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Role of Powder Particle Size on the Encapsulation Efficiency of Oils during Spray Drying

Seid Mahdi Jafari,[1] Yinghe He,[2] and Bhesh Bhandari[3]

[1]*Department of Food Science, Gorgan University of Agricultural Sciences and Natural Resources, Gorgan, Iran; School of Land and Food Sciences, University of Queensland, Brisbane, Australia*
[2]*School of Engineering, James Cook University, Townsville, Australia*
[3]*School of Land and Food Sciences, University of Queensland, Brisbane, Australia*

In this study, emulsions prepared by microfluidizer and ultrasound were spray-dried to produce encapsulated powders containing d-limonene or fish oil (20% w/w). Maltodextrin combined with a surface-active biopolymer (modified starch or whey protein concentrate) was used as the wall material (40% solids w/w). It was shown that volatility of the core material significantly affects the surface oil content of the encapsulated powder. Also, by the classification of encapsulated powders into various sizes, our results revealed that larger particles (>63 μm) retain more volatiles than smaller ones (<38 μm), but at the same time there is more unencapsulated oil at the surface of big particles. In the case of fish oil, although surface oil content obtained by solvent extraction was high in larger particles, X-ray photoelectron spectroscopy analysis showed that different particles had similar surface oil coverage.

INTRODUCTION

The main emphasis of encapsulation of food flavors and oils over the last few years has concentrated on improving the encapsulation efficiency during spray drying; that is, preventing volatile losses and extending the shelf life of the products by minimizing the amount of unencapsulated oil at the surface of powder particles.[1–3] This is intended to produce high-quality encapsulated powders with maximum recovery. The properties of wall and core materials as well as the emulsion characteristics and drying parameters are the factors that can affect the efficiency of encapsulation.

Particle size of the encapsulated powder is primarily determined by physical properties of the infeed emulsion (such as viscosity and solids concentration) and the atomization operating parameters, such as the rotational speed and wheel diameter in the case of centrifugal atomization and the orifice size and pressure in the case of nozzle atomization.[4–7] Particle size can also be influenced by the operating temperatures or infeed solids.[1,8] In practice, depending on the spray dryer design, it is possible to control particle size to some extent based on the mentioned parameters. The influence of powder particle size on encapsulation efficiency of food flavors and oils has not been clear. Several workers have reported that larger particle sizes result in improved flavor retention and lower surface oil contents during spray drying.[7,9,10] On the other hand, Reineccius and Coulter[11] and Finney et al.[6] could find no effect of particle size on retention, as they attributed this result to the high concentration of infeed solids. These controversial data can be mainly related to variations in the spray drying design and methods to control particle size or the properties of the initial emulsion. Recently, Soottitantawat et al.[12] showed that powder particle size alone does not have a significant effect on flavor retention, as other parameters such as emulsion size can have a more considerable influence. Furthermore, Zakarian and King[13] have shown that if both volatile loss and rate of drying are diffusion controlled, volatile retention should be independent of particle size.

The work of Silva and Re[14] suggested the existence of an optimal particle size to achieve maximal volatile retention, similar to the results of Chang et al.,[15] who found that the total oil retention was highest for powder with intermediate particles, while it was lowest for powder with largest particles. The other result was that powders with medium and large particle sizes had about 2.5 and 9 times more surface oil than powder with small particle size, respectively. Although the role of particle size is not clear, it is often desirable to produce large particles to facilitate rehydration. Small particles tend to disperse very poorly, especially in cold water, and instead form lumps on the liquid surface. Large particles can be obtained through

Correspondence: B. Bhandari, School of Land, Crop and Food Sciences, University of Queensland, St. Lucia, QLD 4072, Australia; E-mail: b.bhandari@uq.edu.au

appropriate choice of spray dryer operating conditions or the use of agglomeration techniques.[16–19] Some of the encapsulation parameters could also be changed, such as reduction in surface oil content of the powders due to stripping effect of fluidized bed agglomeration.[17]

All of these works are based on particle size of different encapsulated powders, since they have been obtained by changing atomization conditions or emulsion properties such as viscosity. Encapsulation efficiency of different powder particle sizes within the same powder, however, is unknown, and there is no evidence on how the flavor and oil droplets are retained in various sizes of powder particles and whether the distribution of the emulsion is the same in all powder particles produced by spray drying. On the other hand, information about the influence of volatility of the core material on encapsulation efficiency is scant in the literature. Therefore, the objectives of this work are to determine the influence of different core materials (volatile and non-volatile) and various powder particles within the product on encapsulation efficiency and some other powder properties during spray drying.

MATERIALS AND METHODS

Materials

In this study, d-limonene (Orange Terpen 07231 TT, Quest International, NSW, Australia) or fish oil (HiDHA 25N, Nu-Mega Ingredients, Brisbane, Australia) was used as the core material. The wall material was an aqueous solution of a modified starch (Hi-Cap 100, National Starch and Chemical, NSW, Australia) and/or whey protein concentrate (WPC: 73% protein, 9% fat, 4% moisture, 4% ash; ALACEN, New Zealand Milk Products, Auckland, New Zealand) in a combination with maltodextrin (DE 16–20, Fieldose 17-C AP, Penford Limited, NSW, Australia). Analytical-grade hexane and petroleum ether (BP 40–60°C) were purchased from Sigma Chemicals Company (Sydney, Australia). Distilled water was used for the preparation of all solutions. All general chemicals used in this study were of analytical grade.

Preparation of Emulsions

Wall material powders were dissolved in distilled water one day before emulsification and kept overnight in a shaking water bath (Ratek Instruments, Melbourne, VIC, Australia) to warrant a full saturation of the polymer molecules. Total concentration of dissolved solid was 40% (w/w) composed of 30 wt% maltodextrin and 10 wt% emulsifying ingredients (Hi-Cap or WPC). All emulsions (oil-in-water) were prepared in two stages: (a) pre-emulsions were obtained by a rotor-stator system (Model L2R, Silverson Machines Ltd, London, UK), and the core material (fish oil or d-limonene) in the ratio of 1:4 (core:-wall) was progressively added to the continuous phase

during pre-emulsion preparation and stirred for 10 min at the highest speed. (b) These coarse emulsions were then further emulsified using a microfluidizer (Model M-110 L, Microfluidics, Newton, MA) at 60 MPa for one cycle or a 24-KHz ultrasound probe (Dr Hielscher series, Model UP 400S, Stuttgart, Germany) with 22 mm diameter at the highest power for 100 s. More details about emulsification conditions are presented in our previous works.[20,21]

Spray Drying

A pilot-plant spray dryer (Model SL 20, Saurin Group of Companies, Victoria, Australia) was used to convert infeed emulsions into encapsulated powders, as described in our earlier studies.[21,22] The operational conditions of the spray drying were air inlet temperature of 180°C, air outlet temperature of 65°C, and air pressure at the nozzle of 310 kPa. The dried powder was collected and stored in opaque, airtight containers at 4°C awaiting further analysis.

Size Classification of Encapsulated Powders

In order to classify the encapsulated powders into different classes based on particle size, an analytical sieve shaker (Model AS200, Retsch, Haan, Germany) was used. This machine had a vibrating platform with sieves assembled on the top section. Sieves were perforated plates made of stainless steel and available in different sizes. We used the smallest size (38 μm) and a 63 μm sieve according to the preliminary experiments carried out on the encapsulated powders. These two sieves (with the smaller size underneath) were placed on a container with the same structure as sieves and assembled on the platform of the machine. After adding the powder onto the top sieve (63 μm) and by vibration (for approximately 15 min), some of the particles could pass through this sieve and reach the lower sieve. The particles that were smallest could also pass the lower sieve and fall into the collecting container. The operation was continued until no obvious separation could occur. Therefore, particles remaining on the top sieve were the largest, and those that collected in the container were the smallest size. The fraction that remained between two sieves were of intermediate size.

Emulsion Droplet Size and Powder Particle Size Analysis

The size distribution of the oil droplets in infeed emulsions and the reconstituted emulsions was determined by the laser light scattering method using Mastersizer 2000 (Malvern Instruments, Worcestershire, UK), as described in our previous works.[20,23] Redispersion properties were studied by producing 10% (w/w) aqueous solutions of the powders via mixing the reconstituted emulsions at room temperature with a magnetic stirrer for 30 min. The analysis of powder particle size was performed using the laser light scattering method by an analyzer with a batch cell unit (Mastersizer E, Malvern Instruments). Encapsulated powders were dispersed in propan-2-ol for the particle size

analysis. In all cases, volume mean diameter, or d_{43}, was reported as the emulsion size or powder particle size.

Encapsulation Efficiency Analysis

By definition, encapsulation efficiency (EE) is the amount of core material encapsulated inside the powder particles.[22] For d-limonene–encapsulated powders, we used surface oil content and retention data as indications of the encapsulation efficiency, while for fish oil–encapsulated powders, surface oil content or surface oil coverage were selected criteria for encapsulation efficiency.

Total Oil Measurement

"Clevenger" distillation method was used to determine total oil for d-limonene–encapsulated powders, as described in our previous work.[22] A 500-mL round-bottomed flask was used to dissolve 10 g ($\pm$0.02 g) of the spray-dried powder in 250 mL of deionized water. The solution inside the flask came to boiling underneath the heater and distillation was continued for 3 h. The volume of the oil (d-limonene) was converted to the oil mass via multiplying by the oil density (0.856 g/mL at 20°C), which was measured with a hydrometer. For fish oil–encapsulated powders, no total oil analysis was performed assuming that all the initial oil was retained in the powder because of being nonvolatile.

Surface Oil Content Measurements

The amount of extractable oil on the surface of d-limonene–encapsulated powders was determined according to the method explained in our previous study.[22] One gram of sample, 9 mL of hexane, and 1 mL of a stock solution of cyclohexane in hexane (about 10 µg/mL) as an internal standard were added into a glass bottle. After extraction, the oil content in the organic phase was measured by gas chromatography (GCMS model GC-17A V3, Shimadzu Corp., Kyoto, Japan). The chromatographic conditions were flame ionization detector (FID) at 275°C with helium as the carrier gas; column head pressure 45 kPa; split injection; initial temperature 60°C with initial time of 2 min; program rate 8°C/min to 100°C; and 10°C/min to 200°C was controlled at constant value of 120°C.

The surface oil content of fish oil–encapsulated powders was determined using extraction with petroleum ether (BP 40–60°C), according to the method of Garcia et al.[24] described in our previous study.[21] After the required extraction time, the powder and the solvent were separated by filtration and the powder residue was washed with solvent and the filtrate solution containing the extracted oil was transferred to a round-bottomed flask, which was placed in a rotary evaporator (Model R-114, Buchi Co., Flawil, Switzerland). After evaporation, the flask was dried in an oven (IM550, Clayson Inc., NSW, Australia) at 103°C until constant weight (about 1 h) and the extracted oil value was then calculated based on the difference between the weight of the initial clean flask and that containing extracted oil residue.

Surface Oil Coverage Analysis

We analyzed the surface composition of fish oil–encapsulated powders by X-ray photoelectron spectroscopy (XPS), as explained in our earlier work.[21] The underlying principle of the technique and its application to food powders, particularly dairy powders, have been described elsewhere.[25–30] The XPS analysis was carried out with a Kratos Axis Ultra (Kratos Analytical, Manchester, UK) spectrometer incorporating a 165-mm hemispherical electron energy analyzer. The incident radiation was monochromatic Al X-rays (1486.6 eV) at 150 W (15 kV, 15 ma). Survey (wide) scans were taken at an analyzer pass energy of 160 eV and multiplex (narrow) high-resolution scans at 20 eV. Survey scans were carried out over 1200–0eV binding energy range with 1.0 eV steps and a dwell time of 100 ms. Narrow high-resolution scans were run with 0.05 ev steps and 250 ms dwell time. Base pressure in the analysis chamber was 1.0×10^{-7} Pa and during sample analysis 1.3×10^{-6} Pa. It should be mentioned that for fish oil, the pre-operating temperature of the machine was taken down to transform the liquid oil into a frozen solid, and then surface analysis was performed.

Scanning Electron Microscopy of Encapsulated Powders

A JSM 6400F model scanning electron microscope (JEOL Co. Ltd., Tokyo, Japan) was used to investigate the microstructural properties of the spray-dried encapsulated powders. The samples were placed on the SEM stubs using a two-sided adhesive tape (Nisshin EM Co. Ltd., Tokyo, Japan). The specimens were subsequently coated with Pt using a magnetron sputter coater (Model EIKO IB-5, Eiko Inc., Tokyo, Japan). The coated samples were then analyzed using the SEM operating at an accelerating voltage of 15 kV. The micrographs representing the microstructure of the encapsulated powders were taken by the instrument's software installed on a PC connected to the system.

Experimental Parameters and Statistical Analysis

The parameters considered affecting the encapsulation efficiency were the emulsification method, the type of surface-active ingredient, powder particle size within the same encapsulated powder, and the type of core material. The effect of each operational and compositional parameter was studied using only one system (core or wall material). All the experiments were performed based on a fully factorial design and the results represent the means of two replicates. A general linear model of MINITAB (Version 14, 2004) was used to conduct an analysis of variance (ANOVA) to determine differences between treatments means. Treatment

means were considered significantly different at $P \leq 0.05$ and the difference was considered very significant at $P < 0.01$. Some of the graphs were drawn by Excel (Microsoft Office 2003) and some by MINITAB 14.

RESULTS AND DISCUSSION

Influence of the Oil Type on Encapsulation Efficiency

In order to compare the influence of volatility of the core material on encapsulation efficiency, particularly the surface oil content during the spray-drying process, a full randomized factorial design was planned with two different core materials: a volatile oil (d-limonene) and a nonvolatile oil (fish oil) at a concentration of 20 wt% of total solids. Three methods of emulsification including rotor-stator system (Silverson), microfluidization, and ultrasonication were applied for the preparation of the infeed emulsion and similar spray-drying conditions were used. The overall result was that oil type had a very significant influence ($P < 0.01$) on the encapsulation efficiency and other measured data such as emulsion size before spray drying and after reconstituting the encapsulated powder in water (Table 1).

The surface-active biopolymer only had a very significant ($P < 0.01$) influence on emulsion size and retention of d-limonene or surface oil coverage of fish oil samples, and it did not have any significant effect ($P > 0.05$) on surface oil content and particle size of encapsulated powders. Also, fish oil samples had a much higher surface oil content (5–6 times higher) than their d-limonene counterparts, which could be justified by the volatility of d-limonene, as it will be evaporated and lost from the surface of powder particles during spray drying, while fish oil remains at the

surface more predominantly. For instance, the surface oil content of d-limonene–encapsulated powders containing Hi-Cap and produced from ultrasound emulsions was about 103.1 mg/100 g powder, while for fish oil–encapsulated powders, the surface oil content was about 540 mg/100 g powder. The influence of emulsification method and emulsifying agent on encapsulation efficiency of spray-dried powders containing fish oil and/or d-limonene was discussed in our previous studies.[21,22]

Efficiency of Powder Size Classification

The classification of encapsulated powder based on different particle sizes was done through vibrating screens (sieves) with various pore sizes. Accordingly, encapsulated powders were classified into three classes, $< 38 \, \mu m$, 38–$63 \, \mu m$, $> 63 \, \mu m$, and all the analyses were performed on these powders. Classification with vibrating screens was very efficient, as it significantly ($P < 0.05$) affected the powder particle size analyzed through laser scattering technique (Mastersizer E). For example, powder passed through the smallest screen size ($< 38 \, \mu m$) for microfluidized d-limonene samples containing Hi-Cap had a particle size (d_{43}) of about $19.89 \, \mu m$, while those remaining at the top of the largest screen size ($> 63 \, \mu m$) had a d_{43} equal to $68.12 \, \mu m$. The d_{43} of intermediate powder particles was approximately $32.30 \, \mu m$. This is shown in Fig. 1 with complete particle size distribution of the mentioned samples.

More interestingly, powder particle sizes of bulk samples (without size classification) were comparable to intermediate particles, which was reasonable since bulk sample is a mixed powder containing all different particle sizes. The difference in analyzed size data and pore size of screens

TABLE 1
Influence of the oil type (volatile vs. nonvolatile) on surface oil content and other properties of the encapsulated powders

Core material	Surface active biopolymer	Emulsification method	Emulsion size (d_{43}, μm)	Powder size (d_{43}, μm)	Reconstituted emulsion size (d_{43}, μm)	Surface oil (mg/100 g powder)	Surface oil (mg/m² powder)	Retention (%)
d-Limonene	Hi-Cap	Silverson	1.3^a	27.8^a	1.2^a	55.6^a	7.3^a	82.7^a
		Microfluidizer	0.74^b	37.3^b	0.99^b	55.4^a	9.0^b	86.2^a
		Ultrasound	0.75^b	30.9^a	1.2^a	103.1^b	8.5^b	85.8^a
	WPC	Silverson	3.1^a	31.9^a	4.7^a	899.4^a	158.3^a	65.9^a
		Microfluidizer	0.99^b	21.3^b	0.86^b	170.1^b	18.5^b	76.3^b
		Ultrasound	1.8^c	21.3^b	1.8^c	90.5^c	10.4^c	83.9^b
Fish oil	Hi-Cap	Silverson	4.6^a	41.2^a	48.5^a	2040^a	355.4^a	—*
		Microfluidizer	0.21^b	25.7^b	0.26^b	170^b	22.2^b	—
		Ultrasound	2.2^c	28.8^b	4.1^c	540^c	74.5^c	—
	WPC	Silverson	5.9^a	36.0^a	6.8^a	1270^a	174.8^a	—
		Microfluidizer	0.28^b	32.1^a	0.88^b	690^b	98.3^b	—
		Ultrasound	3.5^c	34.0^a	2.5^c	770^b	103.6^b	—

Means within the same column (for individual biopolymers) followed by different letters[a,b,c] are significantly ($P < 0.05$) different.
*The retention of fish oil is assumed to be 100% during spray drying because of its nonvolatile nature.
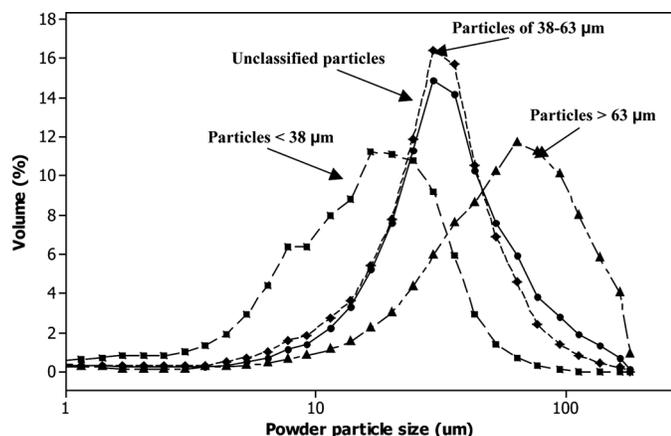
FIG. 1. Powder size distribution (analyzed by laser scattering) of classified d-limonene (20 wt%) encapsulated powders containing Hi-Cap and obtained from microfluidized emulsions compared to the bulk unclassified sample.

$$D43 = \frac{\sum n_i d_i^4}{\sum n_i d_i^3}$$

Our final goal, however, was to classify the powder into different groups according to particle size, which was successful by the mentioned classifying system. The SEM micrographs of the bulk samples of d-limonene–encapsulated powders and the classified ones are shown in Fig. 2. It can be seen that intermediate and small class powders mostly contain smaller particles, while the larger class one contained a minimum number of smaller particles.

d-Limonene Classified Powders

In general, there were significant differences ($P < 0.05$) among various particles within the same encapsulated powder in terms of surface oil content and retention for d-limonene samples, as can be seen from Table 2. Our results showed that surface oil content is progressively increasing when the particle size within the powder is becoming larger, particularly if surface oil data being presented are based on the specific surface area of the powder particles (Fig. 3). For instance, results for powders consisting of Hi-Cap and obtained from Silverson emulsions revealed that small

could be related to various size representations. In other words, pore size of the screens is based on simple circular diameter, while particle size obtained with laser scattering analysis is based on volume mean diameter (d_{43}), which is calculated from the following equation:
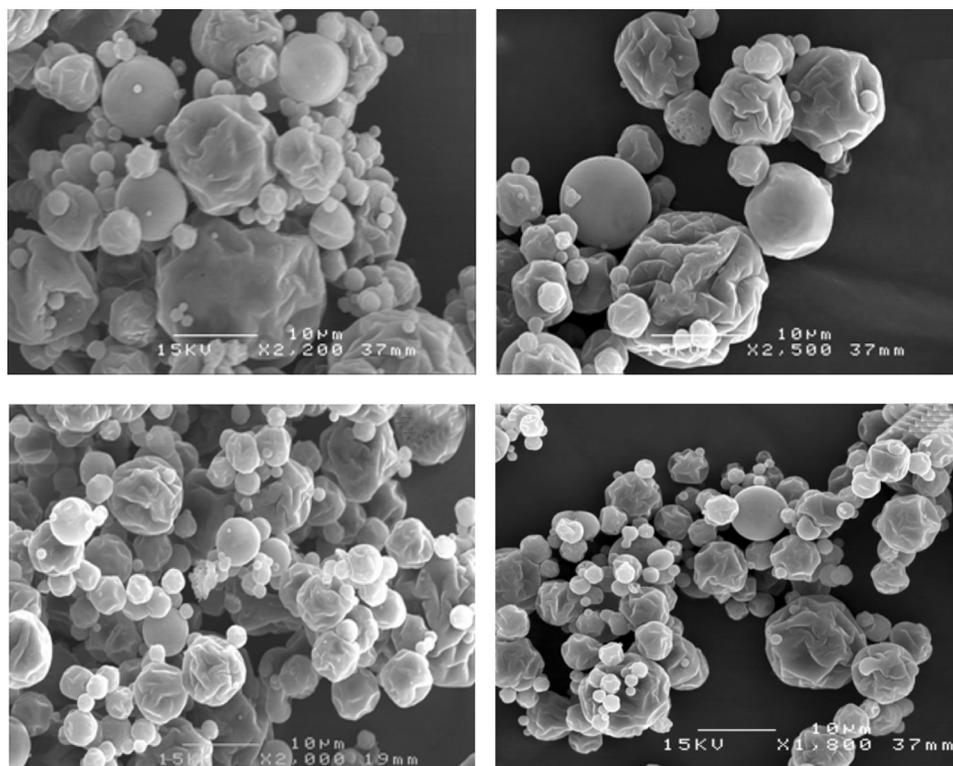

FIG. 2. SEM micrographs of d-limonene (20 wt%) encapsulated powders containing Hi-Cap and produced from microfluidized emulsions: (A) unclassified bulk sample; (B) powder with the largest particles ($>63\,\mu m$); (C) powder with intermediate size (38–63 μm); (D) powder with the smallest particles ($<38\,\mu m$).

TABLE 2

Influence of powder particle size within the same powder on surface oil content and retention and some other properties of d-limonene–encapsulated powder

Powder class[*]	Emulsion size (d_{43}, μm)	Powder size (d_{43}, μm)	Reconstituted emulsion size (d_{43}, μm)	Surface oil (mg/100 g powder)	Surface oil (mg/m^2 powder)	Retention (%)
HLS 1	1.3	15.47[a**]	1.26	24.45	1.39[a]	74.04[a]
HLS 2	1.3	28.38[b]	1.21	53.29	7.60[b]	75.28[a]
HLS 3	1.3	32.40[c]	1.19	77.25	8.58[b]	82.47[b]
HLU 1	0.75	17.64[a]	0.99	93.26	5.67[a]	76.39[a]
HLU 2	0.75	29.37[b]	1.02	73.74	5.93[a]	83.74[b]
HLU 3	0.75	34.44[c]	1.04	91.19	14.36[b]	90.27[c]
HLM 1	0.74	19.89[a]	0.99	49.85	5.48[a]	76.62[a]
HLM 2	0.74	32.30[b]	1.01	49.04	6.81[a]	85.01[b]
HLM 3	0.74	68.12[c]	0.85	83.5	19.92[b]	92.86[c]
WLS 1	3.1	13.59[a]	2.95	336.42	30.88[a]	62.46[a]
WLS 2	3.1	21.96[b]	4.22	337.31	44.14[b]	67.57[a]
WLS 3	3.1	44.63[c]	4.05	279.08	57.39[c]	75.60[b]
WLU 1	1.8	13.08[a]	1.77	68.92	6.04[a]	68.13[a]
WLU 2	1.8	22.58[b]	1.78	86.77	10.58[b]	74.55[a]
WLU 3	1.8	34.34[c]	1.79	109.11	15.02[c]	85.94[b]
WLM 1	0.99	13.80[a]	0.81	56.75	4.79[a]	64.49[a]
WLM 2	0.99	24.31[b]	0.81	61.71	7.85[b]	66.17[a]
WLM 3	0.99	30.99[c]	0.82	71.57	10.20[c]	81.11[b]

[*]The codes are H: Hi-Cap, L: d-limonene, S: Silverson, U: ultrasound, M: microfluidizer, W: whey protein concentrate. Code 1 is classified powder <38 μm, 2 is the powder of 38–63 μm, and 3 is >63 μm according to the sieve pore size.

[**]Means within the same column (for each emulsifying device) followed by different letters[a,b,c] are significantly different ($P < 0.05$).

particles (<38 μm) had a surface oil content of 1.39 mg/m^2 powder (24.45 mg/100 g powder). While the same particles with a larger size (38–63 μm) had a surface oil content of 7.60 mg/m^2 powder (53.29 mg/100 g) and this figure was increased to 8.58 mg/m^2 powder (77.25 mg/100 g) for the largest particles (>63 μm).

This could be explained by the fact that while larger particles have a reduced surface area to volume retention, which would result in better oil retention (Table 2), there would be also a longer time for film formation around the large droplets during the process. The longer the time necessary for film formation, the greater the loss of core material and the higher the surface oil content. Therefore, these two competing parameters can determine the overall effect of particle size on encapsulation efficiency. Also, as can be seen in Fig. 2, large particles generally have more shrinkage than smaller ones, which results in higher surface area and, therefore, increased amounts of surface oil contents. Surprisingly, analyzed data for the bulk sample (without size classification) were almost comparable to middle-sized powders, particularly for those samples containing Hi-Cap, which is reasonable because it is a mixture of particles with different sizes.

Another explanation could be a rapid drying rate of small atomized droplets that would result in a quick formation of

the semi-permeable membrane around droplets and therefore less leaching of the core materials inside the droplets. In fact, smaller droplets undergo higher mass and heat transfers during atomization inside the spray-drying chamber, which means final dried particles will be shaped and formed faster compared with larger atomized droplets. Also, the surface morphology changes (dents, wrinkles) are fewer with smaller particles, so they will contain lower surface oil content as confirmed by our results. There was no significant difference between emulsion droplet sizes retained in different size class of particles, as reconstitution of classified encapsulated powders revealed that almost all samples lead to same emulsion size (Table 2) and size distribution.

It is expected that smaller particles also retain more volatiles than larger particles, as they will have higher drying rates and loss of volatiles could be limited due to the fast formation of a semi-permeable membrane. The results obtained by the analysis of total retention of d-limonene–encapsulated powder contradicted this trend (Fig. 4), as smaller particles within the same powder had significantly ($P < 0.05$) lower retention than bigger particles. For instance, retention of d-limonene in small particles of ultrasound samples consisting of Hi-Cap was about 76.4%, which increased to 83.7 and 90.3% for intermediate and
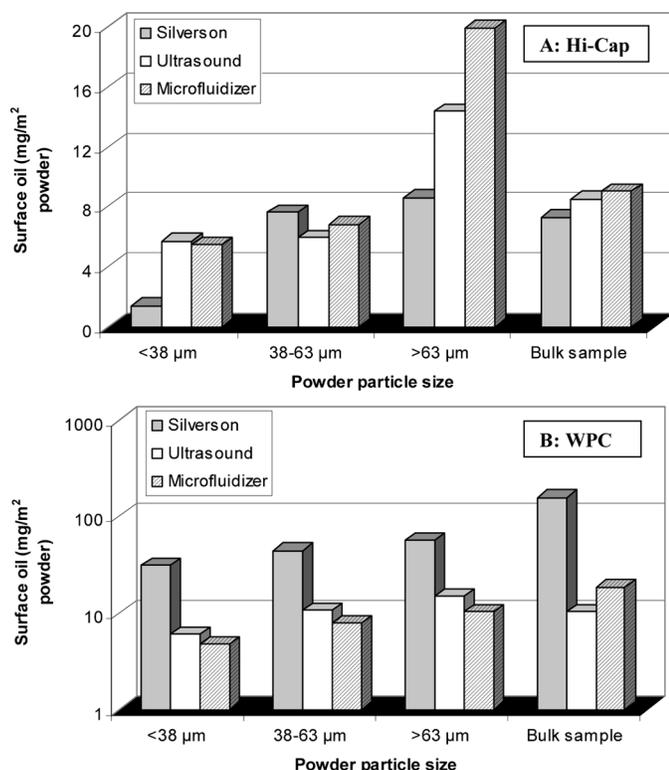
FIG. 3. Surface oil content of classified d-limonene (20 wt%) encapsulated powders containing Hi-Cap (A) or WPC (B) as the surface-active biopolymer and produced from three different emulsifying devices.

larger particles, respectively. The reason could be the smaller surface area and higher volume of larger particles, which helps to retain more core materials during the process. In fact, if the loss of volatiles happens per unit surface area of the powder particles, the amount of loss would be lower for larger particles due to lower surface area
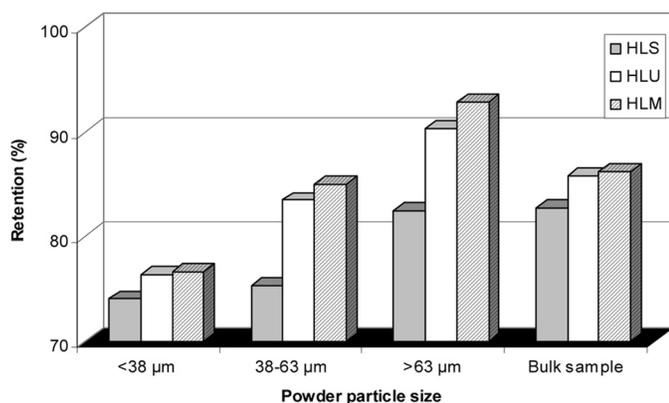

FIG. 4. Retention of classified d-limonene–encapsulated powders containing Hi-Cap compared with the bulk samples (without classification) for emulsions made with Silverson (HLS), ultrasound (HLU), and microfluidizer (HLM).

compared with smaller particles. Also, since the emulsion size is fixed, the larger the powder particle, the higher the difference between emulsion size and the higher capability of particles to enclose more oil droplets and, thus, higher retention. For smaller particles, on the other hand, there is a more chance for some of the oil droplets to be encapsulated incompletely and be lost during the spray-drying process, as the core material (d-limonene) is a volatile compound.

Fish Oil–Classified Powders

In the case of fish oil samples, we found similar results to d-limonene–classified powders; i.e., surface oil content of individual particles within the encapsulated powder increased with increasing particle size (Fig. 5). The important result was that surface oil coverage of classified powders analyzed through XPS showed no difference among various particle sizes (Table 3). For instance, the surface oil coverage of small, intermediate, and large particles from microfluidized fish oil emulsions containing Hi-Cap were equal to 26.53, 26.24, and 25.92%, respectively. This can be attributed to the topography of individual encapsulated particles. Since the XPS system analyzes only the top 10-nm layer of the samples, it cannot show any difference among different size particles, as bigger particles have more shrinkage and, possibly, the surface oil is concentrated more at the dents and indentations rather than on smooth surfaces. Therefore, elemental composition of different particles would be the same, as confirmed by XPS results (Table 3).

CONCLUSION

Comparing volatiles and nonvolatiles, we presented data confirming that surface oil content of nonvolatile (fish oil) encapsulated powders was much higher than their volatile (d-limonene) counterparts, since volatile compounds could be evaporated and removed during spray drying. Also, it
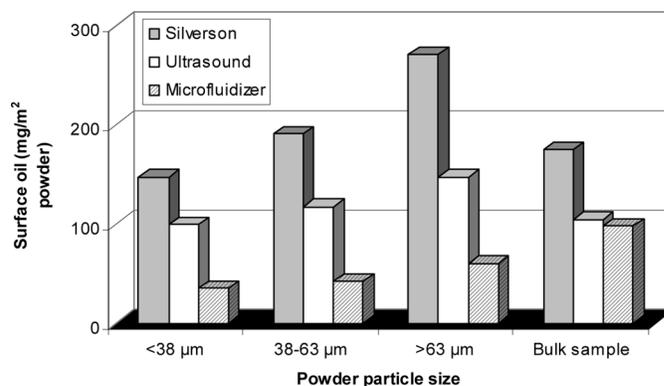

FIG. 5. Surface oil content of classified fish oil (20 wt%) encapsulated powders containing WPC as the surface-active biopolymer and produced from three different emulsifying devices.

TABLE 3

Influence of powder particle size within the same powder on surface oil content and coverage and some other properties of fish oil–encapsulated powder

Powder class[*]	Emulsion size (d_{43}, μm)	Powder size (d_{43}, μm)	Reconstituted emulsion size (d_{43}, μm)	Surface oil (mg/100 g powder)	Surface oil (mg/m^2 powder)	Surface oil coverage (%)
HFS 1	4.6	36.69[a**]	49.18	2380	582.9[a]	63.79
HFS 2	4.6	44.23[b]	50.52	2740	815.9[b]	63.40
HFS 3	4.6	52.72[c]	53.05	3200	1070.9[c]	61.91
HFU 1	2.2	26.07[a]	4.10	370	35.6[a]	25.78
HFU 2	2.2	29.54[a]	4.05	520	57.1[b]	25.14
HFU 3	2.2	34.74[b]	4.13	680	97.5[c]	26.21
HFM 1	0.21	17.23[a]	0.31	220	19.7[a]	26.53
HFM 2	0.21	20.39[a]	0.37	240	23.7[a,b]	26.24
HFM 3	0.21	28.74[b]	0.30	170	25.3[b]	25.92
WFS 1	5.9	26.87[a]	6.78	1230	147.4[a]	33.62
WFS 2	5.9	33.42[b]	6.79	1360	190.9[b]	36.39
WFS 3	5.9	39.89[c]	6.83	1540	271.3[c]	35.41
WFU 1	3.5	23.72[a]	2.60	840	98.8[a]	25.19
WFU 2	3.5	27.17[b]	2.66	900	116.9[b]	27.69
WFU 3	3.5	35.91[c]	2.48	820	147.5[c]	18.14
WFM 1	0.28	20.40[a]	0.88	400	34.7[a]	17.22
WFM 2	0.28	22.44[a]	0.89	420	41.8[b]	16.76
WFM 3	0.28	28.48[b]	0.88	450	59.9[c]	17.69

[*]The codes are H: Hi-Cap, F: Fish oil, S: Silverson, U: ultrasound, M: microfluidizer, W: whey protein concentrate. Code 1 is classified powder < 38 μm, 2 is the powder of 38–63 μm, and 3 is > 63 μm according to the sieve pore size.

[**]Means within the same column (for each emulsifying device) followed by different letters[a,b,c] are significantly different ($P < 0.05$).

was explained that encapsulated powders can be classified according to their particle size using a vibrating system equipped with different sieves. Our results revealed that different powder particles within the same encapsulated product were not similar considering encapsulation efficiency. Smaller particles (< 38 μm) had lower surface oil contents but with poorer retention of d-limonene compared with bigger particles (> 63 μm). Another result was that no difference was found among fish oil–classified powders in terms of surface oil coverage. Finally, it was shown that emulsion droplets encapsulated into different particles generally have the same size and size distribution, independent of the powder particle size. To this end, encapsulation efficiency can be improved by having a narrowly distributed powder particles with optimum size, which was a smaller particle for nonvolatiles (fish oil) and a medium-sized particle for volatiles (d-limonene).

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