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Production of Microencapsulated Phase Change Material by Pilot-scaled Spray Dryer

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Abstract

The use of phase change materials (PCM) as thermal energy storage is one of the passive cooling strategies in building application because PCM can absorb and release massive energy in term of latent heat during phase transition in a small temperature range. Microencapsulation is a process which forms the containment of PCM to prevent the leakage of PCM due to the phase transition between solid and liquid that can be operated by spray drying technology for the microencapsulated PCM powder production. In this research, scaling up of spray drying process from laboratory scale to pilot scale was studied to produce more quantities of PCM powder for higher demand for building application. Methyl palmitate as PCM was encapsulated by silica microcapsules. Two spray drying systems; B-290 (Büchi Labortechik AG, Switzerland) and MOBILE MINOR™ (GEA spray drying, Denmark) were used in the laboratory scale and the pilot scale, respectively. The method is divided into two steps; atomization and drying. In the atomization step, operating condition resulting in matching droplet size was determined. The drying step, process parameters such as drying air flow rate, feed flow rate and temperature in a drying chamber were determined with the key assumption that the constant drying rate per solid content in both scales can produce PCM powder with similar characteristics such as particle size distribution, thermal property, and moisture content. From the experiments, spray dryer in pilot scale can produce a double amount of PCM powder and it also has similar properties compared with laboratory scale.

Keywords: Microencapsulation, Phase change material, Sol-gel synthesis, Pilot-scaled spray dryer, Scale-up methodology

1. Introduction

The increasing energy demand consumes a large amount of fossil fuels that cause climate change and decrease fossil fuels more rapidly. Due to energy prices and environmental conservation, low energy strategy is considered as the goal of world organizations and many countries. From energy consumption in the European Union, the building sector is a primary energy consumer with around 40% of the total energy usage. Large parts of this energy usage are directly used for the temperature control of buildings due to the increase of residential buildings, climate change and higher average temperatures in recent years (Artmann, N., H. Manz, and P. Heiselberg, 2017; Santamouris, M., 1996). To reduce energy demand as well as enhance indoor temperature comfortably, passive cooling strategies have been involved in building (Farid, M.M., 2004). Therefore, thermal energy storage (TES) is one of passive cooling strategy aimed to prevent the increase in indoor temperature in buildings by solar reflectance. Phase change material (PCM) is a latent heat storage material used to absorb and release massive energy in the form of latent heat during phase transition (Farid, M.M., 2004; Sharma, A., 2009).

PCM designed for building applications was used as a liquid phase. Due to the phase transition between solid and liquid, the use of PCM directly may cause the leakage when the materials change from solid to liquid and reducing thermal conductivity. So, Direct incorporation and immersion of PCM in building materials have low latent heat about 35-56 J/g and are not well suited for long-term applications (Feldman, D., D. Banu, and D.W. Hawes, 1995). This can be achieved via the microencapsulation process which forms a coating material around a small droplet of PCM.

There are some researches about the application of microencapsulation of PCM in building materials for thermal energy storage. The experiment conducted by Lei et al. (Lei, J., 2017) showed that PCM applied to skim coat and normal paint for a passive cooling strategy that could absorb the heat and reduced the surface and air temperature. Also, it can achieve the annual energy saving rate throughout the entire year in a local location. In our preliminary experiments, the thermal performance of PCM microcapsules was tested and compared the results with previous researches. One major concern during the experiments is the large quantity of PCM microcapsules. The production of microcapsules in laboratory scale spray dryer can produce in small units that cannot meet the demand for the use for thermal energy storage in building the application. So, pilot-scale spray dryer becomes an interesting way to produce a larger quantity and improve the production of PCM microcapsules.

The aim of this work is to enhance the production of PCM microcapsules from laboratory scale to pilot spray dryer, and to investigate the key assumption to confirm that the constant drying rate per solid content in both scales can produce PCM powder with similar characteristics.

2. Experimental

2.1. Materials

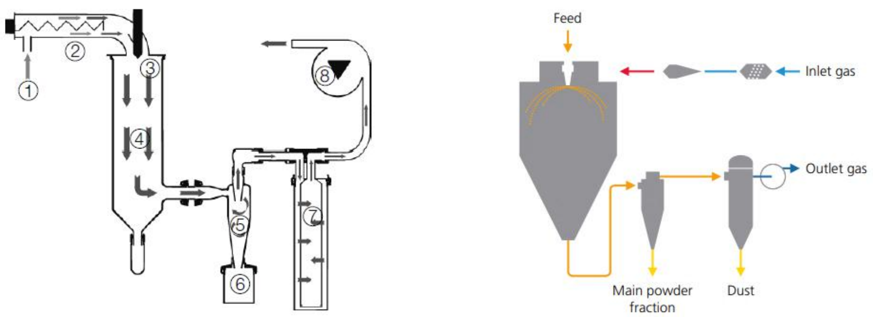
In the microencapsulation process, tetraethyl orthosilicate (TEOS, 99% pure) from Sigma-Aldrich Company Ltd. was used as shell materials for packing PCM in capsules. Methyl palmitate from SAFC (US) was used as PCM for the thermal energy storage. Hydrochloric acid (HCl) was used as an acid catalyst. Cetyl trimethyl ammonium bromide (CTAB) from Ajax Finechem (Australia) was used as the surfactant. All chemicals were used without further purification.

2.2. Microencapsulation Process

Microcapsules containing PCM were prepared by the microencapsulation technique through a sol-gel process that involves two phases: a continuous phase (water phase) and discontinuous phase (oil phase). An encapsulation method is described as follows: Shell materials solution as the continuous phase was prepared by mixing TEOS and deionized water by 1:2 ratio. Then, HCl was added into the solution and stirred at 40 °C unit the homogeneous solution was obtained, indicating that the hydrolysis of TEOS was completed. In the discontinuous phase, PCM was added into an aqueous solution containing CTAB dissolved in deionized water with a ratio of 0.1:1 (CTAB: PCM). The mixture was added into the prepared sol-gel solution to form oil-in-water emulsion by a homogenizer (Model T-25D Ultra-Turrax, IKA, German) with a speed of 6,000 - 8,000 rpm for 3 - 5 minutes. The emulsion solution was made into dry microcapsules by spray drying.

2.3. Spray Drying Process

The laboratory-scale experiments were carried out by a Mini Spray Dryer B-290 (Büchi Labortechik AG; Switzerland) with the capability of generating particles in 2-25 microns. The atomizer type was a two-fluid nozzle with nozzle cleaning function. A peristaltic pump built in the instrument was used to pump the feed to the atomizer. The drying process was carried out in a co-current mode using hot air at a rate of 40 kg/h. This means that the microcapsule product and hot air have the same flow direction. A schematic flow direction of hot air in this spray dryer is shown in Fig.l(a).
The pilot-scale experiments were carried out using a MOBILE MINOR™ spray dryer (GEA Niro A/S, Denmark) in an open- mode design. Particle size can be produced in a 2-80 microns range and have longer the hot air residence time. The atomizer type was a Niro Mobile Minor Rotary Atomizer. The feed was supplied to the atomizer by a peristaltic pump. The drying process was carried out in a co-current mode using hot air at a rate of 90 kg/h. the sprayed product was collected under the cyclone. A schematic flow direction of hot air in the Mobile Minor spray dryer is shown in Fig.l(b).



(a) (b)

Fig.1 A schematic flow direction of hot air in this spray dryer (Cordin Arpagaus, Henrik Schwartzbach, 2008) (a) Mini Spray Dryer B-290 (b) MOBILE MINOR™ spray dryer.

2.4. Characterizations

Characterization of the thermal properties of PCM, including phase change temperature and latent heat, were measured by differential scanning calorimeter (DSC) scanned in the temperature range -10 - 60 C. The phase change temperature range was considered as the range between the onset and end temperature of phase transition peak on DSC curve, and latent heat was determined by numerical integration of the area of the phase transition peak.
The particle size distribution of the PCM microcapsules was analyzed by means of laser diffraction using a Malvern Mastersizer S (Malvern Instruments, Worcestershire, UK). An ultrasonic water bath was used to disperse the particles for 3 minutes. The particle size was given by the volume median diameter of D50. The refractive index of silica and water are 1.455±0.01i and 1.33, respectively.

Fourier Transform Infrared Spectroscopy (FT-IR) were obtained using a Nicolet 6700 FT-IR Spectrometer (Thermo Scientific, US) on a KBr sampling to evaluate the functional group on PCM microcapsule.

3. Results and Discussion

3.1. Preparation and Properties of Feed Solutions

The spray solution prepared by the microencapsulation technique through the sol-gel method should not change in properties before spraying into both scale of the spray dryer. So, chemical stability and physical stability were investigated to confirm the properties of the feed solution. Fig.2(a) shows the FT-IR spectra of TEOS before the hydrolysis reaction in the sol-gel method. In the spectrum, the C-H bonding vibrations at 1366-1442 cm^{-1} and the C-H stretching at 2891-2976 $cm^{-1}$are due to methyl and methylene groups on TEOS structure. In addition, the absorption peak at 1108 and 463 cm^{-1} are attributed to asymmetric and symmetric Si-O-Si stretching vibration of silica precursor. The spectra band at 965 cm^{-1} corresponds to Si-OH vibrations. Fig.2(b) shows the FT-IR spectra of the homogeneous solution after the hydrolysis of TEOS was completed. Compared with Fig.1, there are additional peaks in the spectrum of hydrolysis solution, in which the peaks at 3436 and 1637 cm^{-1} are respectively attributed to Si-OH stretching vibrations and H_2O group. It found that the C-H bonding and stretching vibration are disappeared which confirm the successful of hydrolysis reaction in sol-gel method to form the silica-sol solution.

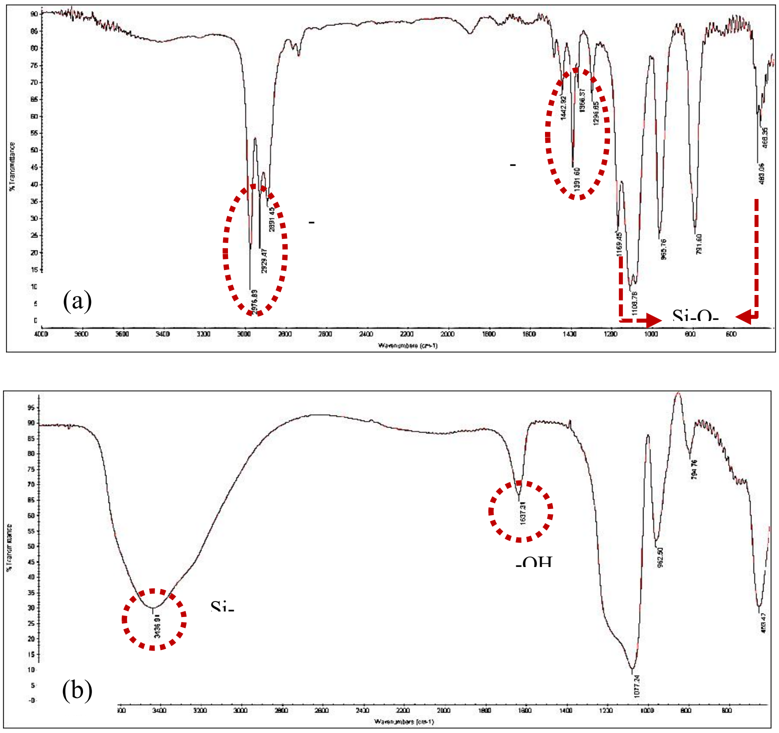


Fig. 2 FT-IR spectra of (a) TEOS solution (b) The homogeneous solution after completely hydrolysis reaction in sol-gel method.

3.2. Spray Drying Process in Laboratory Scale

All characteristics of PCM microcapsules generated in laboratory and pilot-scale spray dryer will be presented in Table 1. Table 1 provides solid content conditions in feed preparation together with the measured particle size and the thermal performances of PCM microcapsules. The spray drying conditions of a laboratory scale, based on previous studies (Cordin Arpagaus, Henrik Schwartzbach, 2018), were calculated based on the key assumption of the constant drying rate per solid content to matching similar characteristics. In laboratory scale, the results indicated that the mean particle size (D_{50}) of 11% solid content is 8.3 µm. The phase change temperatures (T_m and T_c) showed the exothermic and endothermic peak at 22.3 and 29.2 °C, respectively, and latent heat of cooling (ΔH_c) and melting (ΔH_m) are 75 and 76 J/g, respectively. Considering the effect of increasing solid content, microcapsules size varied between 8.3 and 10.4 µm that slightly increased because of the more PCM inside the microcapsules. The phase change enthalpies (ΔH_m and ΔH_c) were found to be unchanged with varying solid content. These results indicated that the operating conditions in laboratory scale calculated based on the key assumption are acceptable.

Table 1. The solid content in feed preparation and the measured properties of microcapsules.

Tests	Scale	Solid content (%wt)	D_{50} (µm)	T_{melt} (°C)	ΔH_m (J/g)	T_{cool} (°C)	ΔH_c (J/g)
1		11	8.3	29.2	77.7	22.3	75.5
2	Laboratory	15	9.3	30.4	77.9	24.6	77.9
3		20	10.4	31.5	79.9	24.7	77.4
4		11	8.6	28.7	79.4	22.7	77.8
5	Pilot	15	9.8	29.3	76.7	14.7	71.3
6		20	9.5	29.6	82.3	13.4	79.5

3.3. Spray Drying Process in Pilot Scale

In the atomization step, feed flow rate and atomization condition are affected droplet size and subsequent the final microcapsules after drying. As an increasing feed flow rate and different atomizer type, the atomization process requires increasing energy input to obtain the same particle size. The energy input is provided from increasing atomizer air pressure used for rotary atomizer. Fig.3 shows the correlation between particle sizes at varying atomizer air pressure. It indicated that the particle sizes decreased with an increase in atomizer air pressure. So, 5 bar of air pressure is suitable for use as an operating condition in pilot-scale.

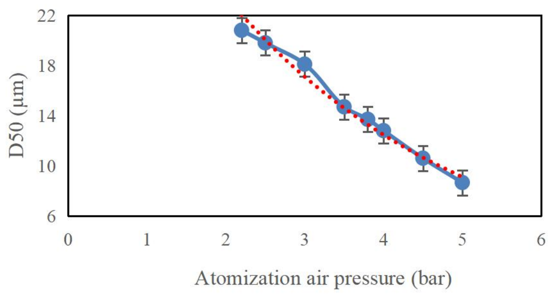


Fig.3 The correlation between particle sizes at varying atomizer air pressure.

In the drying process, a thermodynamic mass and energy balance equations are developed to find operating conditions that can generate PCM microcapsules with similar characteristics such as particle size distribution, thermal property, and moisture content. The calculation of both balance models was also related to the key assumption of the constant drying rate per solid content. The product from pilot scale was characterized as shown in Table 1. The results indicated that microcapsules produced in pilot scale were similar size when compared with the same solid content in laboratory scale. Besides, the microcapsules size slightly increased when solid content increased. This trend was already observed in laboratory results. Moreover, the effect of increasing PCM inside the microcapsules can increase phase change enthalpies and the maximum enthalpy was from 20% solid content at 82.29 and 79.54 J/g of latent heat of cooling (ΔH_c) and melting (ΔH_m), respectively.

4. Conclusions

The production of microencapsulated PCM via the sol-gel method and subsequently spray drying was enhanced from laboratory scale to pilot scale spray dryer. The experiments were designed to confirm the key assumption that the constant drying rate per solid content in both scales of spray dryer can produce PCM powder with similar characteristics. The experimental results indicated that the key assumption is successful to remain the particle properties such as particle size distribution and thermal property. Additionally, an increase in solid content can increase both particle size and phase change enthalpies of PCM microcapsules.

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