Global Journals LaTeX JournalKaleidoscope™
Artificial Intelligence formulated this projection for compatibility purposes from the original article published at Global Journals. However, this technology is currently in beta. *Therefore, kindly ignore odd layouts, missed formulae, text, tables, or figures.*

Preparation of Microcapsules Containing Grape Polyphenols and A-Tocopherol by Spray-Gelling Method

Masato Tanaka[1], Yoshinari Taguchi[2] and Masato Tanaka[3]

[1] Niigata University

Received: 9 December 2013 Accepted: 3 January 2014 Published: 15 January 2014

Abstract

It was tried to prepare the microcapsules containing the (W/O) emulsion or the (S/O) dispersion by the Spray-Gelling method. The shell material was gelled sodium alginate. In the case of preparation of the microcapsules containing the (W/O) emulsion, grape polyphenol as the first core was dissolved in the inner water droplets which were dispersed in ?-tocopherol oil as the second core. In the case of preparation of the microcapsules containing the (S/O) dispersion, grape polyphenol powder as the first core was dispersed in the ?-tocopherol oil as the second core. Two kinds of multiple emulsions such as the (W/O)/W emulsion and the (S/O)/W emulsion, in which the sodium alginate aqueous solution was the continuous water phase, were prepared and sprayed into the calcium chloride aqueous solution as the gelling agent through the nozzle. The microcapsules with the mean diameters from 20 to 70 ?m could be prepared. The microencapsulation efficiency was increased by changing the (W/O) emulsion to the (S/O) dispersion.

Index terms— sodium alginate microcapsule, multiple emulsion, grape polyphenol, spray-gelling method, dual core materials, ?-tocopherol.

1 Introduction

itherto, many kinds of microcapsules have been prepared and applied in the various fields such as cosmetics, food, drugs, paintings, adhesives, textile, electric materials and so on [1][2][3].

The main functions of microcapsules are to protect the core materials from environment for a long time, to optionally release the core materials according to stimuli, to modify the surface of core material [1]. 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. If a few core materials with the different chemical and physicochemical properties could be microencapsulated at the same time, the multiple functions may be given to the microcapsules. For an example, in the case of applying the microcapsules to the food, cosmetics, drugs and textile, the nontoxic materials have to be used as the shell material. Furthermore, if one of the core materials tried to microencapsulate at the same time is hydrophilic and the other hydrophobic, the newly devised microencapsulation procedure has to be developed by using the designated core and shell materials.

It is well known that grape polyphenol is water soluble and has a few physiological effects such as antiaging effect and anti-oxidation effect, but has a few defects such as a bitter taste and reaction activity. Accordingly, if grape polyphenol could be microencapsulated with some edible shell materials, the application fields of grape polyphenol may be considerably extended [4].

On the other hand, ?-tocopherol (vitamin E) has a few physicological effects such as anti-aging effect and prevention effect of lifestyle related disease, but has a few defects such as light and heat destruction and easy oxidization [5][6][7]. Accordingly, if ?-tocopherol oil could be microencapsulated with some nontoxic materials, various application fields for ?-tocopherol may be expected. Furthermore, the microcapsules with the multiple

functions can be prepared by microencapsulating these core materials at the same time. However, as ?-tocopherol is hydrophobic and grape polyphenol is water soluble, in order to prepare the microcapsules containing these core materials as much as possible, it is necessary to develop the preparation method by using the nontoxic shell materials.

The spray methods such as spray drying, spray chilled and spray gelling for preparing the various kinds of microcapsules are well known to be the effective methods, because the selectivity for the core and the shell materials is extremely wide and the continuous production is capable [8,9]. Also, the diameters of microcapsules can be easily controlled by changing the volumetric flow velocity of feed and the spraying pressure [10][11][12]. Accordingly, the spray method may be suitable to the preparation of microcapsules containing the dual cores with the different chemical and physicochemical properties at the same time.

The purposes of this study are to develop the preparation method of microcapsules containing the hydrophilic and the hydrophobic core materials, to characterize the microcapsules and to discuss the formation mechanism.

2 II.

3 Experimental a) Materials

Materials used to prepare the microcapsules containing the dual core materials were as follows.

Grape Polyphenol (GP) (Sunprite Ind. Co., Ltd, Tokyo, Japan) was used as the first core material and ?tocopherol (?-oil) (Wako Pure Chemical Ind. Co., Ltd, Tokyo, Japan) was used as the second core material. Sodium Alginate (Alg?Na) (Wako Pure Chemical Ind. Co., Ltd, Tokyo, Japan) was used as the shell material. Calcium chloride (Wako Pure Chemical Ind. Co., Ltd, Tokyo, Japan) was used as the gelling agent for sodium alginate. Poem PR-100, DAO-7S and Soy bean Lecithin (SL) (Riken Vitamin Ind. Co., Ltd, Tokyo, Japan) were used as the oil soluble surfactants. ML-750 (SY Glystar : Sakamoto Yakuhin Ind. Co., Ltd, Tokyo, Japan) was the water soluble surfactant. b) Verification of effect of oil soluble surfactant species on stability of (W/O) emulsion and (W/O)/W emulsion

In order to increase the content of core materials as much as possible, it is necessary to stabilize the (W/O) emulsion and the (W/O)/W emulsion with the help of surfactant species. For this purpose, the effect of oil soluble surfactant species on the stability of (W/O) emulsion and the (W/O)/W emulsion was estimated as follows.

The GP aqueous solution of a given volume was poured into the ?-oil dissolving the oil soluble surfactant and homogenized to form the (W/O) emulsion.

Then, the (W/O) emulsion was observed by visual confirmation at the constant time intervals. If the stability of (W/O) emulsion is lower, the (W/O) emulsion may be broken and the phase separation may be observed.

Furthermore, the (W/O) emulsion thus prepared was poured into the Alg?Na aqueous solution and homogenized to form the (W/O)/W emulsion. The stability of (W/O)/W emulsion was observed similarily as stated just above. If the stability of the (W/O)/W emulsion is lower, the (W/O)/W emulsion may be broken and the phase separation may be observed. On the other hand, two kinds of the (S/O) dispersions, where GP powder was dispersed in the ?oil, were prepared as follows.

First, the (S/O) dispersion was prepared by directly adding GP powder into the ?-oil and stirred.

Second, the (S/O) dispersion was prepared by adding the GP aqueous solution into the ?-oil, forming the (W/O) emulsion and then, by removing water from the (W/O) emulsion as shown in Figure 3. In this operation, the time for removing water from the (W/O) emulsion was changed from 0 to 12h at 50? in order to change the diameter of inner water droplets, namely the diameter of GP powder. Furthermore polyethylene glycol (PEG 600) was dissolved in the inner water phase in order to try to investigate whether the content of GP could be increased or not. In this fundamental experiment, the spraying conditions such as the volumetric flow velocity of feed and the spraying pressure, the oil soluble surfactant species and the time for removing water from the (W/O) emulsion were changed stepwise. The experimental conditions are shown in Table **??**.

4 Table 1 : Experimental conditions d) Characterization Mean diameter

The diameters of inner water droplets and GP powder were measured by the instrument for measuring the particle size (Otsuka Denshi Co., Ltd, Tokyo, Japan: ELS-8000).

The diameters of microcapsules were measured by the particle size analyzer (Shimazu Seisakusho Co., Ltd, Kyoto, Japan: SALD-3000)

Here, the mean diameters (dp) were obtained as the mean Sauter diameters.

5 Microencapsulation efficiency

The microcapsules of a given weight were added into distilled water of 100 cm3, broken by the homogenizer and then, irradiated supersonic for 10 min to perfectly dissolve GP in the water phase.

The amount of GP dissolved in the sampled water was measured by the spectrophotometer.

For this measurement, the calibration curve between absorption degree and the concentration of GP was obtained beforehand.

The microencapsulation efficiency (Fc) was estimated by the following equation.

6 Observation of emulsion and microcapsules

The (W/O)/W emulsion, the (S/O)/W emulsion and the microcapsules were observed by the optical microscope (OLXMPUS Co., Ltd, Tokyo, Japan: BHT-MV) and their photographs were taken by digital camera. From these photographs, the stability of the multiple emulsions and the formation of microcapsules were observed.

7 III.

Results & Discussion a) Effect of oil surfactant species on stability of (W/O) emulsion and (W/O)/W emulsion Figure **??** shows the results investigating the effect of oil soluble surfactant species on the stability of the (W/O) emulsion (Figure **??**a) and the (W/O)/W emulsion (Figure **??**b). Here, the transient feature of each emulsion was measured to estimate the stability. From Figure **??**a, the following valuable results were obtained.

Just after preparing the (W/O) emulsion, in the case of PR-100 and SL, the (W/O) emulsion was brown color, but in the case of DAO-7S, the (W/O) emulsion was dark brown color. Here, the dark brown color means that the (W/O) emulsion is broken and the GP aqueous solution is separated. 6 shows the optical microscopic photographs of microcapsules prepared under the same conditions as in Figure 5. From these photographs, it was found that there were spherical and irregular microcapsules and many ?-oil droplets were microencapsulated well with the gelled Alg?Na shell. Figure 7 shows the dependences of the mean diameters (dp) and their dispersion degree (?/dp) of microcapsules on the spraying pressure (P). Here, ? is the standard deviation of distribution of diameters. The mean diameters and the dispersion degrees decreased from 63 ?m to 25?m and from 3.2 to 1.7 with increasing the spraying pressure, respectively and almost become constant at P=0.3 **??**MPa]. From these results, it was found that the smaller and more uniform microcapsules could be prepared by increasing the spraying pressure. Figure 8 shows the dependence of the microencapsulation efficiency (?) on the spraying pressure. The microencapsulation efficiency was almost kept constant (?=30%) in spite of increasing the spraying pressure. This lower microencapsulation efficiency should be attributable to the unstable (W/O) emulsion. Figure 11 shows the transient water droplet diameters (dpW) with the time for removing the water phase. From this figure, it was found that the mean diameters of inner aqueous droplets were gradually decreasing with the removing time and become constant at the point of elapsing 6h and the (W/O) emulsion was changed to the (S/O) dispersion. Accordingly, the mean diameters become equal to the diameters of GP powder. Figure 12 shows the dependence of the mean diameter of inner water droplets on the concentration (CL) of SL (Figure 12(a)) and that of the mean diameters (dpS) of GP powder particles on the mean diameter of inner water droplets (Figure 12 (b)). The mean diameters of inner water droplets decreased with the surfactant concentration and the GP powder particle diameters was in proportion to the mean diameters of inner water droplets. It is well known that the content of core materials can be increased by decreasing the diameters of core materials [12][13]. Accordingly, the stability of the (S/O) dispersion could be increased by increasing the surfactant concentration. As a result, the microencapsulation efficiency could be increased.

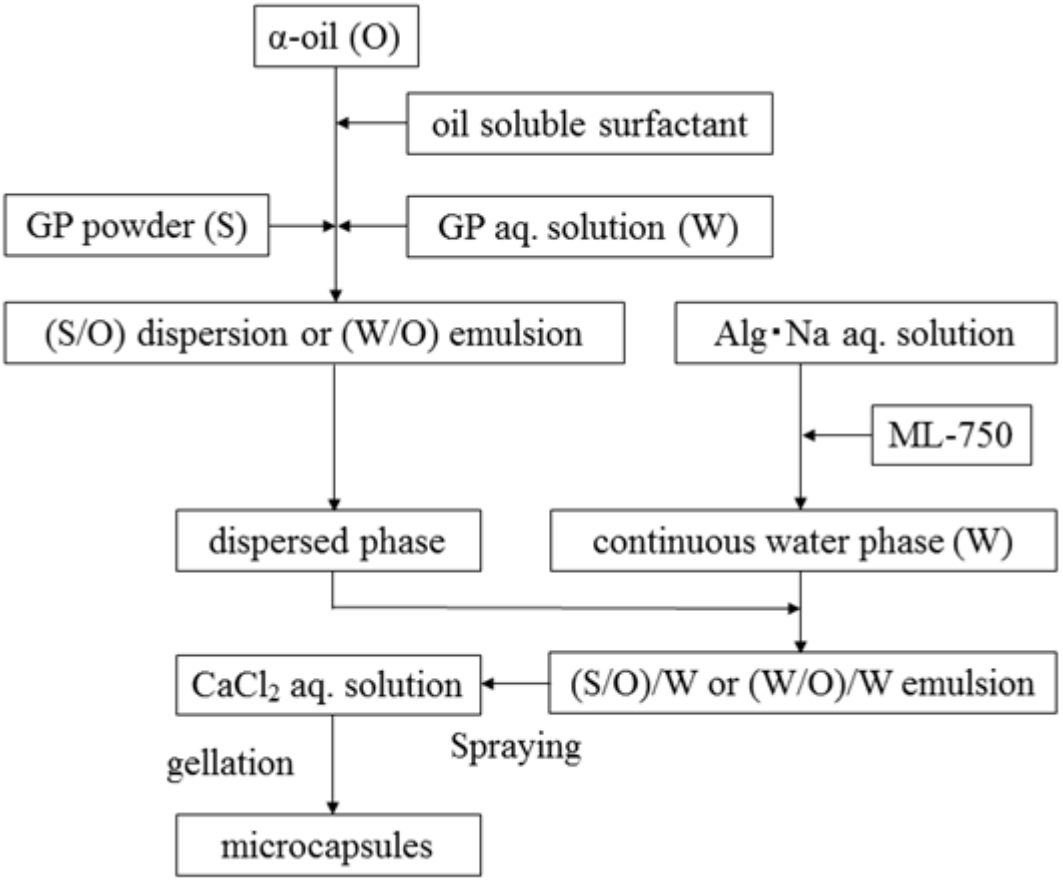
Figure 13 shows the dependence of the microenc-apsulation efficiency (?) on the mean diameters (dpW) of inner water droplets and the GP powder particle diameters (dpS), respectively. From Figure 13, it was found that the microencapsulation efficiency could be increased by decreasing the diameters of inner water droplets and GP powder particles and the microencapsulation efficiencies in the case of the (S/O) dispersion was larger than those in the case of the (W/O) emulsion. Furthermore, the addition of polyethylene glycol (PEG 600) in the inner water phase could considerably increase the microencapsulation efficiency. The effect of addition of PEG may be attributable to the fact that the adsorption layer of PEG on the surface of GP powder particles was formed as shown in Figure 11 (b) and the stability of GP powder in the ?-oil could be increased due to increase in affinity between GP powder and the ?-oil. In order to increase the content of core material, the formation of finer water droplets dissolving the hydrophilic core material, the formation of the (S/O) dispersion by removing water from the inner water and the formation of adsorption layer of stabilizer on the surface of powder particles were found to be considerably effective.

IV.

8 Conclusion

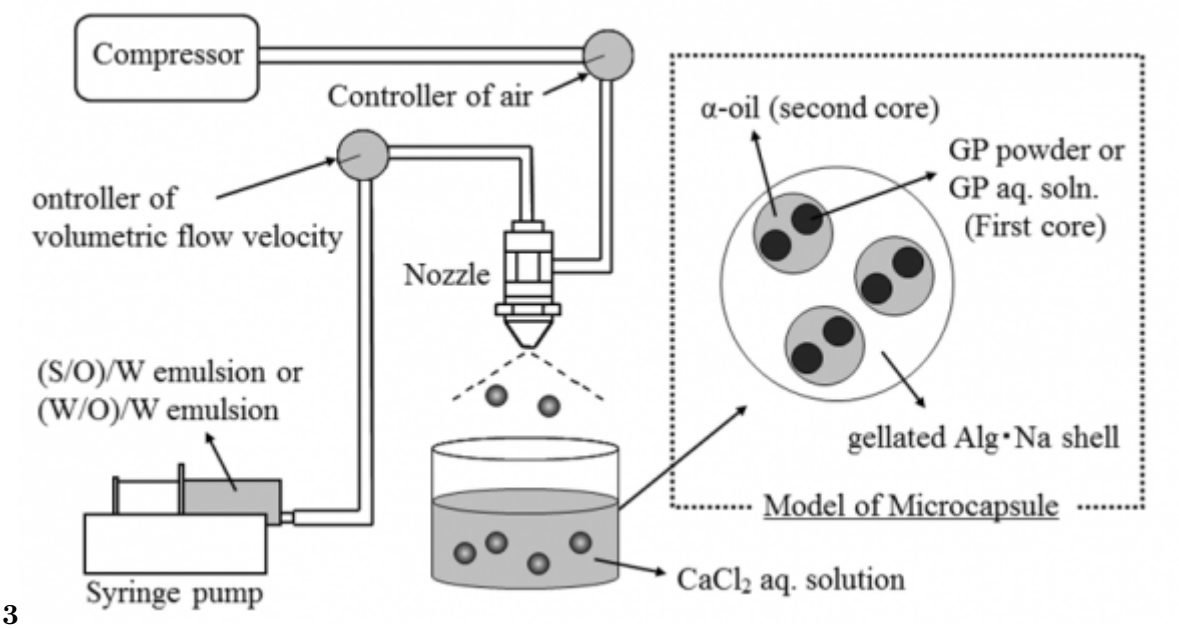
It was tried to prepare the microcapsules containing the dual cores, namely the (W/O) emulsion or the (S/O) dispersion by the spray-gelling method. The following results were obtained. The microcapsules with the mean diameters from 15?m to 70?m could be prepared.

Figure 1:



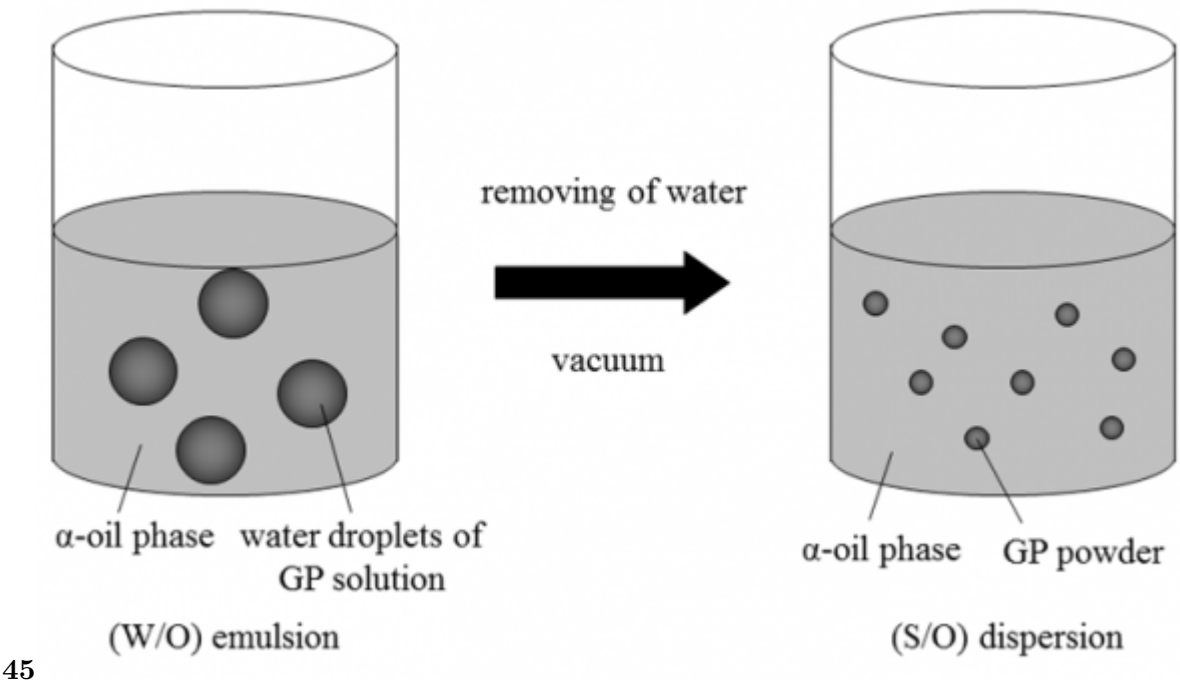
12

Figure 2: Figure 1 :Figure 2 :



3

Figure 3: Figure 3 :



45

Figure 4: Figure 4 :Figure 5

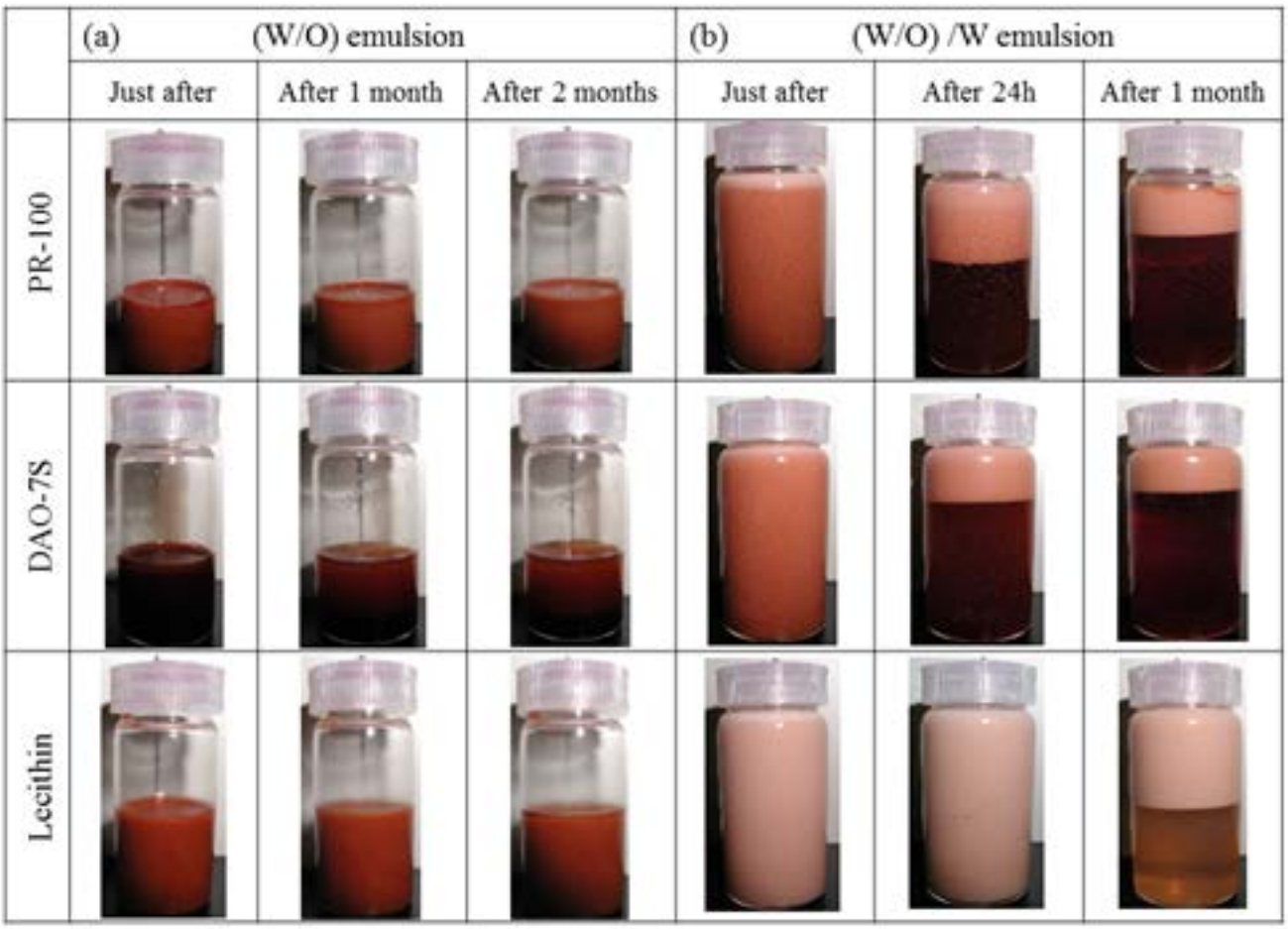


Figure 5: Figure 5 :

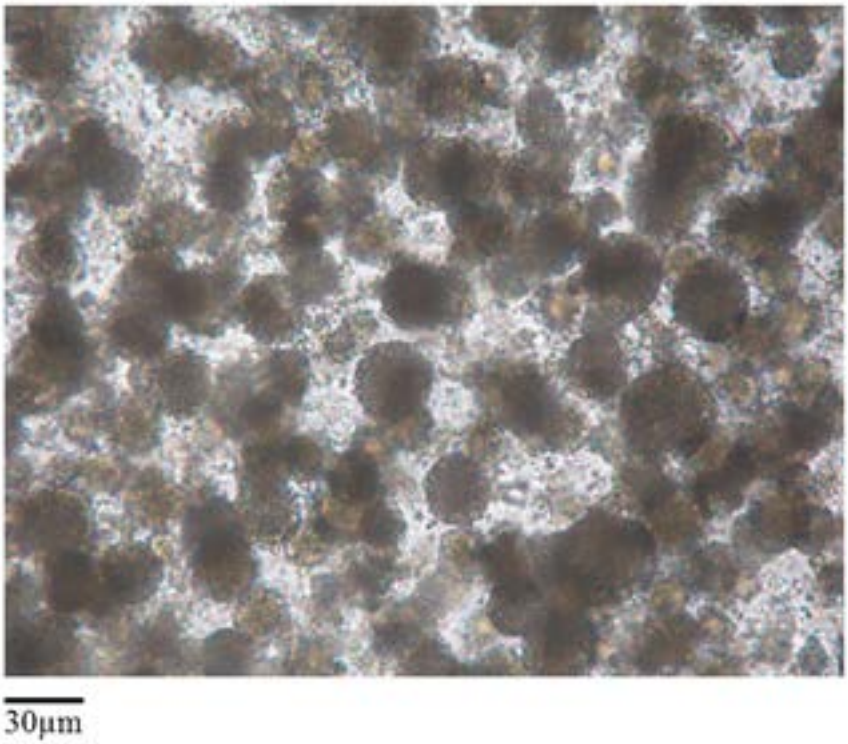
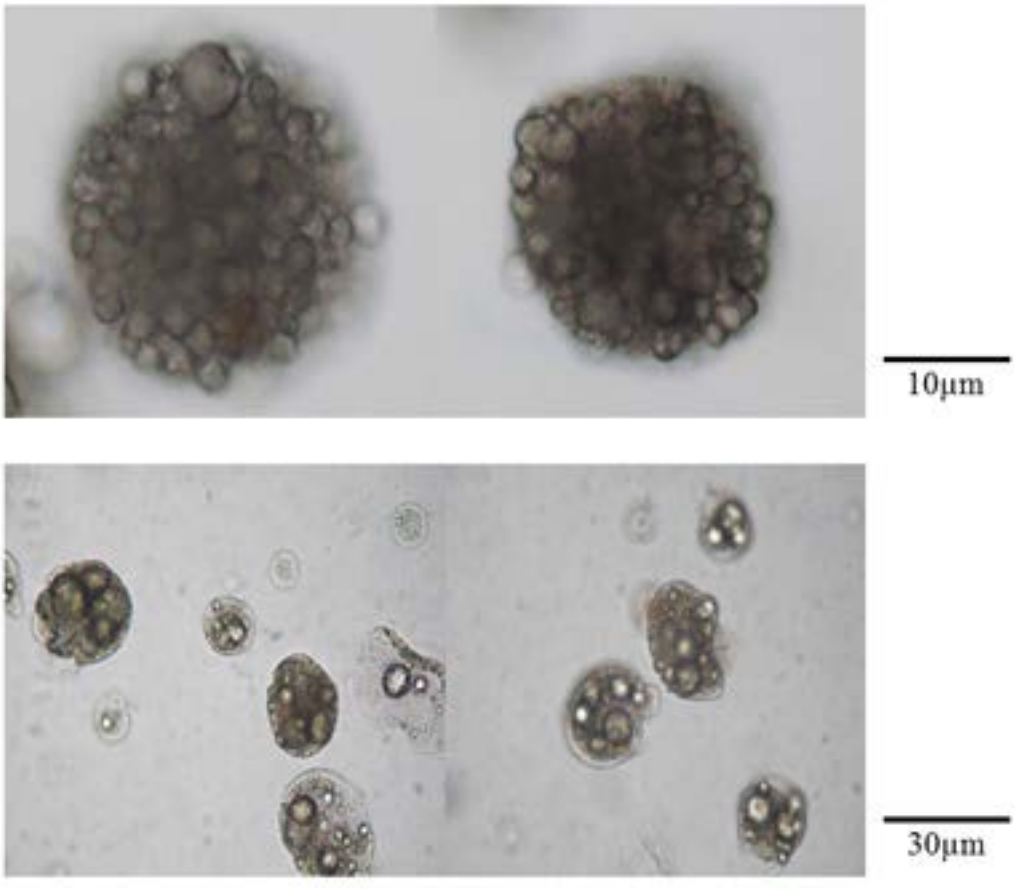
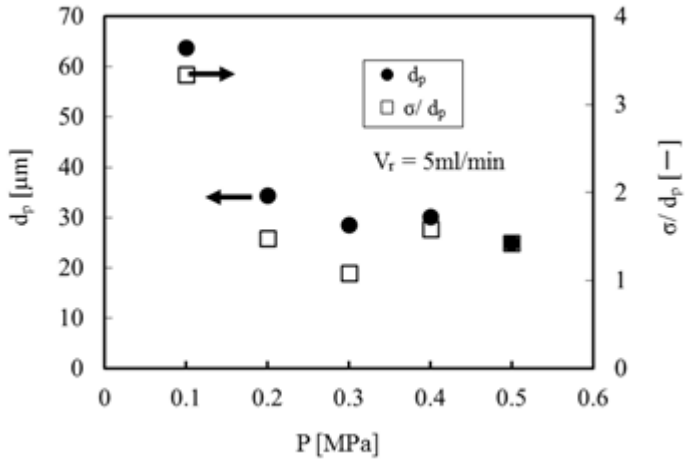


Figure 6: Figure



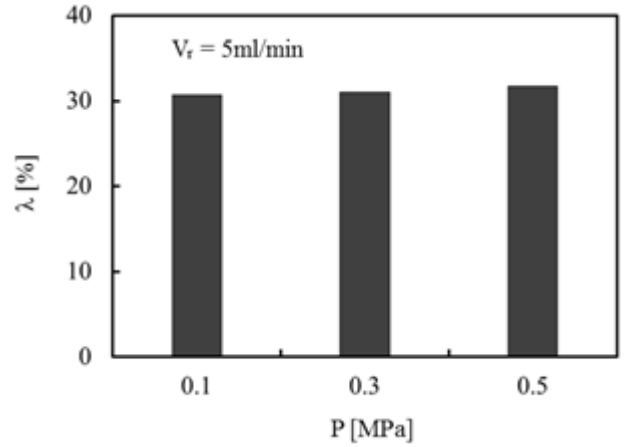
6

Figure 7: Figure 6 :



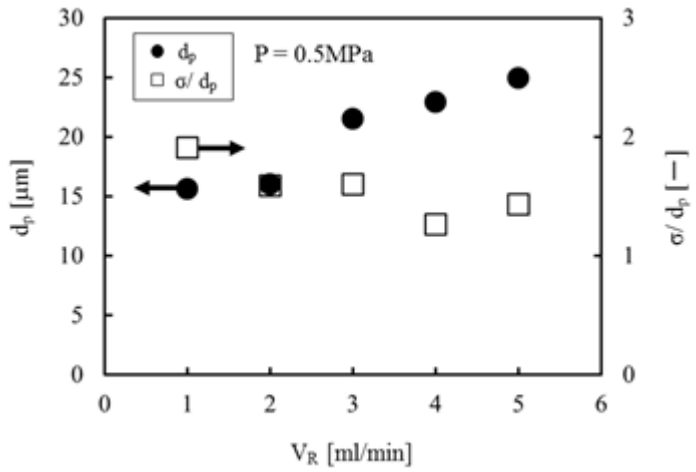
7

Figure 8: Figure 7 :



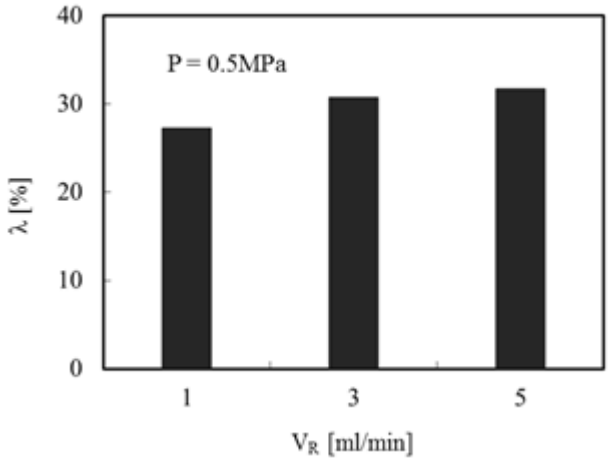
8

Figure 9: Figure 8 :



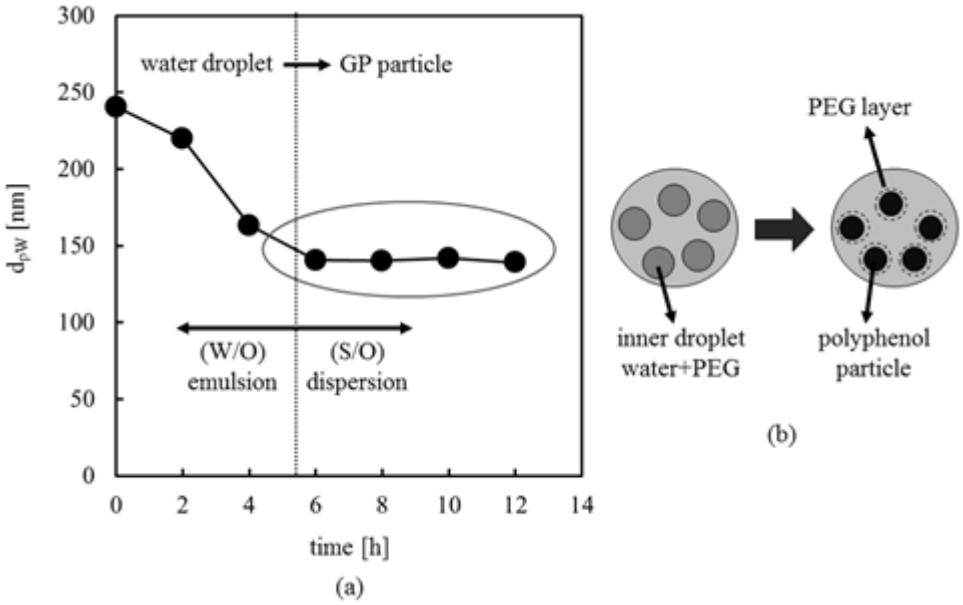
109

Figure 10: Figure 10 Figure 9 :



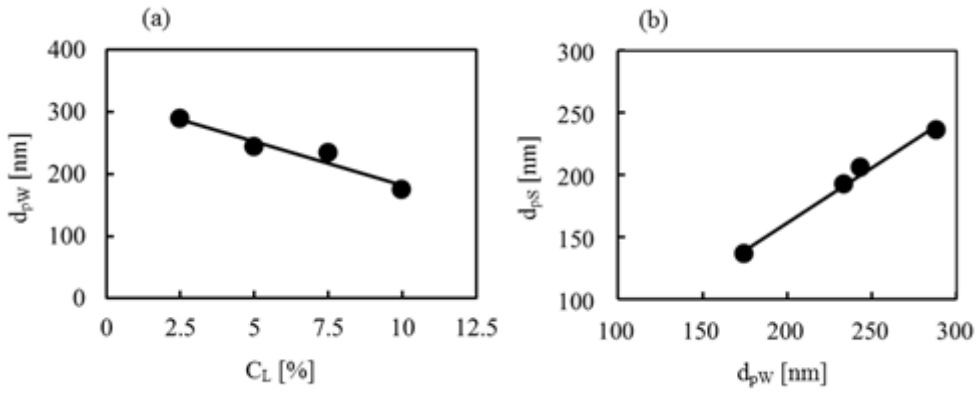
11

Figure 11: Figure 11 :



12

Figure 12: Figure 12 :



1

Figure 13: 1 .

[Song et al. ()] '?-Tocopherol-loaded Ca-pectinate microcapsules: Optimization, in vitro release, and bioavailability'. Y B Song , J S Lee , H G Lee . *Colloids and Surfaces B: Biointerfaces* 2009. 73 p. .

[Xia et al. ()] 'Biological Activities of Polyphenols from Grapes'. E Q Xia , G F Deng , Y J Guo , H B Li . *International Journal of Molecular Sciences* 2010. 11 p. .

[Yokoyama et al. ()] 'Effect of Viscosity of Shell Solution on the Content of Solid Powder as Core Material in Microencapsulation by the Drying-in-Liquid Method'. H Yokoyama , L Mo , Y Taguchi , M Tanaka . *Journal of Applied Polymer Science* 2008. 109 p. .

[Somchue et al. ()] 'Encapsulation of a-tocopherol in protein-based delivery particles'. W Somchue , W Sermsri , J Shiowatana , A Siripinyanond . *Food Research International* 2009. 42 p. .

[Polavarapu et al. ()] 'Impact of Extra Virgin Olive Oil and Ethylenediaminetetraacetic Acid (EDTA) on the Oxidative Stability of Fish Oil Emulsions and Spray-Dried Microcapsules Stabilized by Sugar Beet Figure 13 : Dependence of microcapsulation efficiency on inner water droplet diameter and particle diameter Pectin'. S Polavarapu , C M Oliver , S Ajlouni , M A Augustin . *Journal of Agricultural and Food Chemistry* 2012. 60 p. .

[Soottitantawat et al. ()] *Influence of emulsion and powder size on the stability of encapsulated D-limonene by spray drying. Innovative Food Science and Emerging Technologies*, F Soottitantawat , H Bigeard , T Uoshii , A Furuta , P Ohkawara , Linko . 2005. 6 p. .

[Tanaka ()] *Key Point of Preparation of Nano/Microcapsules*, M Tanaka . 2008. Tokyo: Techno System Publishing Co. Ltd.

[Koishi et al. ()] M Koishi , K Eto , H Higure . *Preparation + Utilization) Microcapsules. Kogyo Chosakai*, (Tokyo) 2005.

[Yoo et al. ()] 'Microencapsulation of ?-tocopherol using sodium alginate and its controlled release properties'. S H Yoo , Y B Song , P S Chang , H G Lee . *International Journal of Biological Macromolecules* 2006. 38 p. .

[Takahashi et al. ()] 'Microencapsulation of Hydrophilic Solid Powder as Fire Retardant Agent with Epoxy Resin by Droplet Coalescence Method'. M Takahashi , Y Taguchi , M Tanaka . *Journal of Applied Polymer Science* 2008. 110 p. .

[Laohasongkram et al. ()] 'Microencapsulation of Macadamia oil by spray drying'. K Laohasongkram , T Mahamaktudsanee , S Chaiwanichsiri . *Procedia Food Science* 2011. 1 p. .

[Shirokawa et al. ()] 'Preparation of Temperature and Water Responsive Microcapsules Containing Hydroquinone with Spray Drying Method'. K Shirokawa , Y Taguchi , H Yokoyama , F Ono , M Tanaka . *Journal of Cosmetics* 2013. 3 p. .

[Gharsallaoui et al. ()] 'Properties of spraydried food flavours microencapsulated with twolayered membranes: Roles of interfacial interactions and water'. G Gharsallaoui , L Roudaut , O Beney , A Chambin , R Voilley , Saurel . *Food Chemistry* 2012. 132 p. .

[Kondo ()] *Saishin Maikurokapseruka Gijutsu (Microencapsulation Technique)*, T Kondo . 1967. TES, Tokyo.