

# Preparation of Thermosensitive Microcapsules Containing Water Soluble Powder by Melting Dispersion Cooling Method

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## Abstract

It was tried to prepare the thermosensitive microcapsules containing the water soluble solid powder by the melting dispersion cooling method and to establish the optimum preparation conditions. As a model water soluble solid powder, sodium hydrogen carbonate was adopted in order to generate carbon dioxide gas and as a thermosensitive shell material, olefin resin with the melting point of ca. 40°C was used. In the experiment, the concentration of olefin resin in the shell material solution was mainly changed together with the concentrations of the oil soluble surfactant species and the  $\alpha$ -tocopherol as a modifier of shell. Addition of  $\alpha$ -tocopherol into the shell material solution could prevent the core from breaking away during the microencapsulation process and result in the higher microencapsulation efficiency, because the dispersion stability of solid powder in the shell material solution could be increased due to the increase in affinity between the shell material solution and solid powder. Also, the microencapsulation efficiency increased with the concentration of olefin resin, became maximum at 50 wt% and then, decreased. The microcapsules were found to begin melting at 36°C and to generate carbon dioxide gas.

## Keywords

Thermosensitive Microcapsules, Microencapsulation, Sodium Hydrogen Carbonate, Melting Dispersion Cooling Method, Olefin Resin

## 1. Introduction

The microcapsule is the generic name of the vessels with the size of  $\mu\text{m}$  order and can contain the core materials such as solid, liquid and gas and handle as the fine solid particles.

For this, numerous kinds of microcapsules have been prepared and applied in the various fields such as agriculture, latent heat storage, cosmetics, food, drugs, paintings, adhesives, textile and electric materials [1]-[4]. The main functions of microcapsules are to protect the core materials from environment for a long time, to release optionally the core materials according to stimuli, to modify the surface of core material and to change the core materials such as liquid and gas to the particulate materials [5]-[7]. The microcapsules with these functions can be prepared by selecting the core and the shell materials with the desired chemical and physicochemical properties and by developing the microencapsulation procedure. Also, the diameter and inner structure of microcapsules can be controlled according to the shape and size of core and the preparation conditions.

It is well known that it is very hard to microencapsulate the gaseous materials and to perfectly prevent them from leaking for a long time. In this case, it may be effective to microencapsulate solid powders generating the desired gas instead of microencapsulating gas directly. Namely, if solid powder could be microencapsulated with some stimuli-responsive shell materials, gas could be generated by breaking the shell due to suitable stimuli and by dissolving solid powder in the solvent. Thus, as solid powder can be relatively easily microencapsulated compared to gas and protected stably for a long time, the desired gas may be generated as occasion demands. However, it is necessary to develop newly the microencapsulation procedure in order to stably microencapsulate solid powder, to prepare the microcapsules with the higher content and to protect solid powder from environment according to the physicochemical properties of solid powder species.

In general, in order to microencapsulate the water soluble core materials, the hydrophobic shell materials are used to increase the content and to protect the core from water or moisture by the help of oil soluble surfactant [8]-[11]. Also, the non aqueous solvents as the continuous phase have been sometimes used to prevent the core from leaking way during the microencapsulating process [12] [13]. Meanwhile, on applying the microcapsules to food, cosmetics, drugs and textile, the nontoxic materials have to be used as the shell material. For example, the chemical preparation methods using polymerizable monomers such as isocyanate, styrene and acryl monomer cannot be used. In this case, the physicochemical preparation methods and the chemical methods using the nontoxic materials may be recommended [14] [15]. Taking these things described above into consideration, it was tried to microencapsulate sodium hydrogen carbonate as a water soluble solid powder instead of microencapsulating carbon dioxide gas. Sodium hydrogen carbonate used generally as a food additive is dissolved well in the acid solution, generate carbon dioxide and change from the acid solution to the alkaline solution. If sodium hydrogen carbonate powder could be stably microencapsulated with the thermosensitive shell material, carbon dioxide gas with a few physiological effects [16] [17] could be generated by breaking down the microcapsules due to heating and by contacting solid powder with the optimum aqueous solution. When the microcapsules thus prepared were used in the cosmetic materials such as the face cream and the basic skin care powder, carbon dioxide gas could be supplied to the face. On preparing the

microcapsules containing the water soluble solid powders as sodium hydrogen carbonate, it may be desired that the hydrophobic shell materials should be used and yet the microcapsules could be prepared by the procedure with the shorter processing time. In this study, it was tried to microencapsulate sodium hydrogen carbonate with the olefin resin as the hydrophobic thermosensitive shell material together with  $\alpha$ -tocopherol as a modifier by using the melting dispersion cooling method in the oil-in-water emulsion. Here, the olefin resin without halogen is completely nontoxic and  $\alpha$ -tocopherol has several physiological effects [18]-[20]. The purposes of this study are to microencapsulate sodium hydrogen carbonate with the melting dispersion cooling method, to characterize thermosensitivity of the microcapsules, to investigate whether carbon dioxide gas could be generated by breaking the microcapsules due to heating or not and to establish the optimum preparation conditions.

## 2. Experimental

### 2.1. Materials

Materials used to prepare the thermosensitive microcapsules were as follows. The thermosensitive shell material was olefin resin (CPAO: Idemitsu Kosan, Co., Ltd., Japan) with the melting point of ca. 40°C.

The additives added to modify the shell material were  $\alpha$ -tocopherol (VE: Pure chemical Co. Ltd., Japan) and the oil soluble surfactants which were polyglycerine polyricinolate (PR-300: Riken Vitamin Co. Ltd., Japan) and Soybean Lecithin (SBL: Pure Chemical Co. Ltd., Japan).

The water soluble stabilizer was methyl cellulose (SM-15) (MC: Shinetsu Kagaku Ind. Co., Ltd., Japan).

Sodium hydrogen carbonate (SHC: mean diameter = 16.8  $\mu\text{m}$ , density = 2.2  $\text{g}/\text{cm}^3$ ; Pure Chemical Co. Ltd., Japan) was adopted as the water soluble solid powder. The continuous phase was distilled water.

### 2.2. Preparation of Microcapsules

**Figure 1** shows the flow sheet for preparing the thermosensitive microcapsules containing SHC.

First, SHC was added into melted CPAO together with VE and the oil soluble surfactants and stirred with magnetic stirrer to form the (S/O) dispersion in the beaker with the volume of 300  $\text{cm}^3$ .

Then, the (S/O) dispersion was added into the continuous water phase dissolving MC and stirred with the six bladed disc turbine impeller to form the (S/O)/W dispersion in the beaker with the volume of 500  $\text{cm}^3$ .

The operation described above was performed at 40°C to dissolve CPAO.

After stirring for a given time, the (S/O)/W dispersion was cooled down to the room temperature in order to solidify CPAO and to form the microcapsules.

The microcapsules were separated with the filtration paper and characterized.

In this fundamental experiment, the concentrations of CPAO and oil soluble surfac-

tants were mainly changed. Also, the effect of addition of VE on the microencapsulation efficiency was investigated. The experimental conditions are shown in **Table 1**.

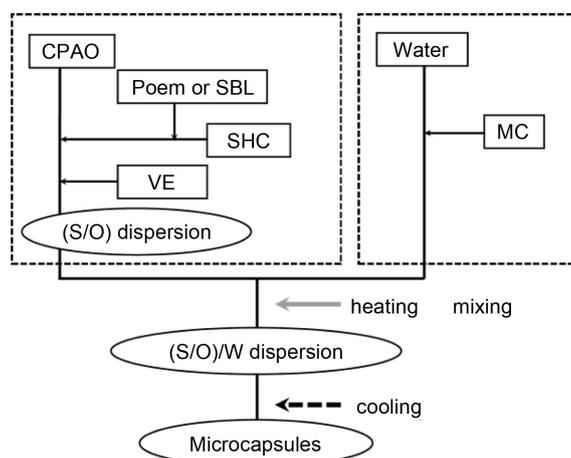
## 2.3. Characterization

### 2.3.1. Thermosensitivity

Thermosensitivity of microcapsules was measured by differential scanning calorimetry (DSC; Shimazu Co., Ltd., DSC-50, Kyoto, Japan) and by observing the change in the color of dye aqueous solution due to carbon dioxide generating on the basis of dissolution of SHC in the aqueous solution.

### 2.3.2. Mean Diameter of Microcapsules

The mean diameters of microcapsules were measured by particle size analyzer (SALD-3000; Shimazu Co., Ltd.). Here, the mean diameters are the Sauter mean diameters.



**Figure 1.** Flow sheet for preparing microcapsules.

**Table 1.** Experimental conditions.

<Dispersed phase>		
<b>Shell material solution</b>		10g
	Olefin resin (CPAO)	$C_{ow} = 15 - 100$ wt%-shell material
	$\alpha$ -tocopherol (VE)	0 ~ 85 wt%-Shell material
	Oil soluble surfactant	PR-300, SBL
		$C_o = 0 - 10$ wt%-Shell material
<b>Core</b>	Sodium Hydrogen Carbonate (SHC)	2 g
<Continuous phase>		
	Distilled water	100 g
	Methyl cellulose (MC)	1 g
<Preparation of (S/O)/W dispersion>		
	Temperature	$T = 40^\circ\text{C}$
	Impeller speed	$Nr = 600$ rpm
	Mixing time	$t = 5$ min

### 2.3.3. Observation of Microcapsules

The microcapsules were observed by scanning electron microscope (SEM) (VE-9800, Keyence Corp. Osaka, Japan) and optical microscope (BH-2; OLYMPOUS Co., Ltd., Tokyo, Japan).

### 2.3.4. Microencapsulation Efficiency

The microencapsulation efficiency ( $\lambda$ ) was estimated by Equation (1).

$$\lambda(\%) = \frac{\text{weight of microencapsulated solid powder}}{\text{weight of solid powder in feed}} \times 100 \quad (1)$$

Here, the weight of microencapsulated solid powder was measured from the change in the weight of microcapsules obtained by TGA (DTG-50/50H; Shimazu Co., Ltd.).

### 2.3.5. Measurement of Viscosity of Shell Material Solution

As the viscosity of shell material solution affects the dispersing stability of SHC, the viscosity of shell material solution was measured by the plate vibrating viscometer (VM-IAL; Yamaichi Denki Ltd., Tokyo, Japan).

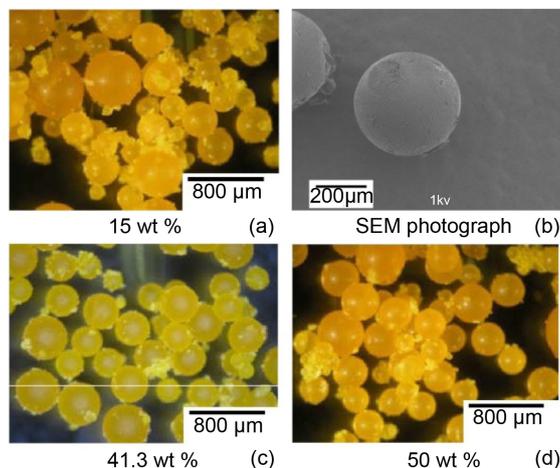
## 3. Results and Discussions

### 3.1. Observation of Microcapsules

**Figure 2** shows the stereo microscopic photographs ((a), (c), (d)) and SEM photograph ((b),  $C_{ow} = 15$  wt%) of microcapsules prepared by changing the CPAO concentration, where PR-300 was used at  $C_o = 5$  wt%.

Also, **Figure 3** shows the stereo microscopic photographs of microcapsules prepared by changing the concentrations of PR-300 (**Figure 3(a)**) and SBL (**Figure 3(b)**) at  $C_{ow} = 50$  wt% together with the SEM photographs ( $C_{ow} = 50$  wt%,  $C_o = 10$  wt%).

From these photographs, it was found that the sound spherical microcapsules could be prepared with the melting dispersion cooling method even by adding the two kinds of oil soluble surfactants and by changing the concentrations of CPAO and oil soluble surfactants.



**Figure 2.** Observation of microcapsules (effect of CPAO concentration).

Moreover, it was observed from the SEM photographs of **Figure 2** and **Figure 3** that the microcapsules had the smooth surface.

### 3.2. Effect of CPAO Concentration on Thermosensitivity and Microencapsulation Efficiency

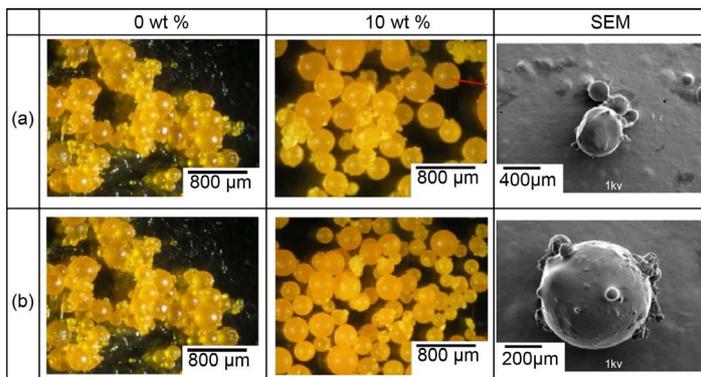
As the shell material is composed of CPAO, the oil solblesurfantant and VE, thermosensitivity of microcapsule must be different depending on the concentrations of them.

So, as it may be important to investigate how the melting behavior of shellmaterial depends on these concentrations, first, the melting temperature of the shell material was measured by changing the concentration of CPAO.

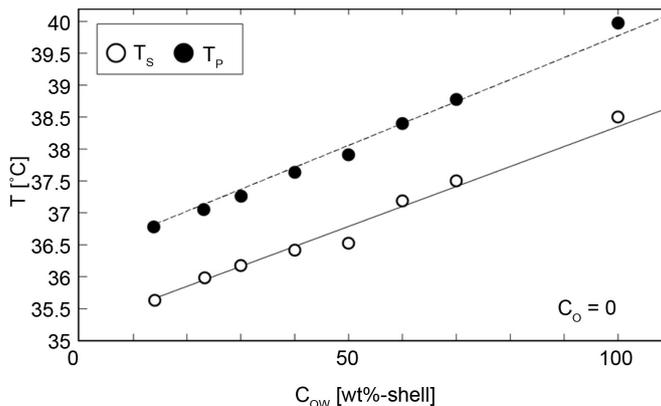
In this investigation, the lowest CPAO concentration required to solidify and to prepare the microcapsules was 15 wt%.

**Figure 4** shows the effect of the CPAO concentration on the melt starting temperature ( $T_s$ ) at which the shell material begins to melt and the peak melting temperature ( $T_p$ ) of shell material at which the melting temperature becomes maximum.

The melt starting temperature ( $T_s$ ) is raising from 35.2°C at  $C_{ow} = 15$  wt% to 38.5°C at  $C_{ow} = 100$  wt% with the CPAO concentration. Also, the peak melting temperature is raising from 36.4°C at  $C_{ow} = 15$  wt% to 40°C at  $C_{ow} = 100$  wt% with the CPAO concentration, too.



**Figure 3.** Observation of microcapsules (effect of oil soluble surfactant species).



**Figure 4.** Effect of the CPAO concentration on melting temperature of shell material.

From these results, it was found that the temperature for breaking the microcapsules, namely for releasing SHC, could be changed optionally from 35.2°C to 38.5°C by changing the CPAO concentration in the shell material solution composed of CPAO and VE.

While, the viscosity of shell material solution at the microencapsulation process may affect the dispersing stability of solid powder in this solution, as a result, the microencapsulation efficiency, because solid powder is prevented from moving in the viscous solution and breaking away from the solution droplet [21].

So, the dependence of the dispersing stability of SHC on the viscosity of shell material solution is estimated as follows.

Figure 5 shows the dependencies of the viscosity of shell material solution ( $\mu_d$ ) and the sedimentation velocity ( $V_s$ ) of a particle of SHC on the CPAO concentration, where the sedimentation velocity is estimated as the movement length of a particle of SHC per min in the shell material solution.

Here, the sedimentation velocity ( $V_s$ ) of a particle of SHC in the shell material solution was calculated by the following Stokes equation.

$$V_s = D_p (\rho_p - \rho_c) g / 18\mu_d \tag{2}$$

where  $D_p$ ,  $\rho_p$ ,  $\rho_c$  are the diameter of solid particle, the density of solid particle, the density of shell material solution, respectively.

The viscosity of shell material solution changed from 0.6 Pa·s at  $C_{ow} = 15$  wt% to 0.8 Pa·s at  $C_{ow} = 100$  wt% with the CPAO concentration.

On the other hand, the sedimentation velocity ( $V_s$ ) of solid powder decreased from  $7.5 \times 10^{-2}$  mm/min (1.25  $\mu$ m/s) at  $C_{ow} = 15$  wt% to  $4.8 \times 10^{-2}$  mm/min (0.8  $\mu$ m/s) at  $C_{ow} = 100$  wt% with the CPAO concentration.

From these results, it may be anticipated that the microencapsulation efficiency could be increased by increasing the viscosity of the shell material solution, because a particle of SHC could be stabilized due to the higher viscosity in the shell material solution droplets.

Figure 6 shows the dependencies of the mean diameters ( $d_p$ ) of microcapsules and the microencapsulation efficiency ( $\lambda$ ) on the CPAO concentration [22].

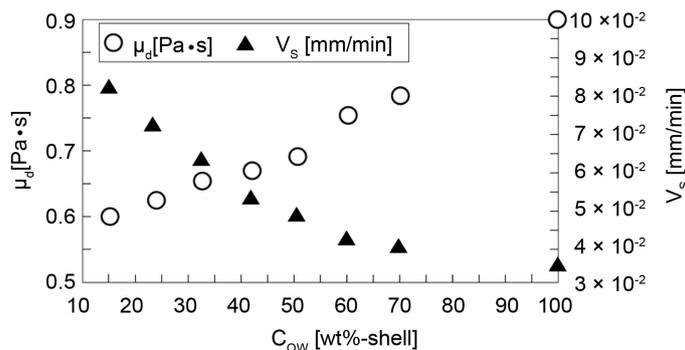
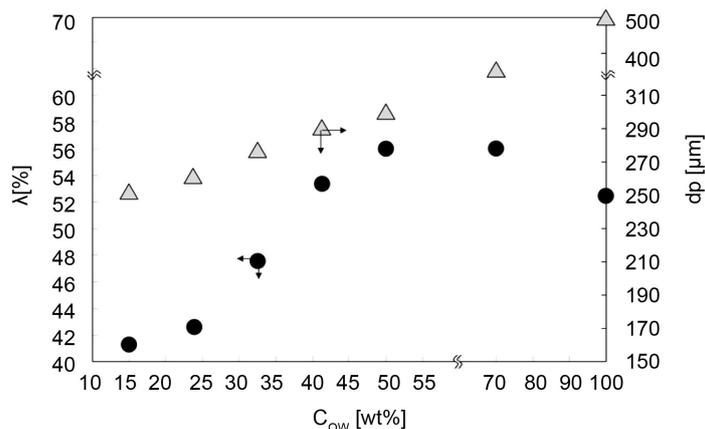


Figure 5. Dependencies of viscosity of shell material solution and sedimentation velocity on CPAO concentration.



**Figure 6.** Dependencies of microencapsulation efficiency and mean diameter on CPAO concentration.

The mean diameter increased from 250  $\mu\text{m}$  at  $C_{ow} = 15$  wt% to 480  $\mu\text{m}$  at  $C_{ow} = 100$  wt%. The increase in the mean diameter is due to the fact that the viscous force against the destruction force acted on a droplet increased with the CPAO concentration [23].

Meanwhile, the microencapsulation efficiency increased from 41% at  $C_{ow} = 15$  wt% to 56% at  $C_{ow} = 50$  wt% and then, decreased to 52% at  $C_{ow} = 100$  wt%.

Here, the dependence of microencapsulation efficiency on the viscosity of the shell material solution may be discussed by use of the mean diameter of a droplet or a microcapsule and the sedimentation velocity as follows.

Namely, if the value of  $d_p/V_s$  is larger than one, a solid particle could have a higher possibility for dispersing stably in a droplet. Contrary, if the value of  $d_p/V_s$  is smaller than one, a solid particle could leave away easily from a droplet. For example, as the values of  $d_p/V_s$  are 200 at  $C_{ow} = 15$  wt% and 375 at  $C_{ow} = 50$  wt%, respectively, the higher the CPAO concentration, the larger the microencapsulation efficiency becomes.

However, the decrease in the microencapsulation efficiency at the CPAO concentration larger than 50 wt% may be due to the decrease in affinity between SHC and the shell material solution owing to the decrease in the VE concentration. As a result, SHC was not able to disperse stably in the shell material solution.

### 3.3. Effect of Oil Soluble Surfactant Species and Their Concentrations

From the results obtained above, as SHC of 59% at  $C_{ow} = 15$  wt% and 44% at  $C_{ow} = 50$  wt% couldn't be microencapsulated, the effects of oil soluble surfactant species and their concentrations were investigated in order to increase the microencapsulation efficiency.

**Figure 7** shows the effects of oil soluble surfactant species and their concentrations on the viscosity of shell material solution and the sedimentation velocity.

The viscosity of shell material solution increased with the concentration ( $C_o$ ) of each oil soluble surfactant. Also, as the viscosity in the case of PR-300 become slightly higher than that in the case of SBL, the sedimentation velocity in the case of SBL become slightly higher than that in the case of PR-300. The sedimentation velocity for PR-300 de-

creased from 0.055 mm/min (0.92 μm/s) at  $C_o = 0$  to 0.048 mm/min (0.80 μm/s) at  $C_o = 10$  wt%. Next, it was investigated whether the concentration of oil soluble surfactant affected the melting behavior of shell material or not.

**Figure 8** shows the effect of concentration of oil soluble surfactant on the melting behavior of shell material at  $C_{ow} = 50$  wt%, where PR-300 was used because of making the viscosity higher and the sedimentation velocity lower as shown in **Figure 7**.

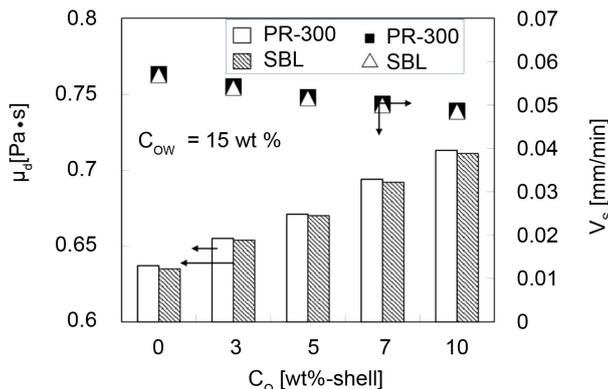
From **Figure 8**, it was found that the melt starting temperature ( $T_s = 36.5^\circ\text{C}$ ) and the peak melting temperature ( $T_p = 37.9^\circ\text{C}$ ) were not almost affected by the concentration of PR-300.

**Figure 9** shows the dependencies of the microencapsulation efficiency and the mean diameter of microcapsules on the concentration of PR-300.

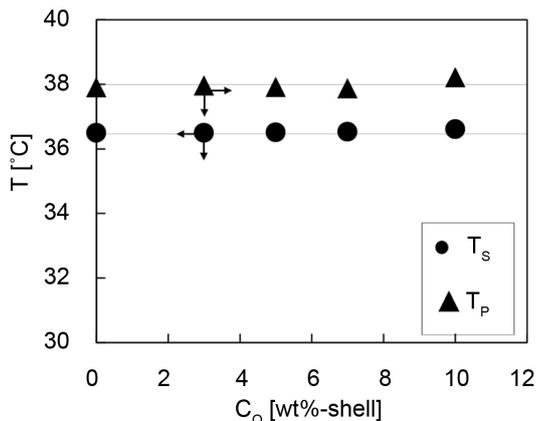
The microencapsulation efficiency could be increased from 42% at  $C_o = 0$  to 54% at  $C_o = 10$  wt%.

Meanwhile, the mean diameter slightly increased with the concentration, too.

Hitherto, as the microencapsulation efficiency could be increased by making the viscosity of shell material solution higher, the effect of the concentration of PR-300 on the microencapsulation efficiency was investigated at  $C_{ow} = 50$  wt%, at which the highest



**Figure 7.** Dependencies of viscosity of shell material solution and sedimentation velocity on oil soluble surfactant species and concentration.



**Figure 8.** Effect of concentration of oil soluble surfactant on melting temperature of shell material.

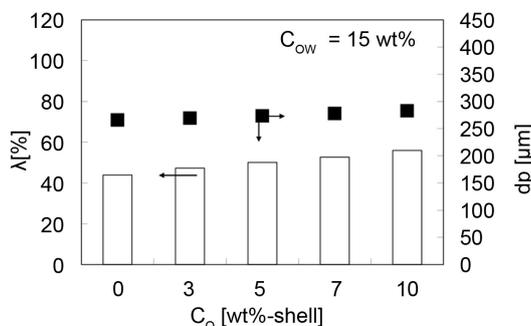
microencapsulation efficiency was obtained as shown in **Figure 6**.

**Figure 10** shows the dependencies of microencapsulation efficiency and the mean diameter on the concentration of PR-300 at  $C_{ow} = 50$  wt%.

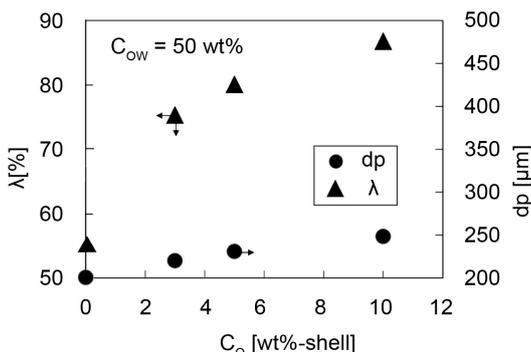
The microencapsulation efficiency could be increased considerably from 56% at  $C_o = 0$  to 88% at  $C_o = 10$  wt% due to the increase in the viscosity of shell solution and the mean diameter, especially due to the increase in the dispersing stability of SHC in the shell material solution by the help of PR-300 [23] [24].

### 3.4. Thermosensitivity of Microcapsules and Carbon Dioxide Gas Generation

**Figure 11** shows the results observing thermosensitivity (**Figure 11(a)**) of microcapsules and carbon dioxide generation (**Figure 11(b)**) by use of the microcapsules prepared at  $C_{ow} = 50$  wt% and  $C_o = 10$  wt% of PR-300, where **Figure 11(b)** shows the color of dye aqueous solution without the microcapsules (left side) at each pH and the color of the dye aqueous solution with the microcapsules at each temperature (right side), respectively. The microcapsules partially melted at 36°C as shown in **Figure 11(a)**. The color of the dye aqueous solution with the microcapsules at 30°C was orange (**Figure 11(b)**—right side), which is the same color as the color of the dye aqueous solution without the microcapsules at pH 1.7 and 4.0 (**Figure 11(b)**—left side), because the microcapsules were not melted at 30°C and SHC was not released as yet.



**Figure 9.** Effect of concentration oil soluble surfactant (PR-300) on microencapsulation efficiency and mean diameter.



**Figure 10.** Dependencies of microencapsulation efficiency and mean diameter on concentration of oil soluble surfactant (PR-300).

However, the color changed from red at 30°C to blue at 36°C and 40°C. This should mean that the microcapsules were melted in the acid solution at 36°C and 40°C, released SHC and generated carbon dioxide to change the dye aqueous solution to the alkaline solution.

From these results, it was confirmed that the microcapsules showed thermosensitivity and could generate carbon dioxide.

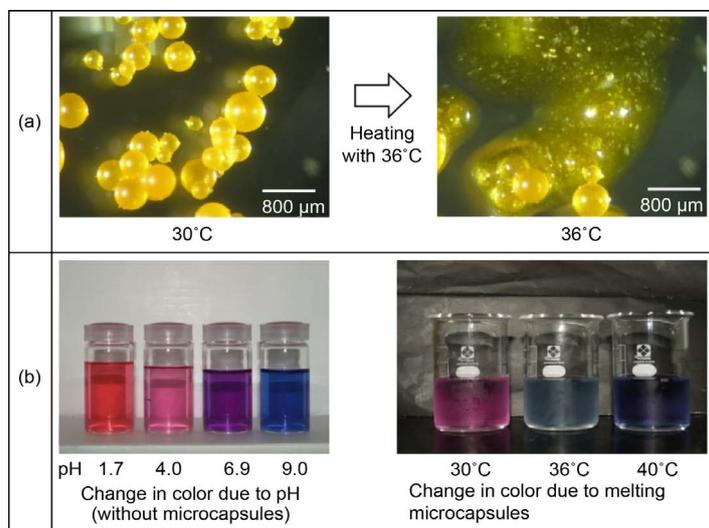
### 3.5. Optimum Preparation Conditions

From the results obtained in this study, it was found that the microencapsulation efficiency was significantly affected by the CPAO concentration, the oil soluble surfactant species and their concentration and the addition of  $\alpha$ -tocopherol as a modifier for shell material, because these affected the physical properties of liquids concerned, the diameter of dispersed droplets and affinity between the shell material solution and SHC.

The optimum preparation conditions to obtain the higher microencapsulation efficiency obtained under the fundamental conditions such as the mixing speed at the microencapsulation process, the volume fraction of the shell material solution, temperature and the experimental apparatus are shown in **Table 2**.

## 4. Conclusions

The microcapsules containing sodium hydrogen carbonate were prepared with the melting dispersion cooling method and the following results were obtained.



**Figure 11.** Thermosensitivity of microcapsules.

**Table 2.** Optimum preparation conditions.

Cow [wt%]	Oil soluble surfactant	Co [wt%]
50	PR-300	10
$dp$	250 $\mu\text{m}$	
$\lambda$	88 [%]	

- 1) The sound spherical microcapsules could be prepared under all the experimental conditions adopted in this study.
- 2) The microencapsulation efficiency could be increased by increasing the viscosity of the shell material solution.
- 3) The microencapsulation efficiency was affected by the CPAO concentration,  $\alpha$ -tocopherol and the oil solution surfactant species and their concentration.
- 4) The optimum preparation conditions to obtain the larger microencapsulation efficiency could be obtained.
- 5) The microcapsules showed thermosensitivity and generated carbon dioxide gas.
- 6) The microencapsulation procedure developed in this study may be applied to microencapsulate the various water soluble solid powders generating the desired gas instead of microencapsulating directly the gaseous core materials. These microcapsules may be applied to the gas delivery system in the future.

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