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INNOVATIVE BIFUNCTIONAL MICROCAPSULE FOR HEAT STORAGE AND ANTIBACTERIAL PROPERTIES

Amorn Chaiyasat[1], Siriluk Namwong[1], Boontida Uapipatanakul[1],
Warayuth Sajomsang[2], and *Preeyaporn Chaiyasat[1]

[1]Faculty of Science and Technology, Rajamangala University of Technology Thanyaburi, Thailand
[2]National Nanotechnology, Thailand Science Park, Thailand

*Corresponding Author, Received: 10 June 2017, Revised: 01 Aug. 2017, Accepted: 02 Sept. 2017

ABSTRACT: It is well known that the microcapsules encapsulating heat storage or phase change materials are coated onto the fabrics for the thermoregulating property. To increase their function on the textile application, in this work, bifunctional microcapsules having both thermal energy storage and antibacterial properties were fabricated. Chitosan was used as surfactant of poly(methyl methacrylate-divinyl benzene) microcapsule encapsulated octadecane (OD) prepared by microsuspension iodine transfer polymerization. The nonspherical with dent microcapsules were prepared in acidic condition without any free polymethyl methacrylate particles nucleated by emulsion polymerization. Due to amino groups in chitosan chains were protonated in which presented positive charge, the microcapsules stabilized with such molecules were then obtained without any coalescence. In addition, percent yield of microcapsules decreased with chitosan concentration in the aqueous medium. It may be due to the high viscosity of the aqueous medium where the monomer droplets are unable to maintain the shape in the early stage of the polymerization. For thermal properties, the latent heats of the encapsulated octadecane (234 J/g-OD) were closed to those (233 J/g-OD) of bulk octadecane. For antibacterial property, 2 millimetres over control sample of the resulting inhibition zones of both *S. aureus* and *E. coli* were observed. Based on bifunctional feature derived from both microcapsule core and shell for thermal energy storage and antibacterial properties, respectively, the developed microcapsules would express a great potential for textile application.

Keywords: Heat storage materials; Antibacterial property; Microcapsule; Microsuspension iodine transfer polymerization

1. INTRODUCTION

Because all seasons of Southeast Asia are hot and humid, thermoregulating fabrics or thermal adaptable cloths and antibacterial fabrics are then interested in the commercial production. For the former case, microcapsules containing phase change materials are often coated onto the fabrics[1, 2]. Because Paraffin waxes can melt and crystallize at a wide range of temperature to absorb and release energy, respectively, they then are preferably used as phase change material and encapsulated in the microcapsules. Encapsulation of paraffin waxes not only increases surface area for providing a larger heat transfer but also prevents phase change material reactivity to the outside environment and controls the volume change as phase change occurs [3]. One of the most famous techniques for phase change material microcapsule preparation is microsuspension conventional radical polymerization due to high encapsulation efficiency of phase change material [4-7]. Various kinds of polymer shell are used for the encapsulation based on microsuspension conventional radical polymerization such as polystyrene[8], poly(styrene-*co*-ethylmethacrylate) [9], poly-divinylbenzene [10, 11], poly(styrene-*co*-divinylbenzene) [12], poly(methyl methacrylate) [6] and poly(methyl methacrylate-*co*-methyl acrylate-*co*-methacrylic acid) [13]. To the best of our knowledge, among these polymer shells, polymethyl methacrylate is the best one to provide high latent heats of phase change material due to completion of phase separation between polymethyl methacrylate shell and wax core [7]. However, using micro suspension conventional radical polymerization, approximately 35% of free polymethyl methacrylate particles respects to total monomer used was formed competition with microcapsules. In previous work, such phenomenon can be overcome with micro suspension iodine transfer polymerization which reduced radical exit from the monomer droplets to an aqueous medium [6, 14]. Chitosan is one of the most famous biopolymers because its beneficial properties include antifungal and antibacterial [15, 16], neuroprotective [17], anti-inflammatory [18] and so on. Since chitosan contains amino group in its chain which was protonated to give positive charges in acidic condition, it may able to be used as the surfactant of micro suspension polymerization in place of the conventional one as

polyvinyl alcohol. To the best of our knowledge, there is a lack of research for phase change material microcapsule preparation using chitosan as a surfactant in microsuspension iodine transfer polymerization.

Therefore, in this research, the preparation of bifunctional microcapsule for both heat storage and antibacterial properties by microsuspension iodine transfer polymerization using octadecane, polymethyl methacrylate and chitosan for phase change material core, polymer shell, and surfactant, respectively, was studied in the first time. The influence of chitosan concentration on the encapsulation efficiency was investigated.

2. EXPERIMENTAL INVESTIGATION

2.1 Materials

Methyl methacrylate (Aldrich, Wisconsin, USA; purity, 99%) was purified by passing through the column packed with basic aluminum oxide to remove the polymerization inhibitors before use. Octadecane (Merck, Munich, Germany; 99.5%) was used as received. Reagent-grade benzoyl peroxide (Merck, Munich, Germany) was purified by recrystallization. Iodoform (CHI_3; Aldrich, Wisconsin, USA; purify, 99%), poly(vinyl alcohol) (Aldrich, Wisconsin, USA; the degree of saponification 87-90%, molecular weight 3-7x10^4 g/mol) and Chitosan were used as received.

2.2 Microcapsule Preparation

The preparation of polymethyl methacrylate/ octadecane microcapsules by microsuspension iodine transfer polymerization using chitosan as the stabilizer was carried out as follows. Firstly, 2.50 g of methyl methacrylate (or methyl methacrylate/divinyl benzene) was homogeneously dissolved in 2.50 g of octadecane, 0.02 g of CHI_3 and 0.20 g of benzoyl peroxide as the oil phase. It was then added to 45 g aqueous solution containing either 1%wt of poly(vinyl alcohol) or chitosan before homogenized at 5,000 rpm for 5 minutes to prepare monomer droplets of oil in water emulsion. Thereafter, the obtained polymer suspension was subsequently transferred to a round bottom flask, sealed with a silicone rubber septum and purged with a vacuum/N_2 cycle for five times (finally in N_2). It was finally polymerized at 80 °C for 3 hours and following at 90°C for 5 hours at a stirring rate of 500 rpm. The concentration of chitosan was varied as shown in Table 1.

Table 1 Recipes for the preparation of polymethyl methacrylate/octadecane microcapsules

Phase	Ingredients		
Oil	MMA (g)	2.50	2.25
	DVB (g)	0.00	0.25
	OD (g)	2.50	2.50
	BPO (g)	0.20	0.20
	CHI_3 (g)	0.02	0.02
Aqueous	Surfactant solution[a] (g)	45.00[a]	45.00[b]

Abbreviations: MMA, methyl methacrylate; OD, octadecane; BPO, benzoyl peroxide; CHI_3, iodoform; CS, chitosan; PVA, polyvinyl alcohol
[a] PVA solution 1 wt% or CS solution at 0.25, 0.50 and 1.00 wt%
[b] 1 wt% of CS

2.3 Characterization of Microcapsules

The microcapsules encapsulated octadecane was observed with an optical microscope (SK-100EB&SK-100ET, Seek, Seek Inter Co. Ltd., Thailand) and scanning electron microscope (JSM-6510, JEOL, JEOL Ltd., Japan) to study the morphology of the particle surface and shape, respectively. For scanning electron microscope observation, a few of dried microcapsules were placed on a nickel stub and dried before Au-coated. For the measurement of thermal properties, the microcapsules were washed with 2-propanol before dried in vacuum oven. The octadecane content of the dried washed microcapsules was determined by a thermogravimetric analyzer (TGA 4000, Perkin-Elmer, USA) at a heating rate of 5 °C/min. The latent heats (ΔH_m and ΔH_c) (J/g-capsule) and the melting (T_m) and crystallization (T_c) temperatures of the encapsulated octadecane were measured with a differential scanning calorimeter (DSC 4000, Perkin-Elmer, USA) under a N_2 flow in a scanning temperature range of -20-40 °C and at a heating/cooling rate of 5 °C/min and shown as average values of three measurements. The ΔH_m* and ΔH_c* (J/g-OD) were, respectively, obtained using the following Eq, (1) from the ΔH_m and ΔH_c and the octadecane content in each washed microcapsule (% loading) obtained from the thermogravimetric analyzer, which did not contain unencapsulated octadecane.

$$A = \left[\frac{B}{C}\right] \times 100 \quad (1)$$

Where A = ΔH_m* and ΔH_c* of the encapsulated octadecane in unit of joules per 1 g of encapsulated octadecane (J/g-OD)

B = ΔH_m and ΔH_c of the encapsulated octadecane in a unit of joules per 1 g of microcapsule (J/g-capsule) measured with differential scanning calorimeter

C = % loading (experiment) of octadecane in the washed microcapsules measured with thermogravimetric analyzer

The theoretical % loading of OD in the washed microcapsules was calculated by Eq. (2)

$$\%\text{Loading}_{(\text{theory})} = \left[\frac{W_{wax}}{(W_{wax}+W_P)}\right] \times 100 \quad (2)$$

Where W_{wax} and W_P are weights of octadecane and polymethyl methacrylate, respectively, in the polymerization recipes shown in Table 1.

Encapsulation efficiency (%) of octadecane was calculated using the Eq. (3).

$$\text{Encapsulation efficiency (\%)} = \left[\frac{\%\text{loading}_{(\text{experiment})}}{\%\text{loading}_{(\text{theory})}}\right] \times 100 \quad (3)$$

2.4 Antibacterial property

The antibacterial property of the prepared microcapsules was tested for *Staphylococcus aureus* (*S. aureus*) and *Escherichia coli* (*E.coli*) using disk-diffusion method at 35°C for 24 h.

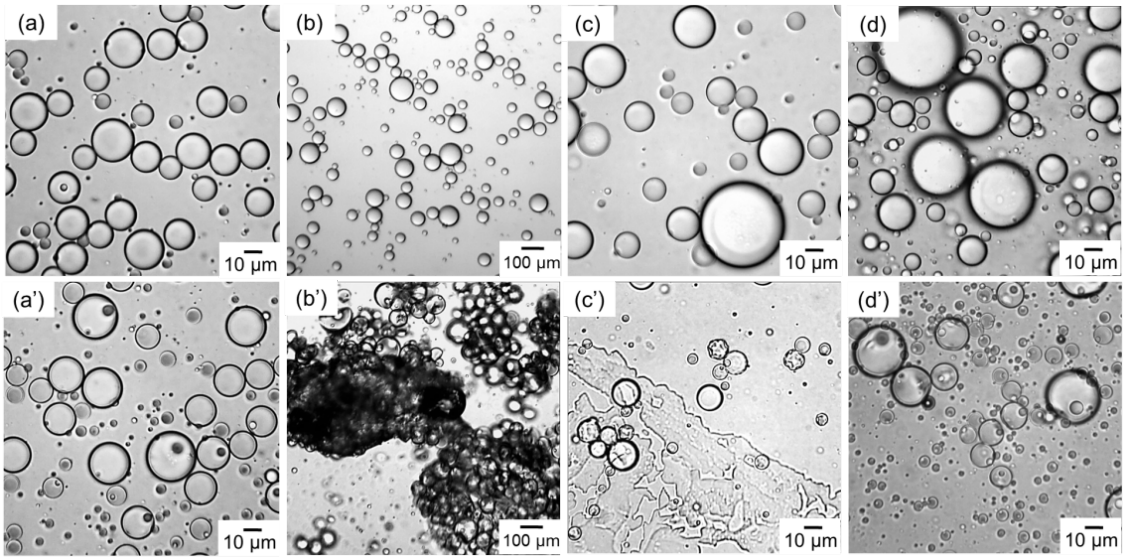

Fig. 1 Optical micrographs before (a-d) and after (a'-d') polymerization of polymethyl methacrylate/octadecane (50:50 w/w%) microcapsules prepared by microsuspension iodine transfer polymerization with 1 wt% of poly(vinyl alcohol) aqueous solution (a, a') and various concentrations of chitosan in aqueous solution (wt%): 0.25 (b, b'); 0.50 (c, c') and 1.0 (d, d')

3. RESULTS AND DISCUSSION

In general, poly(vinyl alcohol) is used as a surfactant in oil in water emulsion system [19-22] especially in microsuspension polymerization [4, 6, 14, 23]. The obtained microparticle/capsules represented high colloidal stability without coalescence. In this work, chitosan was used in place of poly(vinyl alcohol) to increase antibacterial property where the various concentrations of chitosan were studied compared with poly(vinyl alcohol) as shown in Fig. 1. It was found that at all chitosan concentrations (Fig. 1 b-c), monomer droplets were spherical with micrometer-sized. After polymerization, the poly- methyl methacrylate/octadecane microcapsules were unstable and broken in both cases of 0.25 (Fig. 1b') and 0.50 (c') wt% of chitosan. In addition, most of the obtained microcapsules (~75% based on monomer/octadecane in the recipe) were coagulated as shown in Fig. 2a and b. After left overnight, a lot of unencapsulated octadecane dispersed in the aqueous medium (Fig. 2c) whereas octadecane was not detected by a thermogravimetric analyzer (Fig. 2d) in the remained polymer particle precipitated in the bottom. It may be due to insufficient chitosan amount to maintain the microcapsule stability. However, such phenomenon was improved by the increase of chitosan to 1 wt% (Fig. 1d') in which the obtained polymethyl methacrylate/octadecane microcapsules shape was similar to those of using poly(vinyl alcohol) 1 wt% (Fig. 1a'). In addition, microsuspension iodine transfer polymerization represented high performance to reduce free polymethyl methacrylate particles nucleated in an aqueous medium. The aqueous medium of the

obtained polymethyl methacrylate/octadecane microcapsule using 1 wt% of chitosan was changed from milky (Fig. 3a) to transparent (Fig. 3b) after centrifugation at about 3,000 rpm. It accorded with the previous articles [6, 14] where most of the obtained polymethyl methacrylate/ octadecane microcapsule having a total density lower than that of the water floated on the top of the suspension. However, after washing poly-methyl methacrylate/octadecane microcapsules with 2-propanol (Fig. 3c), some microcapsules were broken and having a hole on their surface in which reduced the encapsulation efficiency (~80%) of octadecane. This finding may due to some of the chitosan having high molecular weight absorbed on some microcapsule surfaces as chitosan-riched surface obstructed the polymethyl methacrylate chains to adsorb at the interface. The shell strength then decreased giving a hole after washing. Therefore, to improve the polymer shell strength, polymethyl methacrylate was then copolymerized with 10 wt% related to the monomer of crosslinking monomer as divinyl benzene.

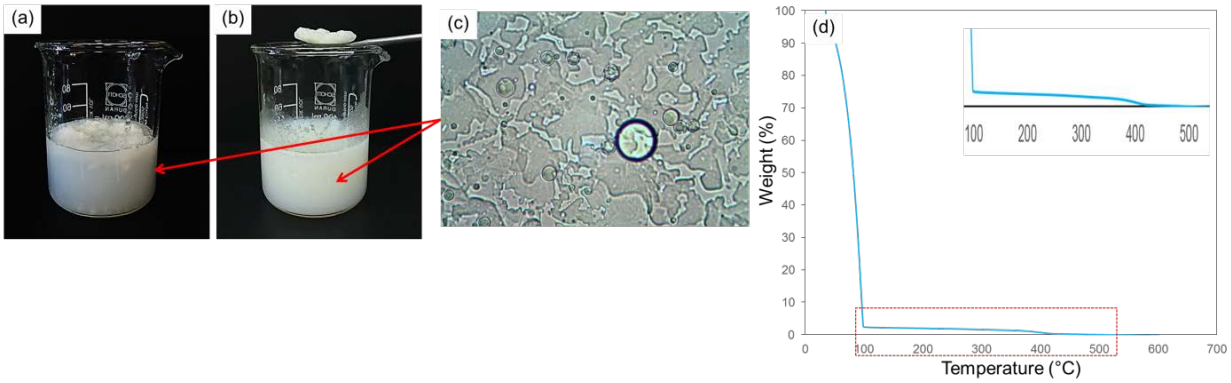

Fig. 2 Suspension photos (a and b), optical micrograph (c) and thermogravimetric thermogram (d) of polymethyl methacrylate/octadecane microcapsules prepared by microsuspension iodine transfer polymerization using various concentrations of chitosan in aqueous medium (wt%): 0.25 (a) and 0.50 (b)

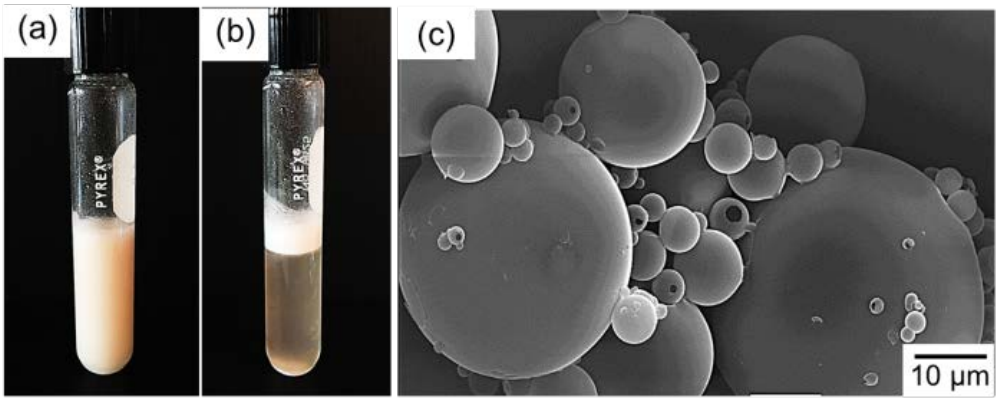

Fig. 3 Suspension photos before (a) and after (b) centrifugation at 3,000 rpm for 15 minutes of polymethyl methacrylate/octadecane microcapsules and scanning electron micrograph (c) polymethyl methacrylate/octadecane microcapsules washed with 2-propanol

The monomer droplets of methyl methacrylate/divinyl benzene/octadecane were spherical (Fig. 4a) similar to methacrylate/ octadecane monomer droplets. After polymerization, the obtained poly(methyl methacrylate-divinyl benzene)/octadecane microcapsules represented nonspherical shape with a big dent (Fig. 4b and c). The dent formation is due to the encapsulated octadecane volume shrinkage based on the decreasing of temperature from 90 °C (polymerization temperature) to room temperature.

The polymer shell with complete enveloping octadecane core would not withstand the external pressure leading to the formation of a dent on their surfaces. In contrast, polymethyl methacrylate/ octadecane microcapsules can reduce the external pressure via a hole where they can maintain their shapes without any dents. Therefore, the encapsulation efficiency (~100%) was improved by copolymer shell where the exited octadecane floated on the top of the suspension after polymerization was not observed.

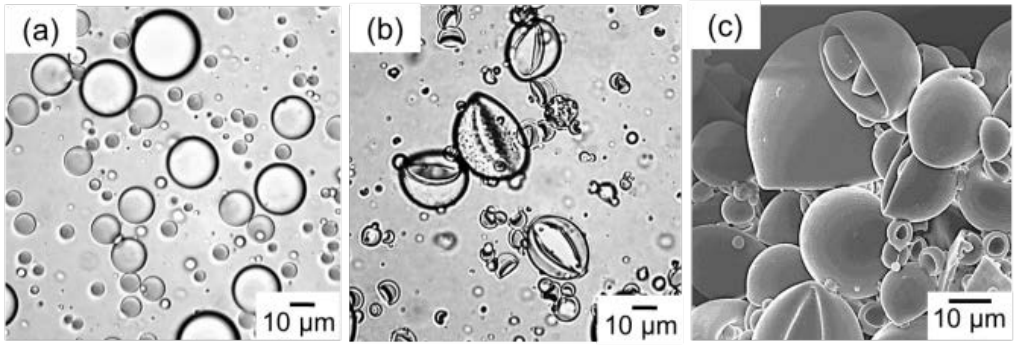

Fig. 4 Optical micrographs of (a) monomer droplets and (b) poly(methyl methacrylate-divinyl benzene)/octadecane microcapsules and scanning electron micrograph of poly(methyl methacrylate-divinyl benzene)/octadecane after washing with 2-propanol

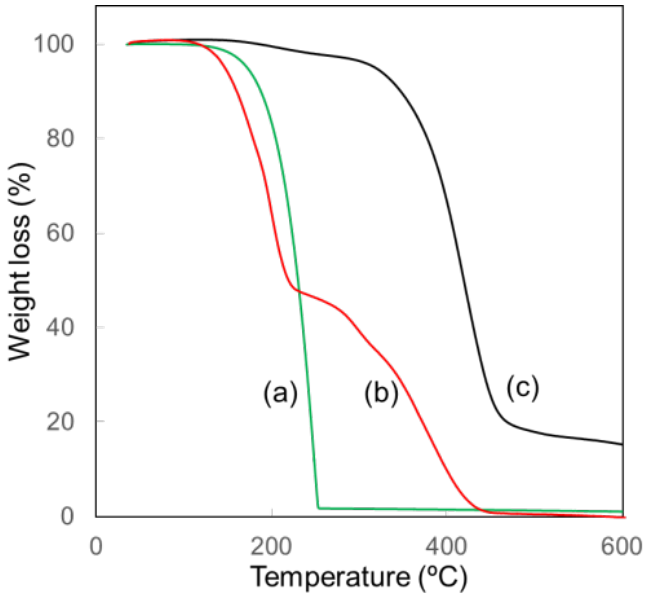

Fig. 5 Thermogravimetric thermograms of bulk octadecane (a), poly(methyl methacrylate-divinyl benzene)/octadecane microcapsules using methyl methacrylate: divinyl benzene ratio of 90:10 (w/w%) (b) and poly(methyl methacrylate-divinyl benzene) particles (c) prepared by microsuspension iodine transfer polymerization

The degradation temperatures of bulk octadecane, poly(methyl methacrylate-divinyl benzene) and the encapsulated octadecane in the poly(methyl methacrylate-divinyl benzene) microcapsules were observed by the thermo-gravimetric analyzer as shown in Fig. 5. The decomposition temperatures of poly(methyl methacrylate-divinyl benzene)/octadecane microcapsule (Fig. 5b) were shown in two respective steps which were octadecane core (130-260°C) and poly(methyl methacrylate-divinyl benzene) shell (220-430°C), respectively. The degradation of bulk octadecane (Fig. 5a) at 140-250°C was closed to that of the encapsulated octadecane. The thermogravimetric thermograms confirmed that octadecane existed in the prepared microcapsules in which about 50% loading of octadecane in poly(methyl methacrylate-divinyl benzene)/octadecane was obtained. In addition, it was close to the theoretical value (50% for 50:50 of monomer: octadecane ratio). This indicates that high encapsulation efficiency (ca. 100%) was obtained.

The latent heats of bulk octadecane (J/g) (Fig. 6a) and the encapsulated octadecane (J/g-sample) (Fig. 6b) were obtained from the heating/cooling peak areas of differential scanning calorimeter thermograms at T_m and T_c, respectively. The latent heats (ΔH_m* = 234 J/g-OD and ΔH_c* = 236 J/g-OD) of the encapsulated octadecane calculated from equation 1 were quite closed to those of bulk octadecane (ΔH_m* = 233 J/g-OD and ΔH_c* = 234

J/g-OD). Because main polymer shell is a hydrophilic polymer as polymethyl methacrylate containing only 10 wt% of the hydrophobic polymer as divinyl benzene, it would increase the phase separation between polymer shell and octadecane core where the encapsulated octadecane behaved similarly to bulk octadecane.

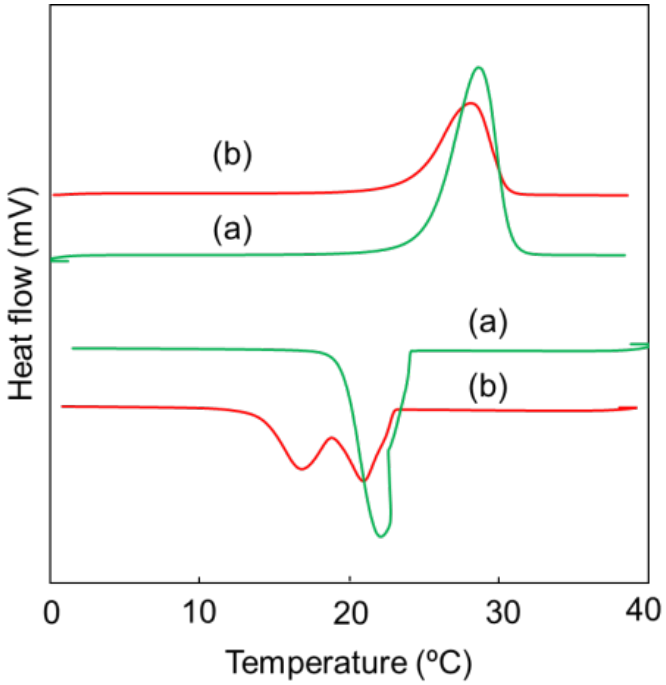

Fig. 6 Differential scanning calorimeter thermograms of bulk octadecane (a) and poly(methyl methacrylate-divinyl benzene)/octadecane microcapsules using methyl methacrylate: divinyl benzene ratio of 90:10 (w/w%) prepared by microsuspension iodine transfer polymerization (b)

The antibacterial property of the obtained poly(methyl methacrylate-divinyl benzene)/ octadecane microcapsules using chitosan as the stabilizer was performed on a bacterial culture of *S. aureus* and *E. coli* by measuring the size of the inhibition zones. It was found that a few antimicrobial activity of poly(methyl methacrylate-divinyl benzene)/octadecane microcapsule was observed based on the small size (2 millimeters) of the resulting inhibition zones of both bacteria. This indicates that amino groups in chitosan chains represented the low performance of the antibacterial property. Therefore, in the future work, amino groups in chitosan chains will be functionalized to be ammonium salt having high performance antibacterial activity and used as the surfactant in microsuspension iodine transfer polymerization.

4. CONCLUSION

The nonspherical shape with a dent poly(methyl methacrylate-divinyl benzene) microcapsules containing octadecane was prepared by microsuspension iodine transfer polymerization using chitosan as a stabilizer without any coagulation. Using polymethyl methacrylate as a polymer shell, microcapsules with a hole were obtained resulting in about 80% encapsulation efficiency. The encapsulation efficiency (100%) was improved by poly(methyl methacrylate-divinyl benzene) copolymer shell. The latent heats of the encapsulated OD were close to those of bulk octadecane. The antibacterial activity of chitosan absorbed on the microcapsule surface was still low (2 millimeters inhibition zone) which needed to be improved in the future work.

5. ACKNOWLEDGEMENTS

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