

# Preparation of Phase Change Materials Microcapsules by Using PMMA Network-Silica Hybrid Shell Via Sol-Gel Process

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Received 18 November 2007; accepted 17 November 2008

DOI 10.1002/app.29742

Published online 11 February 2009 in Wiley InterScience (www.interscience.wiley.com).

**ABSTRACT:** Encapsulation of phase change materials (PCM) using a poly(methyl methacrylate) network-silica hybrid as the shell material has been developed. *n*-Octadecane melted at 28°C was used as PCM. Based on the suspension polymerization process, the microcapsules were prepared successfully by mixing and by the reaction of ethylene glycol dimethacrylate with precopolymer solution with tetraethoxysilane (TEOS), whose resultant microcapsules had higher latent heat ( $\Delta H = 151$  J/g) than those without TEOS ( $\Delta H = 88.3$  J/g). The average size of the PCM microcapsules was about 10  $\mu\text{m}$ . The silica content, *n*-octadecane content, and latent heat of microcapsules were changed with varying ageing conditions, ageing time,

and temperature. The highest amount of latent heat ( $\Delta H = 178.9$  J/g) and *n*-octadecane content (73.3%) of the microcapsule were obtained when the inorganic/organic ratio of the microcapsule was 5%. It was difficult to increase *n*-octadecane content (74% to 55.7–67.9%) and latent heat (180.5 J/g to 135.9–165.7 J/g) of the microcapsules by introducing different functional groups of coupling agents. © 2009 Wiley Periodicals, Inc. *J Appl Polym Sci* 112: 1850–1857, 2009

**Key words:** microencapsulation; phase change material; radical polymerization; ageing; *n*-octadecane; hybrid materials; sol-gel process

## INTRODUCTION

Phase change material (PCM) is one of the most popular energy storage materials due to its relatively higher thermal capacity and storage density, and smaller temperature swing. However, there are also some drawbacks to it, such as the losses between phase changes especially in solid-liquid phase transformations. To reduce the loss and increase its applications, PCM microcapsules have been developed in recent years. In addition, encapsulated PCMs are able to increase the area of heat transfer, reducing reactivity with the outside environment and easily controlling the volume of storage material during phase changes.<sup>1</sup>

Interfacial polymerization and *in situ* polymerization are the most widely used methods for the encapsulation of PCM. For interfacial polymerization, the reaction occurs at the interface between the oil and water phases. Toluene-2,4-diisocyanate (TDI) has been used as an oil-soluble monomer and several water-soluble monomer have also been used, such as

diethylenetriamine (DETA),<sup>2,3</sup> ethylenediamine (EDA),<sup>4</sup> 1, 6-hexane diamine (HDA), or their mixture to encapsulate *n*-hexadecane or *n*-eicosane.<sup>5</sup> For *in situ* polymerization, water-soluble monomers, such as melamine,<sup>6–10</sup> gelatin,<sup>11</sup> or urea<sup>12,13</sup> have also been used. Formaldehyde or glutaldehyde were used as a crosslinker. The polymer produced was precipitated on the surface of oil droplets forming the encapsulating PCM. However, the melamine-formaldehyde, gelatin-formaldehyde, and urea-formaldehyde shell microcapsules usually release poisonous formaldehyde in the application, hence limiting their application.

On the other hand, poly(methyl methacrylate) (PMMA) is a commercially available acrylic resin and has high impact strength and chemical resistance. These properties make it a potential encapsulation material for PCM.<sup>14</sup>

It can be recognized from the literature that many of the studies deal with the encapsulation of paraffin into a polymer network. Moreover, the studies related to PMMA networks are limited. This study, therefore, was aimed at shedding light on the preparation, characterization, and evaluation of the thermal properties of PCM microcapsules. *n*-Octadecane and PMMA network were used as the core and shell material, respectively. Silica prepared by sol-gel process was also

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introduced to PMMA network to improve the PCM content in microcapsules.

## EXPERIMENTAL

### Materials

*n*-Octadecane was purchased from Tokyo Chemical Industry Co., Ltd. (TCI). Methyl methacrylate (MMA) and ethylene glycol dimethacrylate (EGDMA) were purchased from Merck Ltd. Tetraethoxysilane (TEOS), methyl triethoxysilane (MTES), decyltrimethoxysilane (DTMS), and  $\gamma$ -methacryloxypropyltrimethoxysilane (MAPTMS) were purchased from Shin-Etsu Chemical Co. Vinyl triethoxysilane (VTES) was purchased from Acros Organics. 2,2'-Azobisisobutyronitril (AIBN) was purchased from Showa Chemical Co. Polyvinyl alcohol (PVA) was supplied from Chang Chun Chemical Co. All chemicals were used without further purification.

### Preparation of PMMA network shell PCM microcapsules

Three kinds of mixing solutions of *n*-octadecane (PCM) and MMA were used to prepare the suspension solution before polymerization.

#### Solution I—Mixing PCM and monomers directly

A total of 12.5 g of *n*-octadecane, 22.5 g of MMA, 2.5 g of EGDMA, and 3 g of AIBN were first mixed well as the oil-phase solution and then poured into 0.5 wt % of PVA aqueous solution to shear the two-phase system with homogenizer (IKA model T25) for 3 min at 9500 rpm. After which the suspension was heated up to 60°C and stirred vigorously at 300 rpm for 24 h. The PCM microcapsules were collected by filtration and washed with distilled water and *n*-hexane.

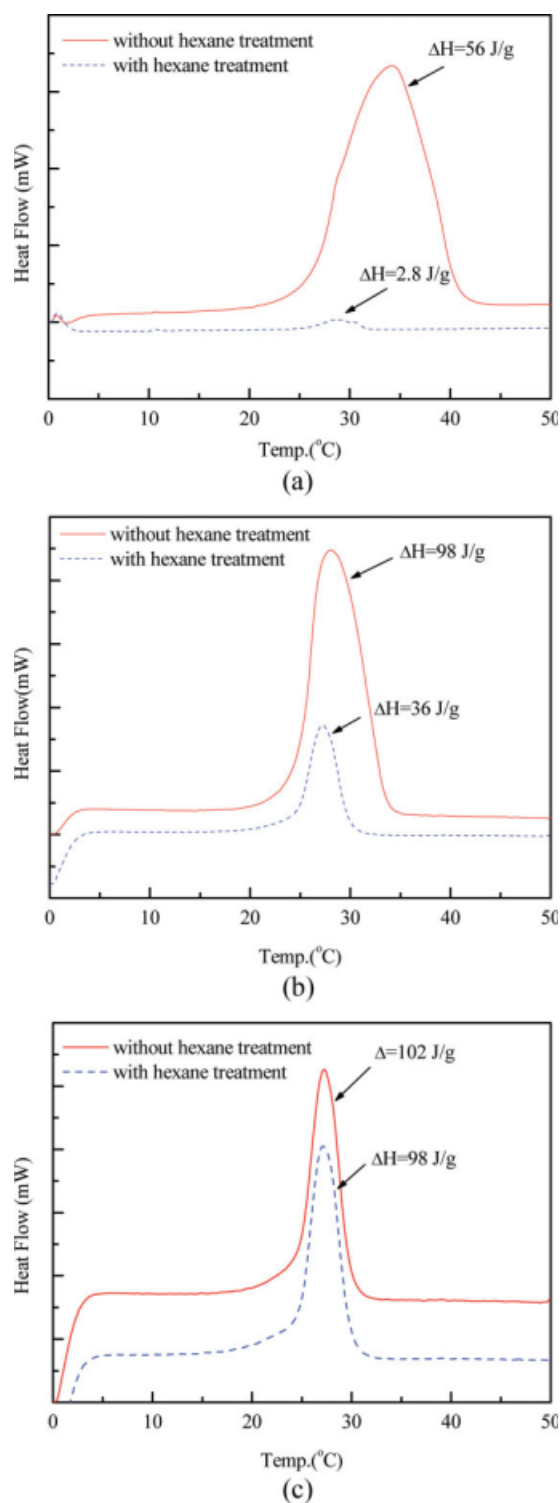
#### Solution II—Introducing PCM, then introducing monomers

In this method, *n*-octadecane was first homogenized for 1 min at 9500 rpm with 0.5 wt % of PVA aqueous solution to form the PCM suspension. MMA, EGDMA, and AIBN were mixed as the oil-phase solution and then poured into PCM suspension followed by homogenizing for 3 min at 9500 rpm. The suspension was then heated up to 60°C and stirred vigorously at 300 rpm for 24 h. The PCM microcapsules were collected by filtration and washed with distilled water and *n*-hexane.

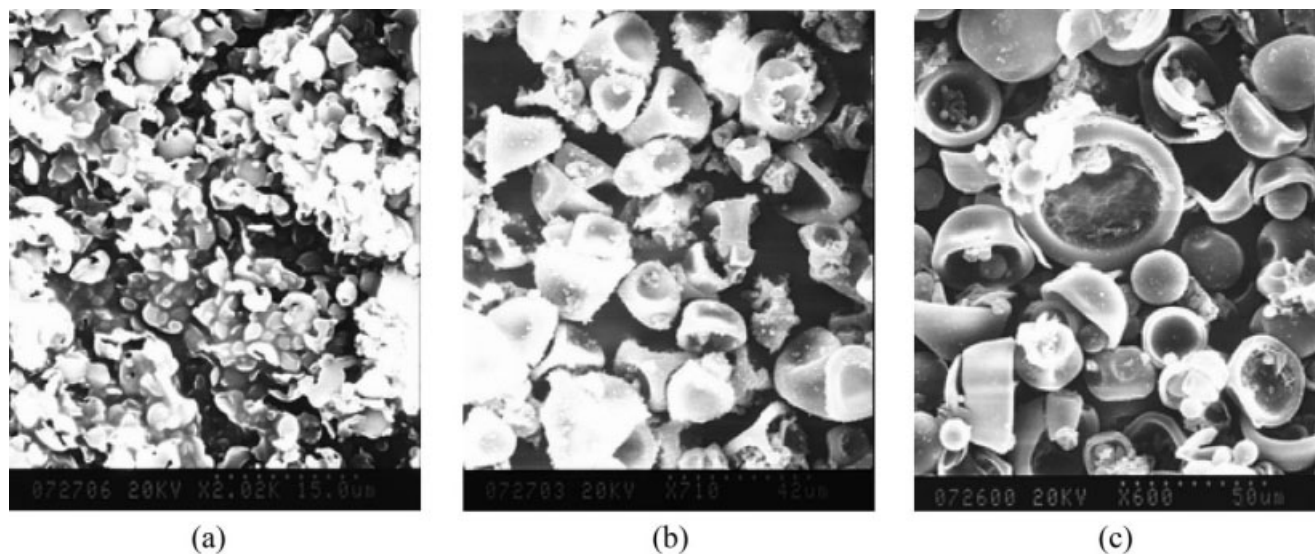
#### Solution III—Introducing PCM, then introducing monomers with prepolymer

*n*-Octadecane was first homogenized for 1 min at 9500 rpm with 0.5 wt % of PVA aqueous solution to

become the PCM suspension. EGDMA, AIBN, and prepolymer solution were mixed as oil-phase solution and then poured into the PCM suspension followed by homogenizing for 3 min at 9500 rpm.



**Figure 1** DSC diagrams of PMMA network shell PCM microcapsules prepared with (a) solution I, (b) solution II, and (c) solution III. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]



**Figure 2** SEM photographs of PMMA network shell PCM microcapsules (after hexane treatment) prepared with (a) solution I, (b) solution II, and (c) solution III.

Prepolymer solution was prepared by mixing 25 g toluene with 20 g MMA and 2.5 g EGDMA which then reacted at 60°C for 1 h and conversion of the prepolymer was 30–65% after reacting. The PCM content of the microcapsules would vary with different conversion of the prepolymer. After homogenizing, the suspension was heated up to 60°C and stirred at 300 rpm for 24 h. The PCM microcapsules were collected by filtration and washed with distilled water and *n*-hexane.

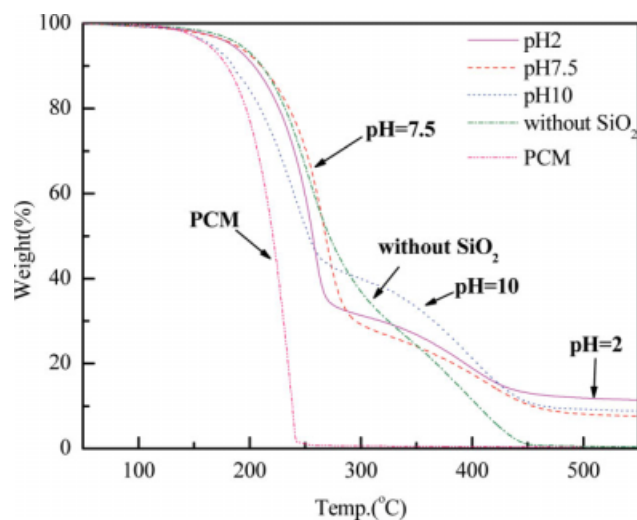
#### Preparation of PMMA network-silica shell PCM microcapsules

*n*-Octadecane was first homogenized for 1 min at 13,500 rpm with 0.5 wt % of PVA aqueous solution to become the PCM suspension. EGDMA, AIBN, TEOS, and prepolymer solution were mixed as the oil-phase solution which then poured into the PCM suspension by homogenizing for 3 min at 13,500 rpm. Prepolymer solution was prepared by mixing toluene, MMA, EGDMA, and then reacted at 60°C for 1 h and conversion of the prepolymer was 30–65% after reacting. After which the suspension was heated up to 60°C and stirred at 300 rpm for 24 h. The PCM microcapsules were collected by filtration and washed with distilled water and *n*-hexane.

#### Characterization of PCM microcapsules

Latent heat of the PCM microcapsules was determined by numerical integration of the area of the

differential scanning calorimeter (DSC; Perkin–Elmer DSC-7) peak at a rate of 10°C/min in a nitrogen atmosphere from 0 to 50°C. The curve of the weight loss ratio and temperature of the PCM microcapsules were measured by thermogravimetric analyzer (TGA, Perkin–Elmer TGA-7) at a rate of 20°C/min in the atmosphere of air from 50 to 550°C. The SiO<sub>2</sub> content was determined by the residues at 550°C of TGA curves. The surface morphology and diameter of the PCM microcapsules were observed by scanning electronic microscope (SEM, Hitachi S-4200).



**Figure 3** TGA diagrams of PCM and PMMA network/SiO<sub>2</sub> shell PCM microcapsules prepared with different pH value. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

**TABLE I**  
The Properties of PMMA Network Shell PCM Microcapsule and PMMA Network/SiO<sub>2</sub> PCM Microcapsules Prepared with Different pH Value

Microcapsule	pH	$\Delta H$ (J/g)	PCM content (%) <sup>a</sup>	SiO <sub>2</sub> content (%)	SiO <sub>2</sub> content in shell (%) <sup>b</sup>
PMMA	–	88.3	36	0	0
	2	151.0	64.7	10.5	29.7
PMMA/SiO <sub>2</sub>	7.5	129.3	55.4	7.2	16.0
	10	92.3	39.6	8.2	13.6

<sup>a</sup> PCM content = ( $\Delta H$  of sample)/( $\Delta H$  of *n*-octadecane).

<sup>b</sup> SiO<sub>2</sub> content in shell = (SiO<sub>2</sub> content)/[100–(PCM content)].

## RESULTS AND DISCUSSION

### Preparation of PMMA network shell PCM microcapsules

It is necessary to encapsulate PCM using a PMMA network as the shell to keep the PCM in the microcapsules when the ambient temperature is higher than the melting point of PCM. Three kinds of mixing solutions of PCM and MMA were used as the suspension solution before polymerization. Solution I was a suspension formed by mixing PCM and MMA directly. Solutions II and III were solution formed by introducing PCM in an aqueous solution. The MMA monomer and polymer solutions were introduced into the above solution, respectively. Figure 1 shows the DSC diagram of the PMMA network PCM microcapsule prepared using the three kinds of mixing solutions with or without hexane treatment. The latent heat of the PCM microcapsules prepared by Solutions I and II decreased significantly when the microcapsules were washed with hexane. This finding indicates that PCM was easily washed from the microcapsules by hexane when using these two kinds of solutions. The latent heat of the PCM microcapsules prepared with Solution III was almost not changed (98 J/g to 101 J/g) after the microcapsules were washed with hexane or other-

**TABLE II**  
The Properties of PMMA Network/SiO<sub>2</sub> Shell PCM Microcapsules with Different Ageing Period at Room Temperature

Day	$\Delta H$ (J/g)	PCM content (%) <sup>a</sup>	SiO <sub>2</sub> content (%)	SiO <sub>2</sub> content in shell (%) <sup>b</sup>
0	96.6	39.6	1.52	2.5
1	93.7	38.4	2.48	4.0
2	103.9	42.6	2.92	5.1
4	107.8	44.2	3.49	6.2
6	102.0	41.8	4.12	7.1

<sup>a</sup> PCM content = ( $\Delta H$  of sample)/( $\Delta H$  of *n*-octadecane).

<sup>b</sup> SiO<sub>2</sub> content in shell = (SiO<sub>2</sub> content)/[100–(PCM content)].

**TABLE III**  
The Properties of PMMA Network/SiO<sub>2</sub> Shell PCM Microcapsules with Different Ageing Period at 60°C

Day	$\Delta H$ (J/g)	PCM content (%) <sup>a</sup>	SiO <sub>2</sub> content (%)	SiO <sub>2</sub> content in shell (%) <sup>b</sup>
0	96.6	39.6	1.52	2.5
1	94.4	38.7	1.82	3.0
2	98.3	40.3	1.89	3.2
4	100.5	41.2	2.60	4.4
6	105.4	43.2	2.92	5.1

<sup>a</sup> PCM content = ( $\Delta H$  of sample)/( $\Delta H$  of *n*-octadecane).

<sup>b</sup> SiO<sub>2</sub> content in shell = (SiO<sub>2</sub> content)/[100–(PCM content)].

wise. It was found that the PCM content in the microcapsules prepared using Solution III was 40% and the PMMA network in microcapsules was 60%. The results show that the higher content of the PCM microcapsules could be prepared by using Solution III. SEM photographs of PCM microcapsules prepared by the three kinds of solutions with hexane treatment are shown in Figure 2. The core-shell structure may be observed and is shown in Figure 2(b,c) as prepared with Solutions II and III, respectively. The microcapsules prepared using Solution I [Fig. 2(a)] were cracked corresponding to the absence of PCM to sustain the structure of the microcapsules. The core-shell structure of the microcapsules was obtained using Solutions II and III. The smoother surface of microcapsules was especially observed using Solution III.

### Preparation of PMMA network-silica shell PCM microcapsules

To improve the dense structure of the PCM microcapsules and increase the PCM content in it, TEOS was added into the prepolymer solution. TEOS then reacted with water from the disperse phase by sol-gel process and PMMA network-silica shell PCM microcapsules were obtained.

**TABLE IV**  
The Properties of PMMA Network/SiO<sub>2</sub> Shell PCM Microcapsules with Different Ageing Period at 80°C

Day	$\Delta H$ (J/g)	PCM content (%) <sup>a</sup>	SiO <sub>2</sub> content (%)	SiO <sub>2</sub> content in shell (%) <sup>b</sup>
0	96.6	39.6	1.52	2.5
1	102.2	41.9	1.65	2.8
2	95.2	39.0	2.45	4.0
4	96.6	39.6	3.54	5.9
6	94.9	38.9	4.56	7.5

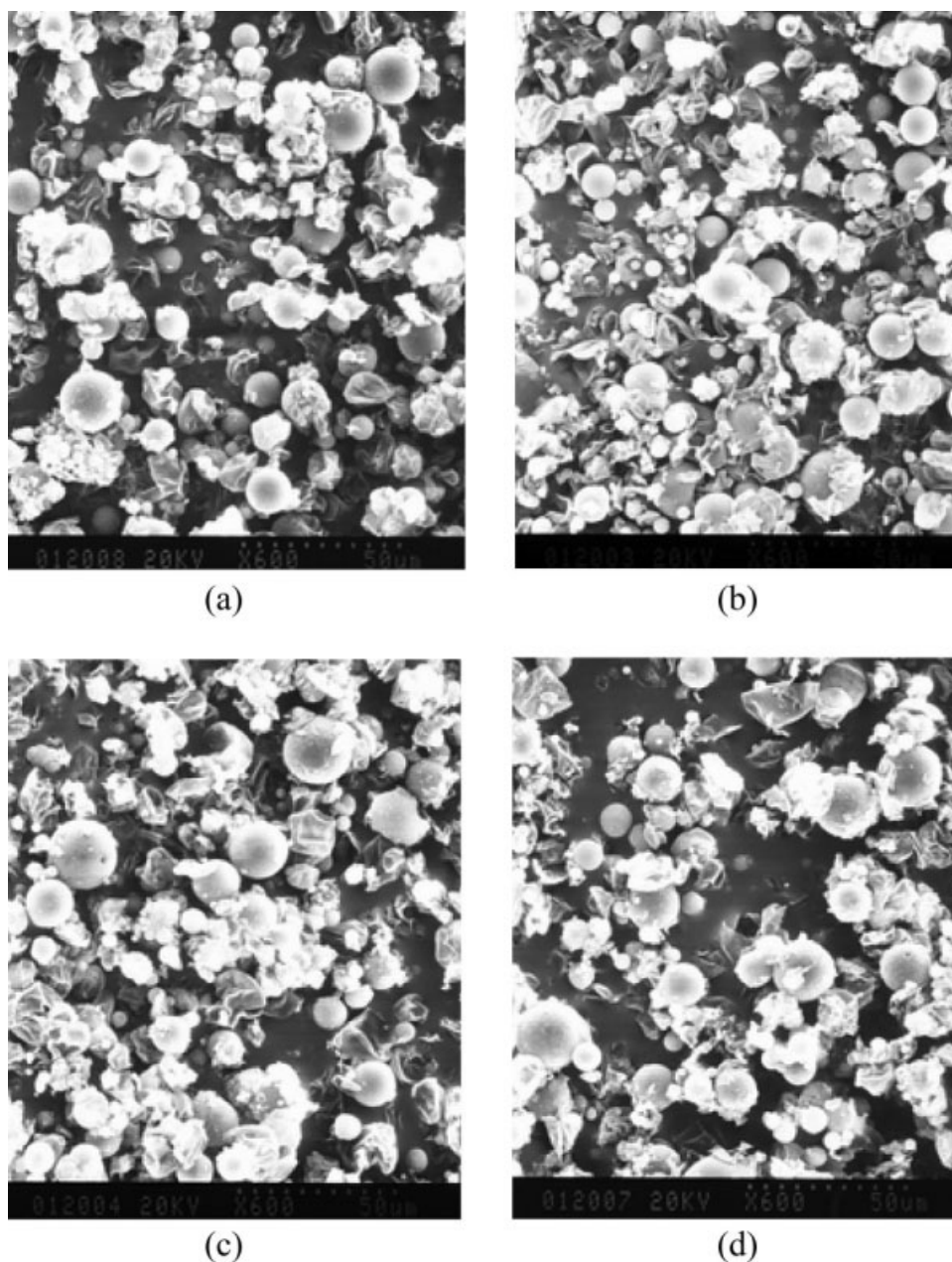
<sup>a</sup> PCM content = ( $\Delta H$  of sample)/( $\Delta H$  of *n*-octadecane).

<sup>b</sup> SiO<sub>2</sub> content in shell = (SiO<sub>2</sub> content)/[100–(PCM content)].

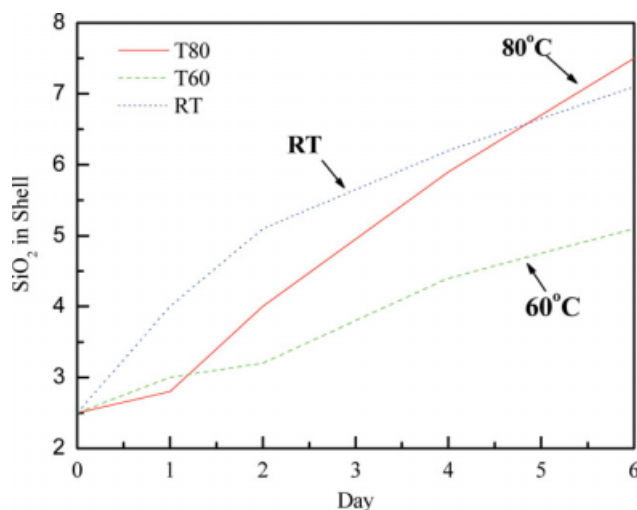
### Effect of pH value in water phase

The surface structure of the shells of the microcapsules was affected by the pH value in the sol-gel process. In the acidic sol-gel process, many hydroxyl groups were produced after the hydrolysis of silicon alkoxide and entangled linear chains were formed in sols, thus resulting in a dense network structure of the shell of the microcapsules. Three different pH values, 2, 7.5, and 10, were chosen in the preparation of the PCM microcapsules and there was 46% prepolymer in the prepolymer solution. The TGA diagrams of the PCM microcapsules prepared with

different pH values are shown in Figure 3. The findings indicate that the TGA curves shifted to higher temperatures when the PCM microcapsules were prepared with TEOS at any pH value. The latent heat, PCM content, and SiO<sub>2</sub> content of PCM microcapsules are listed in Table I. The latent heat of PMMA network-silica shell PCM microcapsules (92–151 J/g) is higher than that of the PMMA network shell PCM microcapsule (88 J/g). This finding means that the PCM content of PMMA network-silica shell PCM microcapsules (39.6%–64.7%) are higher than that of PMMA network shell PCM microcapsules (36%). In the sol-gel process, acid catalysis caused



**Figure 4** SEM photographs of PMMA network/SiO<sub>2</sub> shell PCM microcapsules at varying ageing period and temperatures: (a) 0 day, (b) 4 days, room temperature, (c) 4 days, 80°C, and (d) 6 days, 80°C.



**Figure 5** Effect of SiO<sub>2</sub> content in shell of PMMA network/SiO<sub>2</sub> shell PCM microcapsules on the ageing period and temperature. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

faster hydrolysis of TEOS and the morphology of the product was weakly branched. On the contrary, faster condensation occurred in a basic catalytic system and the morphology of the product was of a particle type. Under acid conditions, the morphology of the precursor is similar to that of a “branched polymer” and can exist in an interpenetrating network (IPN) with PMMA network.<sup>15</sup> Therefore, with decreasing pH value, the dense structure of the shell was easily obtained so that the latent heat and PCM content of the microcapsules increased.

#### Effect of ageing time and temperature

The sol-gel process is a dynamic system that evolves with time, temperature, and storage conditions. There is evidence that ageing has a marked effect on the final properties of the microcapsules. Ageing likewise results in further chemical reactions, producing strengthening, stiffening, and shrinkage of the network. It could be controlled by using varied pH values, temperature, pressure, and time.

At room temperature, the SiO<sub>2</sub> content of the PCM microcapsules increased with increased ageing days (Table II). The maximum latent heat, 107.8 J/g, and PCM content, 44.2%, were obtained when ageing time was 4 days. After 4 days, the latent heat and PCM content of the microcapsules decreased because excessive shrinkage compressed PCM out of the microcapsules. Table III also shows that the SiO<sub>2</sub> content of the microcapsules increased with increasing ageing days at 60°C. The maximum latent heat and PCM content of the microcapsules were obtained with 6 days ageing time. The SiO<sub>2</sub> content of microcapsules was also increased with increasing ageing days at 80°C. However, the PCM content of the microcapsules was decreased with increasing ageing days at 80°C (Table IV). This finding was because microcapsules were shrunk violently and excessively when ageing at high temperature. On the other hand, the appearance and structures of the PCM microcapsules without ageing were similar to those with different ageing time and temperature, indicating that the PCM microcapsules were formed before the ageing process as shown in Figure 4. The effect of ageing temperature and period on the SiO<sub>2</sub> content of microcapsules is shown in Figure 5. When the microcapsules were aged for 1 day, the SiO<sub>2</sub> content of the microcapsules raised slowly at 60°C and 80°C, respectively. It was considered that further reaction of the unreacted monomer occurred in the solution. With the increase in both the organic and inorganic polymer of the microcapsules during the aging process, the SiO<sub>2</sub> content of the microcapsules increased gradually. After a 1 day ageing process, the SiO<sub>2</sub> content of the microcapsules began to grow significantly at 80°C due to the absence of unreacted organic monomer. However, the unreacted organic monomer still remained in the solution at 60°C. Therefore, the SiO<sub>2</sub> content of the microcapsules grew gradually when the ageing time was more than 1 day at 60°C. On the other hand, the unreacted organic monomer did not easily further react when was ageing at room temperature. Hence, there was only a limited increase in the SiO<sub>2</sub> content of the microcapsules. The most suitable preparation

**TABLE V**  
The Preparation Conditions of PMMA Network/SiO<sub>2</sub> Shell PCM Microcapsules with Varying Inorganic/Organic Ratios

Inorg./org. ratio (%)	PCM (g)	Prepolymer (g) <sup>a</sup>	EGDMA (g)	AIBN (g)	TEOS (g)	H <sub>2</sub> O (g)	PVA (g)
5	25	44.0	4.75	0.475	4.30	150	0.75
10	25	41.6	4.50	0.450	8.60		
15	25	39.3	4.25	0.425	12.9		
20	25	37.0	4.00	0.400	14.4		

<sup>a</sup> Prepolymer-Toluene : MMA : AIBN = 20 : 16 : 1, react at 60°C, 1 h. Ageing condition: 80°C, 4 days.

**TABLE VI**  
The Properties of PMMA Network/SiO<sub>2</sub> Shell PCM Microcapsules Prepared with Varying Inorganic/Organic Ratios

Inorganic/organic ratio (%)	$\Delta H$ (J/g)	PCM content (%) <sup>a</sup>	SiO <sub>2</sub> content (%)	SiO <sub>2</sub> content in shell (%) <sup>b</sup>
5	178.9	73.3	0.94	3.5
10	130.3	53.4	2.32	5.0
15	124.2	50.9	3.55	7.2
20	125.4	51.4	5.37	11.0

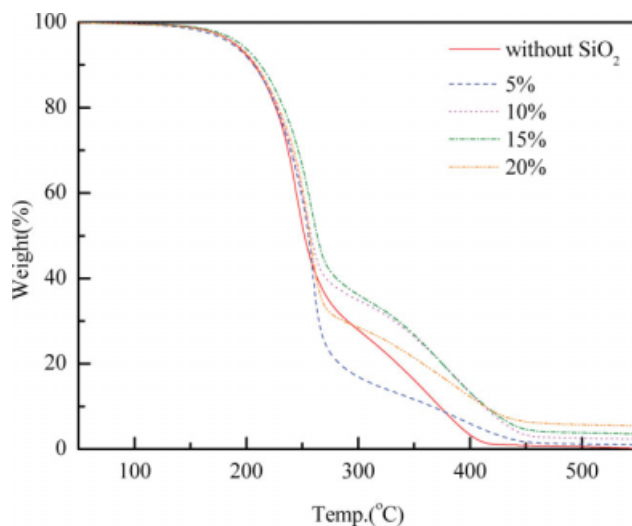
<sup>a</sup> PCM content = ( $\Delta H$  of sample)/( $\Delta H$  of *n*-octadecane).

<sup>b</sup> SiO<sub>2</sub> content in shell = (SiO<sub>2</sub> content)/[100-(PCM content)].

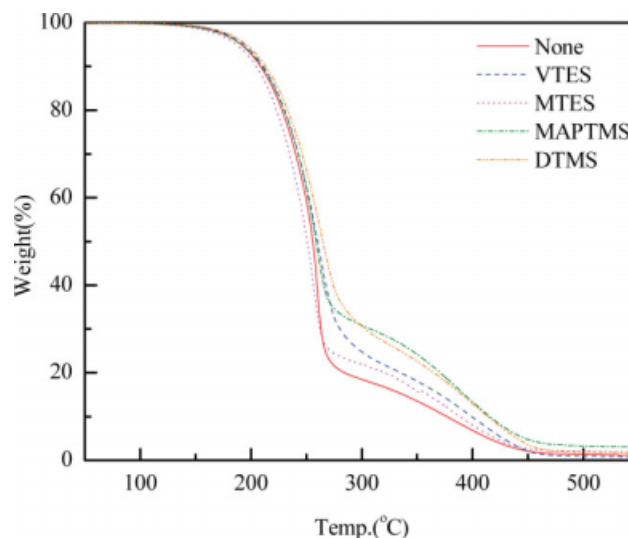
condition was ageing at 80°C for 4 days. Under this condition, the unreacted monomer completely reacted and the SiO<sub>2</sub> and PCM content of the microcapsules increased as much as possible.

#### Effect of SiO<sub>2</sub> content in PCM microcapsules

As mentioned earlier, PMMA network-silica shell PCM microcapsules were prepared by adding TEOS into the prepolymer solution, later reacting with water from the disperse phase via sol-gel process. The preparation conditions and results of the PMMA network/SiO<sub>2</sub> shell PCM microcapsules with different inorganic/organic ratios are shown in Tables V and VI, respectively. Conversion of the prepolymer was 30%. The latent heat and PCM content of microcapsules decreased with increasing inorganic/organic ratio. The maximum latent heat 178.9 J/g and PCM content 73.3% in the microcapsules were obtained when the inorganic/organic ratio was



**Figure 6** TGA diagrams of PMMA network/SiO<sub>2</sub> shell PCM microcapsules prepared with different inorganic/organic ratios. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]



**Figure 7** TGA diagrams of PMMA network/SiO<sub>2</sub> shell PCM microcapsules prepared with different coupling agents or not. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

5%. This indicates that a suitable amount of inorganic precursor could be easily interpenetrated with organic substrate to form hybrid and denser shells of the microcapsules. The TGA curves of the PCM microcapsules with different inorganic/organic ratios are shown in Figure 6. No matter what the inorganic/organic ratios were, there was no distinct improvement of the thermal stabilities of the microcapsules.

#### Effect of coupling agents

Four different functional groups of coupling agents were chosen for introduction into the PCM microcapsules. The inorganic/organic ratio was 5% and the ratio of the coupling agent/TEOS was 5 wt %. Conversion of the prepolymer was 30%. The TGA curves of the PCM microcapsules with different coupling agents are shown in Figure 7. The thermal stabilities of the microcapsules did not improve with

**TABLE VII**  
The Properties of PMMA Network/SiO<sub>2</sub> Shell PCM Microcapsules Prepared with different Coupling Agents or Not

Coupling agents	$\Delta H$ (J/g)	PCM content (%) <sup>a</sup>	SiO <sub>2</sub> content (%)	SiO <sub>2</sub> content in shell (%) <sup>b</sup>
None	180.5	74.0	1.08	4.2
VTES	162.4	66.6	0.72	2.2
MTES	165.7	67.9	1.76	5.5
MAPTMS	135.9	55.7	2.98	6.7
DTMS	147.9	60.6	1.71	4.3

<sup>a</sup> PCM content = ( $\Delta H$  of sample)/( $\Delta H$  of *n*-octadecane).

<sup>b</sup> SiO<sub>2</sub> content in shell = (SiO<sub>2</sub> content)/[100-(PCM content)].

the introduction of the coupling agents. The maximum latent heat 180 J/g and PCM content 74% of the microcapsules were obtained without introducing any coupling agents (Table VII). The methoxyl groups of the coupling agents, DTMS and MAPTMS, were more reactive than the ethoxyl group in TEOS. The methoxyl group hydrolyzed easily to become hydroxyl groups which diffused into the aqueous phase as soon as possible. Therefore, microcapsules prepared by the introduction of DTMS or MAPTMS had incomplete surface resulting in the lowest latent heat and PCM content. On the other hand, the latent heat and PCM content of microcapsules prepared with the introduction of the ethoxyl group of coupling agents, VTES and MTES, were higher than those with the introduction of the methoxyl group of coupling agents, MAPTMS and DTMS. Nonetheless, the latent heat and PCM content of the microcapsules prepared with introduction of any coupling agents were not higher than those without introduction of any coupling agents. It was found that the introduction of any coupling agent in the microcapsules did not improve the properties of microcapsules.

### CONCLUSIONS

Encapsulation of the phase change material, *n*-Octadecane, by using PMMA network-silica hybrid as the shell material has been developed. The most effective method to prepare microcapsules is through the use of prepolymer solutions. Ageing conditions and the pH value in aqueous phase affect the surface structure and the PCM content in microcapsules. The SiO<sub>2</sub> content in microcapsules increases with increasing ageing time. The most suitable condition for producing high

latent heat and PCM content in microcapsules is by the introduction of 5% SiO<sub>2</sub> in microcapsules. The highest latent heat (180 J/g) and PCM content (74%) of microcapsules may be obtained without adding any coupling agents.

The authors gratefully acknowledge the Precision Instrument Center of the College of Engineering, National Central University for offering them to do the research.

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