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Optimization of the conditions for producing water-in-oil-in-water microemulsions and spray-dried microcapsule of tomato extract powder

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Abstract

The objective of this study was to investigate optimized conditions for producing water-in-oil-in-water (W/O/W) microemulsions and spray-dried microcapsules of tomato extracts. The condition of highest yield for W/O/W microemulsification was optimized by response surface methodology (RSM). The independent variables were ratio of tomato extract to MCT (X_1: 1:9-2:8), concentrations of emulsifier (X_2: 0.50-1.00), and ratio of W/O to secondary aqueous coating material (X_3: 1:5-1:9). According to the RSM results, the ratio of tomato extract to MCT at 1.24:8.76 (v/v), concentration of emulsifier at 1.00% (w/v), and ratio of W/O phase to secondary aqueous coating material of 1.00:6.16 (v/v) were found to be the optimal conditions for producing microemulsion of tomato extracts. At these conditions, the yield of W/O/W microemulsion determined as approximately 99.69%. After optimizing the conditions for producing microemulsion of tomato extract by RSM, the emulsion was spray-dried. The average particle size ($D_{(4,3)}$) and zeta-potential value of the spray-dried microcapsules under optimized condition were approximately 5 μm and -29.68 mV, respectively. Results of the present study provide practical information on optimum conditions for producing W/O/W microemulsions and spray-dried microcapsules of tomato extracts.

Keywords: tomato extract; microemulsion; spray-dried microcapsule; response surface methodology.

Practical Application: W/O/W microcapsules can be applied to various food systems such as beverage and dairy food because dosage can be changed by using microencapsulation technique.

1 Introduction

Tomato is traditionally recognized as the plentiful source of lycopene (Edwards et al., 2003). Lycopene is a polyunsaturated (polyene) straight-chain molecule, which contained 13 double bonds with a molecular weight of 536.9 daltons. It has strong antioxidant and other beneficial effects, including free radical scavenging (Hernandez-Marin et al., 2013). Antioxidant effects of lycopene might include scavenging free radicals from reactive oxygen species (ROS) because lycopene is very effective to remove singlet oxygen in *in vitro* and *in plasma* (Zhang et al., 2014). However, lycopene in tomato is hardly to obtain since tomato extract has poor water solubility and is easily oxidized by light or heat exposure. To combat those issues, microencapsulation can be a good approach and it could be a good solution to be utilized tomato extracts into food system.

Microencapsulation can be an approach to enhance stability of lycopene, and to enable its dispersion in an aqueous media (Rocha et al., 2012). This technology has been successfully used in the food system to preserve its core materials that are susceptive to temperature, light, oxygen and humidity. Encapsulation is one of economical packaging technique for active ingredients in foods for maximum protection, targeted delivery and controlled release of nutrients (Leclercq et al., 2009; Ubbink & Schoonman, 2013; Weinbreck et al., 2004). The development of delivering sensitive and active ingredients has been newly emerged for the food industry (Shah & Ravula, 2000; Siegrist et al., 2008).

Spray drying is the most conventionally utilized technique of encapsulation for food products. The process of spray drying is economical and flexible, readily available equipment and highly qualified (Reineccius, 1988; Rosenberg et al., 1990). The effect of suitable coating materials, preparation of emulsions with core and coating materials and the drying processes affect the shape and type of capsules formed; the efficiency and rate of retention of core materials are also influenced (Graves & Weiss, 1992). The typically used coating materials in spay-drying process are maltodextrin (MD), gum arabic (AG), milk protein, soy protein and modified starch.

In the previous study, lycopene was embedded effectively in the layers of gelatin/sucrose microcapsules by spray drying with a loading of over 80% core-to-wall ratio. The benefit originated from embedding lycopene into gelatin and sucrose matrices enhanced storage stability of lycopene equal to tomato paste (Shu et al.,

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2006). Rocha et al. (2012) reported that the stability of lycopene was improved by microencapsulation. Also, Pelissari et al. (2016) showed that the microparticles loaded with lycopene by spray chilling could enhance its stability approximately 90% or more. For more bioavailability, the encapsulated bioactive ingredients should be easily released from the food, and then absorbed in the human (Shu et al., 2006; Sanz & Luyten, 2006). Ideally, the spray-dried microcapsules for tomato extracts should have instant properties or served as a source of lycopene-rich functional food ingredient for supplement into food products. Recently, Pu & Tang (2017) suggested that stability of the lycopene in *chlorella pyrenoidosa* was enhanced by encapsulation. Shi et al. (2015) also reported that the formulation of microemulsion of lycopene might be an effective way to prevent the environmental stresses on stability. According to Celli et al. (2016), the stability of lycopene from watermelon was increased in microemulsion. However, there is limited information in the literature on water-in-oil-in-water (W/O/W) microemulsions and spray-dried microcapsules of tomato extract powder containing high amount of lycopene as functional foods. Therefore, the purpose of the present study was to optimize conditions for preparing W/O/W microemulsion and spray-dried microcapsules of tomato extract powder and investigate physicochemical characteristics of such emulsions and microcapsules.

2 Materials and methods

2.1 Materials

Tomato extracts which is contained 6% lycopene (Lyco-O-Mato) was provided by Lycored Ltd. (Bear shave, Israel). Medium-chain triglyceride (MCT) was purchased from Wellga Co., Ltd. (Seongnam, Korea). As the coating materials, MD (DE 18, Samyang Genex, Seoul, Korea), AG (Samchun Pure Chemical Co., Ltd., Pyeongtaek, Korea), whey protein isolates (WPI, Davisco Foods International, Eden Prairie, MN, USA) or whey protein concentrate (WPC, Davisco Foods International, Eden Prairie, MN, USA) were used. As the emulsifiers, the polyglycerol fatty acid ester (PFAE, HLB 13), Tween 60 (HLB 14.9), Tween 80 (HLB 15) and polyoxyethylene sorbitan monooleate (PSML, HLB 16.7) were provided by Il-Shin Co., Ltd. (Seoul, Korea). All chemicals were of reagent grade unless otherwise specified.

2.2. Preparation of W/O/W microemulsion and spray-dried tomato extract microcapsules

The procedure of W/O/W tomato extract microemulsion was shown in Figure 1. To produce a primary emulsion, the different ratios (0.5:8.5, 1:9, 1.5:8.5, and 2:8, w/w) of the tomato extract to MCT were mixed in distilled water (DW), stirred at 500 rpm for 3 h, and filtered through a filter paper (Whatman No. 4). For the preparation of the secondary coating materials, different concentrations (10, 15, 20, 25 and 30%, w/v) of the secondary coating materials, such as MD, WPC, AG and WPI, were dissolved in DW. The coating materials and various emulsifiers, such as 0.25, 0.50, 0.75, 1.00 and 1.25% (w/v) PFAE (HLB 13.0), Tween 60 (HLB 14.9), Tween 80 (HLB 15.0) and PSML (HLB 16.7), were mixed at a magnetic stirrer at 400 rpm for 30 min. Each primary emulsion was gradually added to the coating material-mixed solution to make the ratios of W/O to secondary coating materials of 1:3, 1:5, 1:7 and 1:9 (v/v) with a high speed homogenizer (HMZ-20DN, Poonglim Co., Seoul, Korea) at 10,000 rpm for 3 min.

2.3 Determination of emulsion stability index (ESI)

The volumetric method was used to elucidate the emulsion stability index (ESI) for W/O/W microemulsion of tomato extracts according to Chang et al. (1994). Briefly, the emulsion was incubated at 60 °C for 2 h, and then the separated layer was formed to measure the volume. All samples were measured in triplicate. ESI was calculated by the following formula Equation 1:

$$\text{ESI } (\%) = \left[1\text{-}\left(\text{volume of separation layer / total volume of emulsion}\right)\right] \text{X}100 \quad (1)$$

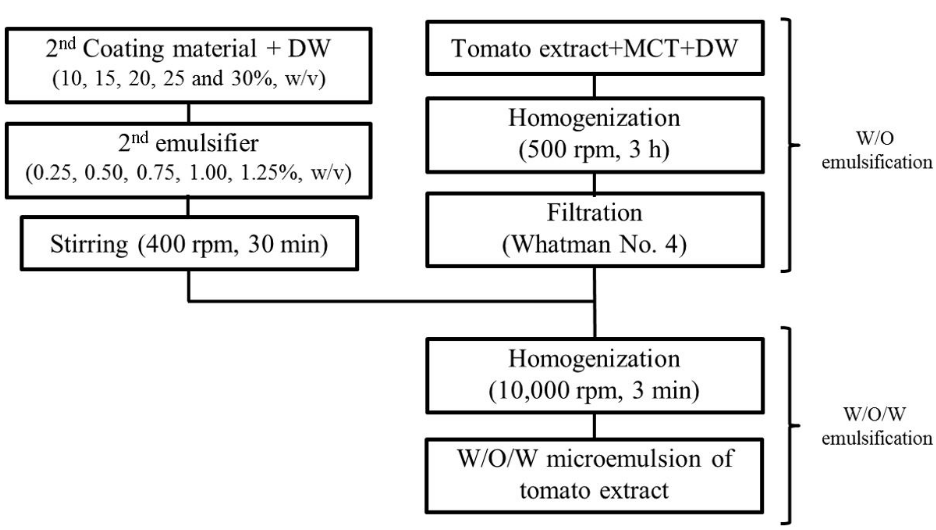

Figure 1. Procedure of W/O/W microemulsification of tomato extract. DW: distilled water; MCT: medium-chain triglyceride.

Table 1. Coded level for independent variables used in experiment design for W/O/W microencapsulation of tomato extract.

Variables	Coded X_i	Coded level		
		-1	0	+1
Ratio of tomato extract to MCT[1)] (w/w)	X_1	1:9	1.5:8.5	2:8
Concentrations of emulsifier (%, w/v)	X_2	0.50	0.75	1.00
Ratio of W/O to secondary aqueous coating material (v/v)	X_3	1:5	1:7	1:9

[1)]MCT: medium-chain triglyceride.

2.4 Optimization of conditions for producing microemulsion of tomato extract by response surface methodology (RSM)

Response surface methodology (RSM) was utilized to determine the effect of three independent variables as shown in Table 1.

Twenty experiments were designed with the principal of RSM using Minitab release 16 (Minitab, 2010) and Design Expert 7 (Stat-Ease, 2007). The quadratic polynominal regression model was estimated for the effective yield of the microencapsulation. The effective yield of the microencapsulation was calculated by following Equation 2:

$$Y = \beta_{k_0} + \sum_{i=1}^{3} \beta_{k_i} x_i + \sum_{i=1}^{3} \beta_{k_{ji}} x_i^2 + \sum_{i=1}^{2} \sum_{j=1+1}^{3} \beta_{k_{ij}} x_i x_j \quad (2)$$

where, the effective yield of the microencapsulation is response, β_{k0}, β_{ki}, β_{kiii}, and β_{kiij} are constant coefficients of intercept, linear, quadratic and interaction terms, respectively. X_i and X_j are uncoded independent variables (ratio of tomato extracts to MCT, emulsifier concentration, and secondary coating materials to W/O phase). The regression model from RSM was utilized to estimate the result by iso-response three-dimensional (3-D) surface plots.

2.5 Yield for microemulsification of tomato extract

The lycopene content was determined from the microcapsules of tomato extract by high performance liquid chromatography (HPLC) based on the method suggested by Lin & Chen (2003). The microencapsulation yield was calculated as followed Equation 3:

$$\text{Microencapsulation yield } (\%) = (\text{total lycopene in the emulsion / total lycopene in powder}) \text{X}100 \quad (3)$$

2.6 Spray drying

After optimization of conditions for producing microemulsion of tomato extract by RSM, the emulsions were spray-dried using lab scale spray-dryer (SD-1000, Tokyo Rikakikai Co., Ltd., Tokyo, Japan) under following conditions: feed flow rate was 0.5 L/h, inlet temperature was 170 °C, air flow rate was 0.70 m^3/min, atomizing pressure was 50 kPa, and outlet temperature was 90 °C.

2.7 Optical microscopy

Optical microscopy images for microemulsions of tomato extract was using a digital microscopy system (Eclipse 80i, Nikon, Tokyo, Japan). The microstructure of the microemulsion was magnified by 1,000-folds.

2.8 Scanning electron microscopy (SEM)

The morphological features of powdered microcapsules of tomato extracts were observed using the scanning electron microscopy (SEM, S-4300, Hitachi, and Tokyo, Japan). The SEM was operated at an accelerating voltage of 15.0 kV.

2.9 Particle size distribution

Particles size distribution of the powdered microcapsules of tomato extracts was analyzed by a Malvern Mastersizer 2000 (Malvern Instruments Ltd., Worcestershire, UK). The suspension was poured into a cuvette and its particle size was measured at 25 °C with fixed scattering angle at 165°. All samples were measured in triplicate and the mean particle size and distribution span were computed on the basis of a volume median diameter.

2.10 Zeta-potential measurement

Zeta-potential was measured to observe electrical stability of the spray-dried microcapsules of tomato extracts using a particle size analyzer (Delsa nano C, Beckman coulter, Inc., Fullerton, CA). All measurements were done in triplicate at fixed temperature and an angle of 25 °C and 15 °C, respectively.

2.11 Statistical analysis

Analysis of variance (ANOVA) and Duncan's multiple tests were performed to analyze the differences between groups. Data was expressed as mean ± SD and statistical significance was set at $p<0.05$. All statistical analyzes were done using SAS 9.0 (SAS, Institute Inc., NC, USA) and the graphical representations were performed using Sigmaplot 10.0 (Systat Software Inc., San Jose, CA, USA).

3 Results and discussion

3.1 Determination of ESI and emulsifier for producing W/O/W microemulsion of tomato extract

According to Almeida et al. (2015), most of common indicators for investigating deterioration of an emulsion are layer separation, and the emulsion stability is the most important among main factors which could be influenced to final products. In general, emulsion stability is influenced by type or amount of emulsifier. To obtain stable W/O/W microemulsions of tomato extract, the simple and rapid method was needed to evaluate emulsion stability. Therefore, ESI measurement could be a suitable method in this study. The results of ESI, which is investigated to optimize the ratio of W/O to aqueous phase and to select the suitable emulsifier, are shown in Figure 2.

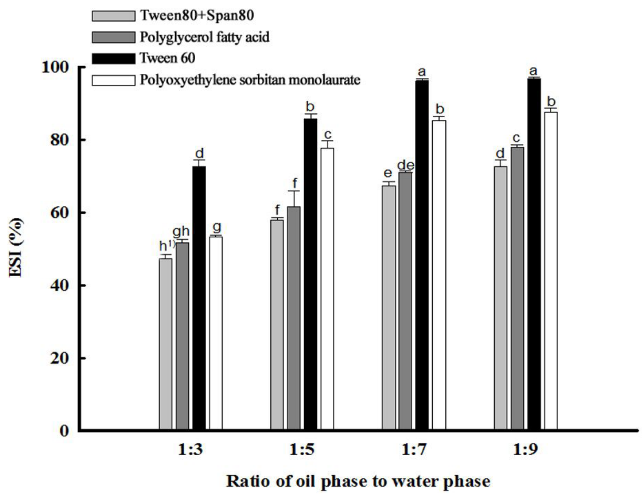

Figure 2. Effects of HLB-value of emulsifiers [Tween80+Span 80 mixed (HLB12), polygycerol fatty acid (HLB 13), Tween 60 (HLB 14.9), polyoxythylene sorbition monolaurate (HLB 16.7)] and mixing ratio of water phase to oil phase on emulsion stability index (ESI) of W/O emulsion cored with tomato extracts and coated with 10% maltodextrin. 1) Values with different superscripts are significant at p<0.05 by Duncan's multiple range test.

It is known that emulsifiers with HLB value higher than 10 are predominantly hydrophilic, thus favouring formation of W/O/W emulsion. Tween 60 (HLB 14.9) at ratios of 1:7 and 1:9 for W/O to secondary coating materials showed more than ESI of 96%. It was highly stable. ESI values were not significantly different between 1:7 to 1:9 (p>0.05).

Similarly, Spernath et al. (2002) have reported that microemulsions of lycopene can be stabilized by three emulsifiers (Tween 60, sucrose fatty acid ester, and ethoxylated monodiglyceride). However, only Tween 60 increased the solubilization of lycopene (Spernath et al., 2002). Results of the present study also indicated that Tween 60 (HLB 14.9) at ratio of tomato extract to MCT of 1:9 in W/O/W could be useful for generation of W/O/W microemulsions of tomato extract. It was reported that an increase of coating material led to an increase in the microencapsulation yield and stability of core material. Wu et al. (2014) studied microencapsulation of sulforaphane by spray-drying. In the result, as the ratio of coating material was increased and the ratio of core material decreased, the yield of microencapsulation and core material stability increased. Furthermore, Nhu Quynh et al. (2016) reported that as the amount of wall material increased, the microencapsulation yield increased in the Gac oil microencapsulation by spray-drying. Although this study has limitation which it did not optimize condition of spray-drying, it is desirable to increase the ratio of coating material from the results of these previous studies (Wu et al., 2014; Nhu Quynh et al., 2016). Therefore, it is considered that more suitable ratio of core to coating material was 1:9 (v/v).

Based on the result of the optimized ratio of W/O to secondary coating material (1:9) and emulsifier (Tween 60) as shown in Figure 2, optimized Tween 60 concentration was determined by ESI using various concentrations of Tween 60 (0.25, 0.50, 0.75, 1.00, and 1.25%) in the emulsion during keeping at 60°C for 2 h (data are not shown). As the Tween 60 concentration increased, the ESI was symmetrically increased. The ESI level reached to 95.1% at 1.00% (w/v) of Tween 60; however, the concentrations of 1.00 and 1.25% (w/v) of Tween 60 were not significantly different (p>0.05). On the other hand, it was observed that 0.25, 0.50, and 0.75% (w/v) of Tween 60 were creamed or rapidly segregated. Thus, 1.00% (w/v) of Tween 60 was selected for the emulsifier concentration in W/O/W microemulsions of tomato extracts in the present study.

To choose the optimum coating material in the concentration of 30% (w/v), ESI of the W/O/W microemulsions coated with various coating materials such as MD, WPC and AG was measured during incubation at 60 °C for 2 h (data are not shown). Among the coating materials used, MD exhibited the highest ESI value (95.3%) and significantly differ from others (p<0.05). According to Raja et al. (1989), MD with dextrose equivalence (DE) between 10 and 20 was suitable as coating material. When oil droplets were encapsulated by complex coacervation of WPI or AG, it was noticed that the emulsion was highly unstable against creaming. Therefore, it was indicated in this study that MD was the optimal coating material for the manufacturing of W/O/W microemulsion of tomato extracts.

Finally, ESI was measured to optimize the concentration of MD as the coating material for W/O/W microemulsions of tomato extracts during incubation at 60°C for 2 h (data not shown). The concentrations of 25 and 30% (w/v) MD showed higher ESI values than those of 10 to 20% (w/v), and even though the ESI values were not significantly different from 25 and 30% (p>0.05), 30% MD was higher than 25% MD in the ESI value. Yoshii et al. (2001) have showed that MD concentration played important role for spay-drying of ethyl butyrate microcapsules. Klinkesorn et al. (2004) have also reported that as the MD concentration increased further to 25 or 35%, the creaming rate decreased remarkably. Therefore, the highest concentration (30%, w/v) of MD was decided as the optimum concentration in coating material.

3.2 Optimization of conditions for producing microemulsion of tomato extract by RSM

The yield (Y) of microemulsification was optimized using statistical experimental design based on coded level from three independent variables, such as ratio of tomato extract to MCT (X_1), concentration of emulsifier (Tween 60, X_2), ratio of W/O to secondary aqueous coating materials (X_3). The mathematical relationship in the form of regression equation was given below in terms of coded factors Equation 4:

$$Y\,(\%) = 98.5901 - 2.0135X_1 + 0.9906X_2 - 0.4591X_3 - 1.3671X_1^2 \quad (4)$$

R^2 (R-squared) describes the variation in the observed responses that is explained by the model (Minitab, 2010). R^2 of the yield was described by the model as 91.93% (data are not shown). It means that this model can be explained the responses approximately 91.93% accuracy

The 3-D response surfaces for yield of microencapsulation were shown in Figure 3A, B and C. In the ratio of W/O phase, tomato extract as a core material, and concentration of Tween 60 were increased, the microencapsulation yield was increased. According to Gharibzahedi et al. (2012), RSM is a collection of useful mathematical and statistical procedures, and it needed to scrutinize multiple factors and their interactions in comparison to other approaches for possessing the advantage of reducing number of experimental trials. Min et al. (2018) optimized the nanoencapsulation efficiency with different emulsifiers using RSM in red ginseng extract nanoencapsulation. In the study, the increase of emulsifiers led to nanoencapsulation increase. In addition, the report of Lee et al. (2013) accorded with this study. The researchers reported that as the secondary emulsifier increase, the yield of microencapsulation increased in the microencapsulation of peanut sprout extract.

As apparently indicated in Figure 4, the optimum combination of factors for achieving the desired response was revealed to be the ratio of tomato to MCT (1.24:8.76, v/v), concentration of Tween 60 (1.0%, w/v) and ratio of W/O to secondary aqueous coating materials (1.00:6.16, v/v). Combining those factors makes sure the maximum yield (99.6911%) of microencapsulation.

In this study, spray-drying process variables, such as drying temperature, feed flow rate, atomizing pressure, and so on were not considered yet since lycopene is known to be more stable and have heat resistance compared to ascorbic acid in food processing (Nguyen & Schwartz, 1998; Tola & Ramaswamy, 2015) and it is considered that optimization of emulsification is more important than that of spray-drying condition.

3.3 Morphological observation

The morphology of microemulsions of tomato extracts at the optimum microencapsulation condition was analyzed with light microscopic after suspension with DW, as shown in Figure 5A. They were appeared spherical and well individualized.

After spray-drying to produce microcapsules of tomato extract, the morphology of the microcapsules was observed using SEM and presented in Figure 5B. These images revealed that spray-dried microcapsules aggregated. They showed spherical

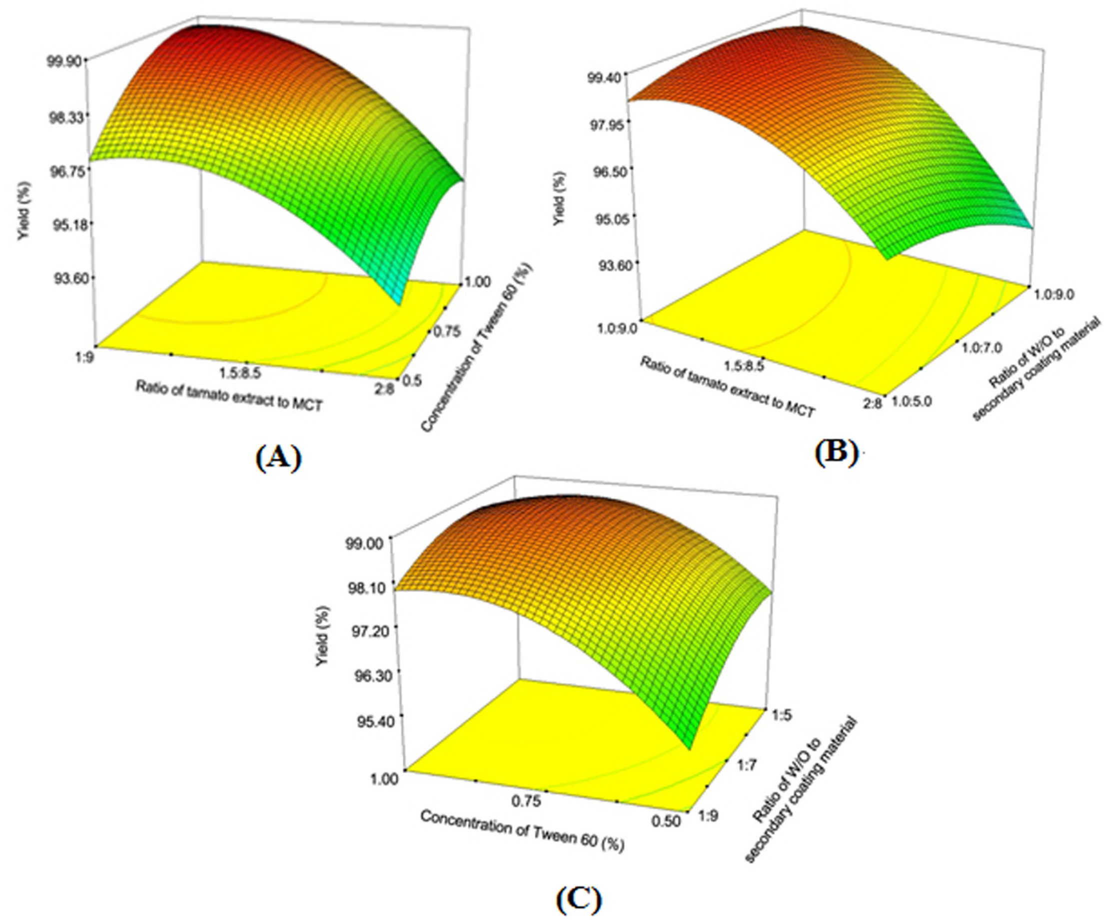

Figure 3. Response surface plots for the effects of variables on the yield (%) of microencapsulation of tomato extract. (A) ratio of tomato extract to medium-chain triglyceride (MCT) and concentration of Tween 60; (B) ratio of tomato extract to medium-chain triglyceride (MCT) and ratio of W/O to secondary coating material; (C) concentration of Tween 60 to ratio of W/O to secondary coating material.

Range	Ratio of tomato extract to MCT[1)]	Concentration of Tween 60 (%)	Ratio of W/O to secondary coating material
High	2:8	1.0	1:9
Current	[1.24:8.76]	[1.0]	[1.00:6.16]
Low	1:9	0.5	1:5

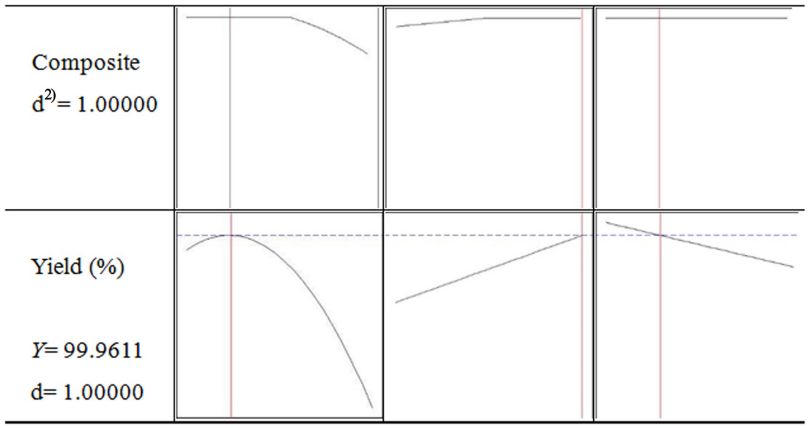

Figure 4. Optimum conditions of selective microencapsules of tomato extracts by response surface methodology. [1)]MCT: medium-chain triglyceride; [2)]d: desirability.

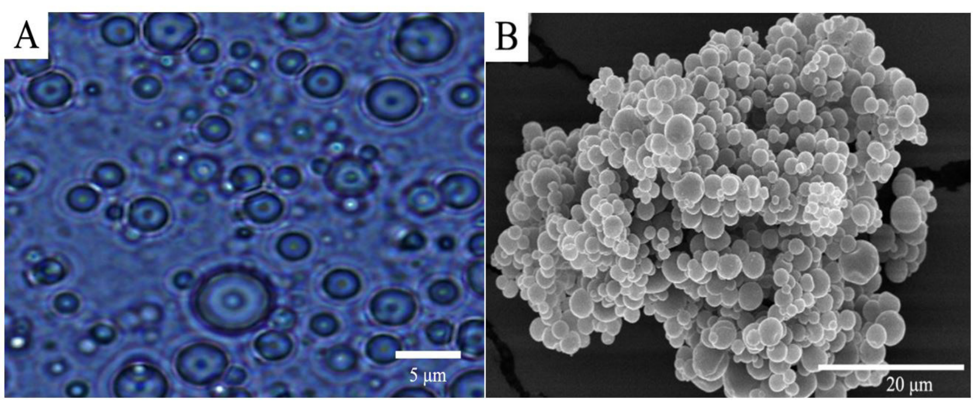

Figure 5. Microphotographs of W/O/W emulsion for tomato extracts coated with 30% maltodextrin (A) and scanning electron microphotographs of spray-dried microcapsules of tomato extracts coated with 30% maltodextrin (B).

and smooth surfaces. Ersus & Yurdagel (2007) reported that morphology of the microcapsules coated with MD showed smooth surfaces. According to Fang & Bhandari (2010), the typical form of encapsulated particles produced by spray drying was globular.

3.4 Particle size distribution

Size distribution profiles of spray-dried microcapsules of tomato extract were measured with a particle size analyzer (Figure 6). Size distributions of microcapsules coated with MD at 30% concentration ranged from 1 to 10 µm, with an average particle size ($D_{(4,3)}$) of about 5 µm. Fang & Bhandari (2010) have reported that the typical form of encapsulated particles by spray drying is globular with size of 10-100 µm. Baranauskienė et al. (2006) have demonstrated that the particle size of microcapsules coated with 30% WPC varies from 2 to 556 µm. According to Choi et al. (2006) and Ahn et al. (2010), the microcapsules, which have 50-200 µm average size, were enough small to offer soft texture and superb dispersibility in milk. In addition, milk fat globules in homogenized milks have approximately 2-5 µm size, and milk fat globules are stable in milk serum. In view of these facts, the microcapsules which have 5 µm-average size are uniformly distribute in milk or other beverage systems.

3.5 Zeta-potential

Zeta-potential is recognized as an important parameter for the stability of colloidal system (Lin & Chaudhury, 2008). Results of zeta-potential for microemulsions and spray-dried microcapsules were compared (Figure 7). Average zeta-potential

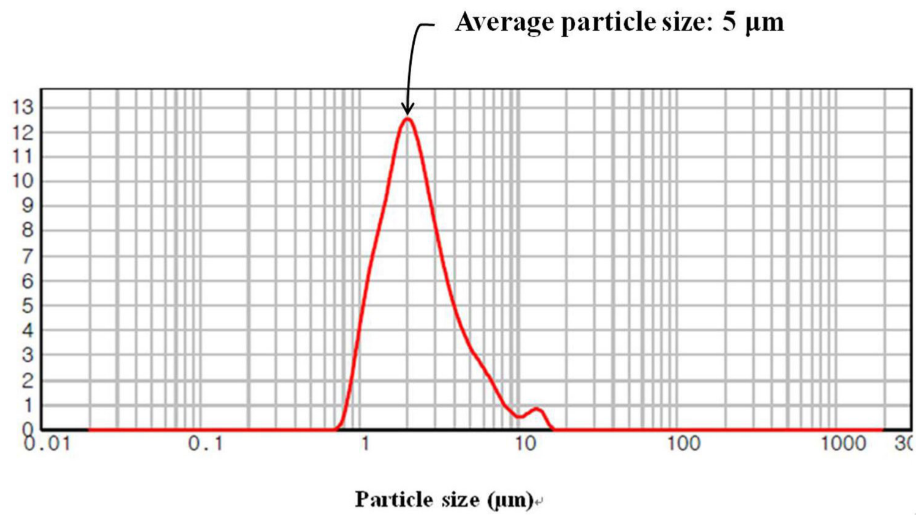

Figure 6. Particle size distribution of spray-dried microcapsules of tomato extracts coated with 30% maltodextrin.

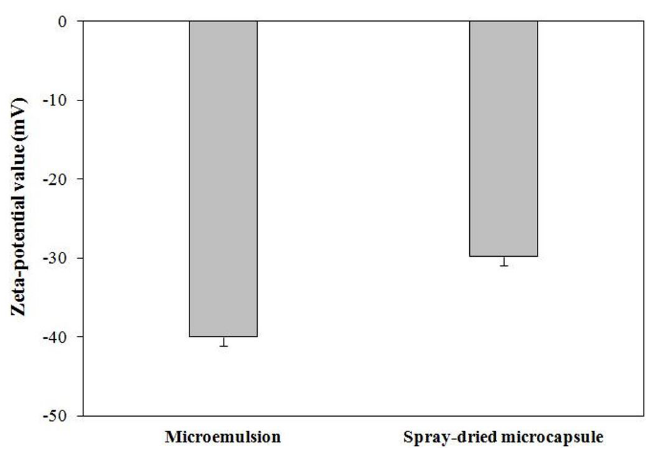

Figure 7. Zeta-potential values of microemulsions and spray-dried microcapsule powders of tomato extracts.

values of microemulsions and the spray-dried microcapsules were approximately -40 mV and -29.68 mV, respectively. The border line between stable and unstable suspensions is generally taken at ±30 mV. Particles with an absolute value of zeta-potential of more than 30 mV are normally considered as stable (Van Nieuwenhuyzen & Szuhaj, 1999). On the other hand, as the absolute values of zeta-potential approached to zero or low-zeta potential value, the dispersion of the particles tended to be electrically unstable and rapidly coagulate or flocculate (Hanaor et al., 2012). Accordingly, both microemulsions and spray-dried microcapsule of tomato extracts can make stable suspension.

4 Conclusion

The microencapsulation technique is known to be good for protect vulnerable core material from light, oxygen, enzymes or other various environment. In the present study, the microemulsification of lycopene was conducted successfully with ratio of tomato to MCT at 1.24:8.76 (v/v), concentration of Tween 60 at 1.0% (w/v), and ratio of W/O to secondary aqueous coating materials at 1:6.16 (v/v), and spray-dried the microemulsion to produce microcapsules. However, the advantages of microencapsulation could not confirm yet in this study. Therefore, it is necessary to perform further study on its release and storage properties and application into various food systems.

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