen, the mean particle size of Micro-PCM decreases with increasing emulsification time.

As the concentration of emulsifier is increased, the mean particle size shows a minimum value at 4-5 wt% at 9,000 RPM and changes differently with the emusification speed. Also, the agglomeration or break of the microcapsule occurs at below 2 wt% of emulsifier concentration; therefore, a low concentration of emulsifier causes difficulty of encapsulation. In the SEM pictures of Lee et al. [4] and Khudhair and Farid [9], the shape of single Micro-PCM is almost spherical. From the analysis of particle size (FRITSCH), the average diameter of produced capsules is found to be 5-20 μ m as shown in Fig. 6.

A typical result of DSC (DSC550, Instrument Specialists, Inc.) analysis of PCM sample is shown in Fig. 7 where two peaks appear due to the endothermic process. One peak appears at around 1.75

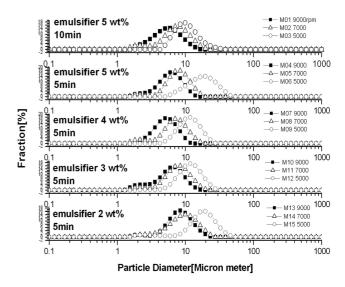


Fig. 5. Particle size with emulsifier and emulsification speed.

Korean J. Chem. Eng.(Vol. 24, No. 2)

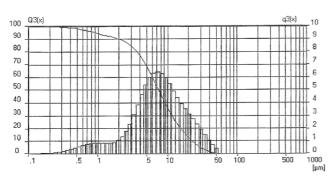


Fig. 6. Particle size distribution of Micro-PCM sample.

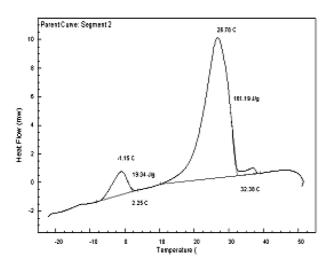


Fig. 7. Typical DSC analysis of Micro-PCM sample.

Table 1. Thermal conductivity of PCM gypsum wallboard

Sample	Thermal conductivity W/m·K
Raw materials	0.144
PCM gypsum wallboard (K19, 10 wt%)	0.163
PCM gypsum wallboard (K19, 20 wt%)	0.150
PCM gypsum wallboard (C18, 10 wt%)	0.128
PCM gypsum wallboard (C18, 20 wt%)	0.144
PCM gypsum wallboard (J18, 10 wt%)	0.128
PCM gypsum wallboard (J18, 20 wt%)	0.138

°C, that may be caused by the change of thermal flow due to the co-nucleating agent and the other peak appears at 28 °C due to phase change of the core materials. As the Micro-PCM diameter is increased, peak temperature of the core material increases because the thickness of shell affects the heat transfer [4]. The latent heat of Micro-PCM samples is 210 J/g (23 °C), 200 J/g (24 °C) and 150 J/g (28 °C) with the core materials respectively. So, a Micro-PCM with higher latent heat capacity could be used to increase the heat capacity of building materials.

The heat storage characteristic of PCM building applications, such as gypsum wallboard, film and so on, was determined. The thermal conductivities of the various PCM gypsum wallboards in Fig. 3 were analyzed by using the KS L 9016: 2005 method and

the results are shown in Table 1. Thermal conductivity of the gypsum wallboard without PCM is 0.144 W/m·K, but that of PCM gypsum wallboards is between 0.128 W/m·K and 0.163 W/m·K. As the weight fraction of PCM particles in gypsum wallboard increases, the thermal conductivity increases decrease except K19 PCM. This result is due to the decrease of heat flux between gypsum wall thickness from almost constant temperature during phase change. Therefore, the thermal storage performance of PCM wallboard increases with increasing the weight fraction of PCM. Neeper [3] found that thermal storage provided by the PCM wallboard is sufficient to store large solar heating. The PCM wallboard shows tenfold increase in capacity of storage and heat discharge compared with the original gypsum wallboard alone [10,11].

In the heat storage experiments of olefin film using PCM particles, temperature profiles of the experiment boxes $(1.5 \text{ m} \times 1 \text{ m} \times 1 \text{ m})$ are shown in Figs. 8 and 9. The PCM concentration in olefin film of these experiments is 100 g/m² and the film thickness was 3.0 mm and 9.0 mm, respectively. As can be seen in Fig. 8 (outside temperature: 10 °C), the time of temperature drop from 30 °C to 20 °C in the simulation box is 94 min. In the ordinary box, temperature drop time is only 20 min. So the temperature drop time of

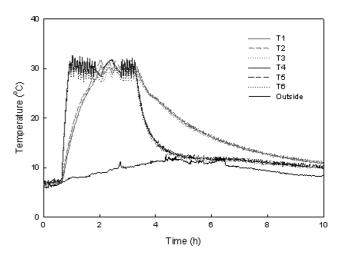


Fig. 8. Temperature profiles of 3 mm PCM olefin film.

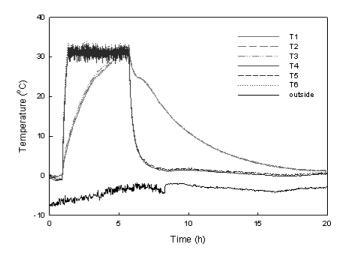


Fig. 9. Temperature profiles of 9 mm PCM olefin film.

CONCLUSIONS

Micro-PCM was prepared by using *in-situ* polymerization for PCM building materials. The average particle size of Micro-PCM was between 5 and 20 µm. As the concentration of emulsifier is increased, the mean particle size shows a minimum value at 4-5 wt% at 9,000 RPM and changes differently with the emusification speed. Also, the agglomeration or break of the microcapsule occurs at below 2 wt% of emulsifier concentration; therefore low concentration of emulsifier causes difficulty of encapsulation. The latent heat of Micro-PCM samples is 210 J/g (23 °C), 200 J/g (24 °C) and 150 J/g (28 °C) with the core materials respectively. The thermal conductivity of the gypsum wallboard without PCM is 0.144 W/m·K, but that of the PCM gypsum wallboard is between 0.128 W/m·K and 0.163 W/m·K. As the thickness of PCM olefin film increases, the capacity of ther- mal storage inside chamber increases.

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