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Research Article

Effect of Feed Material Properties on Microencapsulation by Spray Drying with a Three-Fluid Nozzle: Soybean Oil Encapsulated in Maltodextrin and Sugar Beet Pectin

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The relationship between the properties of wall material and spray-dried microcapsules was investigated. Soybean oil was encapsulated with various concentrations of maltodextrin (10 and 20% *w*/*w*) and sugar beet pectin (0.5, 1, 2, 3, and 4% *w*/*w*) solution via spray drying with a three-fluid nozzle (3FN). The rheological properties of the wall material solution were characterized using a controlled strain rheometer before spray drying and were found to fit the power-law model. The rheological properties of a wall material within the range of the samples tested had a limited impact on the morphology of the microcapsules. Most spray-dried samples had a wrinkled surface with some microcapsules having a spherical shape with a hollow core. As the consistency index of the wall material solution increased, the particle size distribution of microcapsules became wider. The surface oil content of the microcapsules was between 1.3 and 3.4%, generally increasing as the consistency index of the wall material solution increased. The encapsulation efficiency (EE) was between 74.7 and 91.2%. When compared with the sample produced with a two-fluid nozzle (2FN), 3FN microcapsules were larger and had higher effective EE and higher oil loading than 2FN microcapsules. This study revealed that oil could be encapsulated by wall materials, without the preparation of an emulsion, with a 3FN, which will save time and energy for the encapsulation process.

1. Introduction

Encapsulation is an effective method to entrap core compounds within a wall material [1], which can control delivery, mask off flavor, and improve the stability of core compounds [2]. Spray drying is one of the most widely used techniques to produce microcapsules. The process includes the atomization of a feed liquid and dehydration of the atomized droplets in a stream of hot air, typically 150-220°C, to form a powder in 15-30 seconds [3, 4].

A spray nozzle is an essential part of a spray dryer to atomize liquid feed. Two-fluid spray nozzles (2FN) are one of the conventionally used atomization nozzles, which contain two concentric channels for compressed gas and a premixed homogeneous liquid (Figure 1(a)). Three-fluid nozzles (3FN) have a channel for compressed gas and two liquid channels separating the flows of, for example, core material and wall material until they make contact at the nozzle tip (Figure 1(b)), thus creating an opportunity for the creation of an emulsion during spray drying [5, 6]. This design creates an opportunity to decrease production time and cost by eliminating a separate emulsification step in a spray drying process, which is a unique opportunity for this 3FN technology not available for other nozzle/atomizer types, such as 2FN, high pressure nozzle, and rotary disks.

The properties of spray-dried microcapsules are controlled by the material properties of liquid feed and various processing parameters, such as feed viscosity, feed rate, feed

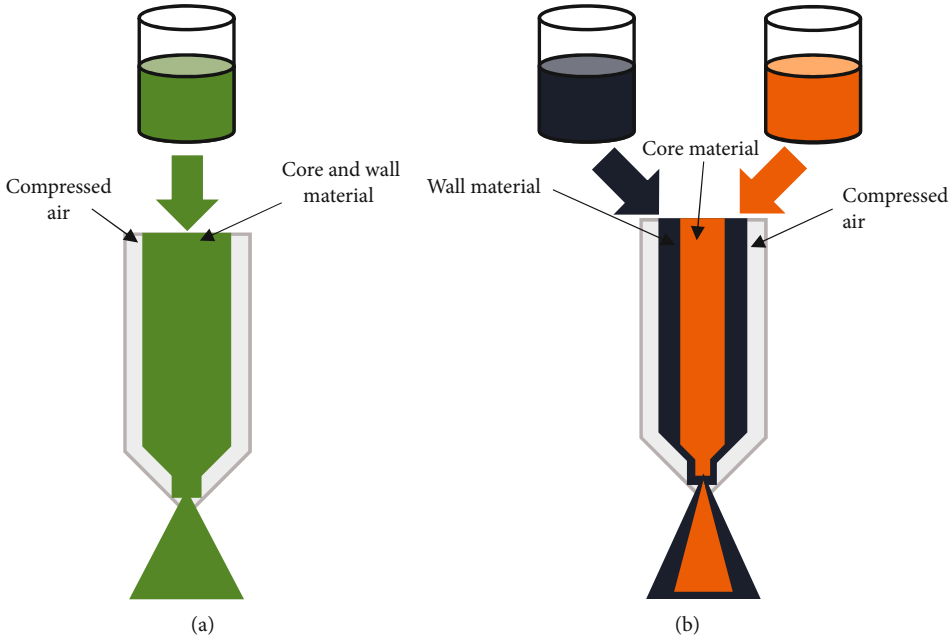

Figure 1: Schematic diagram of two types of nozzles for a spray dryer: (a) two-fluid nozzle (2FN); (b) three-fluid nozzle (3FN).

temperature, inlet air temperature, and atomization pressure [7]. Many encapsulation studies using spray drying have been completed with a 2FN [8–10].

Recently, many other researchers have been exploring novel areas in microencapsulation of food materials as well, such as Shamaei et al. [11]. In their study, the effect of total solid concentration, dry air temperate, and oil/wall material mass fraction on the morphology of an individual droplet of skim milk powder encapsulated walnut oil. And Shamaei et al. [12] investigated the physiochemical effect of wall material composition of walnut oil microcapsules in a 2FN. Also, the use of Pickering emulsions and chitosan was investigated for the production of submicron-sized spray-dried particles [13].

As another novel tool in microencapsulation, there has been limited information reported using 3FNs for spray drying to create microcapsules for food applications. Kašpar et al. [14] used a 3FN to microencapsulate tripolyphosphate with chitosan. Legako and Dunford [6] encapsulated fish oil with whey protein isolate using a 3FN. A 3FN was also used to encapsulate α-amylase [15]. Cáceres et al. [16] reported an encapsulation of purified walnut oil with Capsul® (encapsulating agent), sodium alginate, and ascorbic acid using a 3FN. In Table 1, recent studies investigating the use of 3FN technology in microencapsulation have been listed. These further illustrate the novelty of our present study as there are very limited studies on the application of 3FN for oil microencapsulation.

In our previous study, the controlled release of encapsulated sodium was investigated using maltodextrin and octenyl-succinic-anhydride-modified starch by spray drying with 2FN and 3FN [5]. 3FNs have been also used in the pharmaceutical and chemical industries [7, 22–24]. Overall, few studies have explored the effect of material properties or processing conditions on microencapsulation performance and the properties of spray-dried microcapsules using a 3FN [24].

The design of 3FN provides the possibility for two immiscible liquids to be spray dried without premixing, especially when the mixing happens in the nozzle body. To better understand the physical mechanisms that occur in a 3FN during spray drying, the flow-focusing configuration of a microfluidic device can be used as a reference. Due to the difference in flow rates, the fluid with a higher flow rate can be considered a continuous phase and the one with a lower flow rate can be a dispersed phase. Similar to flow-focusing devices, two immiscible fluids are hydrodynamically focused and thereafter in elongating flows when passing through a contraction [25].

When two separate fluids of wall and core materials are fed into the 3FN with controlled flow rates, the flows can transform into dripping in the nozzle, which is the basic mechanism of encapsulation in a 3FN. In this study, hydrophobic core material and hydrophilic wall materials were chosen as a model to explore the application of the 3FN for encapsulation. Soybean oil was selected as the model core material due to its unsaturated nature and ease of accessibility. It is classified as polyunsaturated oil and an important source of vitamin E which has many health benefits including lowering serum cholesterol and low-density lipoprotein (LDL) levels and preventing atherosclerosis and heart disease. Polyunsaturated oil is unstable when exposed to oxygen, light, moisture, and heat, so encapsulation of soybean oil is a possible way to maintain its biological and functional properties during storage and consumption [26].

For the wall material, typically a wide variety of natural or synthetic polymers have been used based on the characteristics desired in the final microcapsules. Particularly, for oil encapsulation, an ideal wall material should have good emulsifying properties, be a good film former, have a bland taste, and have low hygroscopicity as well as have good rheological properties and solubility [27]. Sugar beet pectin is a

Table 1: Recent studies using the three-fluid nozzle technology for microencapsulation.

Study title	Particle type	Year	Ref.
Application of a three-fluid nozzle for the preparation of amorphous solid dispersions	Amorphous solid dispersions	2022	Corell et al. [17]
Development of iron-vitamin multilayer encapsulates using 3 fluid nozzle spray drying	Multilayer particles for targeted nutrient delivery	2023	Nimbkar et al. [18]
A general strategy for efficiently constructing multifunctional cluster fillers using a three-fluid nozzle spray drying technique for dental restoration	Complex nanoparticles for dental resin composites	2022	Yang et al. [19]
A modified 3-fluid nozzle spray drying approach for co-encapsulation of iron and folic acid	Coencapsulated nutrient fortification	2023	Nimbkar et al. [20]
Development of inhalable spray dried nitrofurantoin formulations for the treatment of emphysema	Inhalable nanoparticles for medical applications	2022	Leslie et al. [21]

polysaccharide extracted from sugar beet pulp and works as a suitable wall material for producing spray-dried oil powders due to its excellent emulsifying properties [28]. It has been used to encapsulate fish oil [8], and the encapsulation efficiency can reach higher than 90% [28].

The emulsifying properties of sugar beet pectin are attributed to its high protein content (up to ~10%) and high level of acetylation (up to 5%) compared to other pectins [29, 30]. Due to the limited solubility of sugar beet pectin in water, a bulk agent is required to increase the solid concentration of the wall material solution. Maltodextrin is commonly used as a wall material for microencapsulation. It has a high solubility in water and low viscosity [31]. By adjusting the concentration of sugar beet pectin and maltodextrin, the rheological and chemical properties of the wall material will be altered which may affect the microencapsulation performance.

Therefore, the current study is aimed at determining the impact of the formulation and rheological properties of the wall material solution (sugar beet pectin and maltodextrin) on the microencapsulation of soybean oil by spray drying with a 3FN. The wall material solutions were characterized for rheological properties, while the microcapsules were evaluated for morphology, internal structure, particle size distribution, flowability, color, surface oil, and encapsulation efficiency. It was hypothesized that increasing the viscosity and pectin content of wall materials will increase the particle size of microcapsules and encapsulation efficiency due to the greater surface-active properties of pectin and larger and denser droplets formed at the nozzle tip when spray dried with a 3FN. Any abbreviations, acronyms, or symbols encountered in this paper can be found with their definition in Table 2.

2. Material and Methods

2.1. Material. Soybean oil (Carlini Pure Vegetable Oil) was purchased from a local grocery store (ALDI Inc., Batavia, IL, U.S.A). Sugar beet pectin (GENU® BETA Pectin, high methyl pectin, dextrose equivalent- (DE-) 55) was provided by CP Kelco Company (CP Kelco, Atlanta, GA, U.S.A.). Maltodextrin DE-10 (PenNovo® MD 10) was obtained from Ingredion Company (Ingredion, Westchester, IL, U.S.A.).

Hexane (ACS Reagent) and ethyl ether (ACS Anhydrous) were purchased from Fisher Scientific Company (Fisher Scientific International, Inc., Pittsburgh, PA, U.S.A.).

2.2. Preparation and Rheological Characterization of Wall Material Solutions. Solutions were prepared by dissolving the required amount of pectin and maltodextrin in deionized (DI) water by stirring on Cole-Parmer Advanced Stirring Hot Plate at 45°C (Cole-Parmer Instrument Company, LLC., Vernon Hills, IL, U.S.A.) until the wall materials completely dissolved. After that, the solutions were kept stirring at 200 rpm with a 1-inch magnetic bar at room temperature overnight for analysis and spray drying. The compositions of all the samples are summarized in Table 3. The concentrations of wall material were selected from preliminary tests based on the solubility of the materials and flowability of the solutions through the 3FN.

Before spray drying, the rheological properties of wall material solutions were measured using a controlled stress rheometer (ARES-G2, TA Instruments, New Castle, DE, U.S.A.). The geometry used was the DIN concentric cylinder (bob diameter of 27.7 mm and cup diameter of 30 mm), and approximately 30 mL of the sample was loaded. The measurements were performed at 25°C from 0.1 to 100/s of shear rates. The collected shear rate–shear stress data best fit with the power-law model: $\tau = K\gamma^n$, where τ is shear stress (Pa), K is the flow consistency index (Pa·s^n), γ is the shear rate (/s), and n is the flow behavior index (dimensionless, $n = 1$ for Newtonian fluid) [32]. Measurements were made in triplicate on three individual samples.

2.3. Spray Drying and Process Yield. The samples were spray dried using a spray drier (Mini Spray Dryer B-290, BÜCHI Labortechnik AG, Postfach, Switzerland) with a 3FN at the inlet temperature of 160°C and the aspirator rate of 100%. The outlet temperature varied from 90 to 105°C. The wall and core materials were pumped through two separate syringe pumps (Harvard Apparatus PHD 2000 Infusion Syringe Pumps Harvard Apparatus, Cambridge, MA, U.S.A.) into the 3FN. The 3FN used an inner nozzle insert for the core material with an inner diameter at the outlet of 0.254 mm. The outer nozzle tip had an inner diameter of 0.7 mm at the outlet, and the atomizing air cap at the end of the 3FN had an inner diameter of 1.5 mm. Soybean

Table 2: Abbreviations, notations, nomenclatures, and symbols.

Term	Meaning
3FN	Three-fluid nozzle
2FN	Two-fluid nozzle
EE	Encapsulation efficiency
LDL	Low-density lipoprotein
DE	Dextrose equivalent
DI	Deionized
°C	Degree Celsius
rpm	Rotations per minute
mL	Milliliter
mm	Millimeter
s	Second
τ	Shear stress
Pa	Pascal
K	Flow consistency index
γ	Shear rate
n	Flow behavior index
%	Percent
g	Gram
w/w	Weight/weight
Y	Yield
W_p	Weight of the sample powder collected
V_{oil}	Volume of soybean oil
ρ_{oil}	Density of soybean oil
SEM	Scanning electron microscope
kV	Kilovolt
mg	Milligram
pH	Potential of hydrogen
nm	Nanometer
L^*	Lightness
a^*	Red and greenness
b^*	Blueness to yellowness
mPa	Millipascal
ρ_{bulk}	Bulk density
W_{bulk}	Bulk mass of the sample
V_{bulk}	Bulk volume of the powder
ρ_{packed}	Packed density
W_{packed}	Packed mass of the sample
V_{packed}	Packed volume of the powder
HR	Hausner's ratio
CI	Carr's index
v/v	Volume by volume
μm	Micrometer
Eq	Equation
MD	Maltodextrin
CLSM	Confocal scanning laser microscopy
α	Alpha
ANOVA	Analysis of variance
LSD	Least significant difference
P	Probability value

Table 3: Concentrations of pectin and maltodextrin in wall material solution.

Wall material	Pectin concentration (%, w/w)	Maltodextrin concentration (%, w/w)
0.5 pectin-10 MD	0.5	10
0.5 pectin-20 MD		20
1 pectin-10 MD	1	10
1 pectin-20 MD		20
2 pectin-10 MD	2	10
2 pectin-20 MD		20
3 pectin-10 MD	3	10
3 pectin-20 MD		20
4 pectin-10 MD	4	10
4 pectin-20 MD		20

Sample codes represent the following: pectin concentration %, w/w-maltodextrin concentration %, w/w (MD: maltodextrin).

oil density was taken as 0.926 g/mL. The flow rates of wall and core materials are shown in Table 4. The mass ratio of core to wall material in the nozzle was 0.15 : 0.85, which was equivalent to 15% w/w of theoretical oil loading in the microcapsule. After spray drying, powders were collected in Falcon 50 mL conical centrifuge tubes (Fisher Scientific International, Inc., Pittsburgh, PA, U.S.A.) for characterization and analysis.

The yield (Y) of the spray drying process was calculated using

$$Y = \frac{W_p}{V_{oil} * \rho_{oil}/0.15} * 100\%, \quad (1)$$

where W_p (g) represents the weight of the sample powder collected from the collecting vessel at the end of the spray drying process, V_{oil} (mL) is the total volume of soybean oil used during spray drying, and ρ_{oil} (g/mL) is the density of soybean oil.

2.4. Characterization of Spray-Dried Powders

2.4.1. Morphology Using Scanning Electron Microscopy. The morphology of the spray-dried samples was observed with a scanning electron microscope (SEM) (FEI Company, Hillsboro, OR U.S.A.) in high-vacuum mode. The voltage used was 3.00 kV with several levels of magnification. The samples were coated with gold-palladium by Denton Desk II TSC turbo-pumped sputter coater (Denton Vacuum, Inc.,

Table 4: Flow rates of wall and core materials during spray drying and process yield. The target oil loading in dried powder was 15% *w*/*w*.

Wall material	Solid content in wall (%, *w*/*w*)	Vol. flow rate of wall (mL/min)	Mass flow rate of wall solids (g/min)	Oil flow rate (mL/min)	Yield (%)
0.5 pectin-10 MD	10.5	5.00	0.525	0.1	30.8 ± 7.70^{bc}
0.5 pectin-20 MD	20.5	2.56			23.9 ± 2.06^{ab}
1 pectin-10 MD	11	4.77			31.3 ± 2.83^{bc}
1 pectin-20 MD	21	2.50			26.5 ± 6.99^{bc}
2 pectin-10 MD	12	4.38			40.6 ± 5.42^{d}
2 pectin-20 MD	22	2.39			29.5 ± 3.11^{bc}
3 pectin-10 MD	13	4.04			32.6 ± 3.32^{cd}
3 pectin-20 MD	23	2.28			23.1 ± 2.42^{ab}
4 pectin-10 MD	14	3.75			29.8 ± 9.12^{bc}
4 pectin-20 MD	24	2.19			21.3 ± 4.73^{a}

Sample codes represent the following: pectin concentration %, *w*/*w*-maltodextrin concentration %, *w*/*w* (MD: maltodextrin). One-way ANOVA, Fisher's LSD ($P < 0.05$). Numbers sharing the same letter are not significantly different.

Moorestown, NJ, U.S.A.) for 70 seconds prior to scanning. Representative images were selected for presentation.

2.4.2. Confocal Scanning Laser Microscopy. The internal structure of the dried powders was characterized using a Zeiss LSM7 Live confocal laser scanning microscope (CLSM) (Carl Zeiss AG, Oberkochen, Germany) of the samples dyed with fluorescein sodium. A fluorescein sodium solution with a 1 mg/mL concentration at a pH of 8.5 was added to the wall solution in the mass ratio of feed solid of 1 : 6000 before spray drying. The laser supplies a 488 nm excitation wavelength, which allows the emission of fluorescein sodium, with excitation/emission wavelengths of 485/530 nm (Apinan [33]). A 63x objective was used for the images.

2.4.3. Color. The color of spray-dried samples was measured by a colorimeter (HunterLab, Reston, VA, U.S.A.). The values of L^*, a^*, and b^* were determined. L^* values represent the lightness of the samples and range from darkness to lightness (0-100). Negative a^* values represent greenness, and positive a^* values represent redness. Similarly, b^* values represent blueness when negative and yellowness when positive. Readings were done in triplicate for each sample.

2.4.4. Particle Size and Size Distribution. The particle size of the samples was measured using a laser scattering particle sizer (Shimadzu SALD-2300 Laser Particle Size Analyzer, Shimadzu, Kyoto, Japan) in dry mode with a Cyclone Injection Type Dry Measurement Unit (SALD-DS5). The average signal count of one measurement was 64, the signal accumulation count was one, and the interval was 2 seconds. About 1 g of the powder sample was placed in a cyclone, and an elevator speed of 10 mm/sec with an air pressure of 0.30 MPa was used. The measurements were made in triplicate.

2.4.5. Flowability. To represent the flowability, the bulk density and packed density of the samples were measured first. As for the bulk density, the sample was poured into a Kimble Kimax 25 mL graduated cylinder (Kimble Chase Life Science and Research Products LLC, Vineland, NJ, U.S.A.), and the volume was read from the cylinder directly as the weight was measured by an analytical balance (Mettler Toledo, Columbus, OH, U.S.A.).

The bulk density was calculated using

$$\rho_{\text{bulk}} = \frac{W_{\text{bulk}}}{V_{\text{bulk}}}, \tag{2}$$

where W_{bulk} is the mass of the sample in the cylinder and V_{bulk} is the volume of the powder. To measure the packed density, after pouring the sample into a 20 mL graduated cylinder, the cylinder was packed until the powder volume did not change; then, the volume and weight were recorded.

The packed density was calculated using

$$\rho_{\text{packed}} = \frac{W_{\text{packed}}}{V_{\text{packed}}}, \tag{3}$$

where W_{packed} is the mass of the sample in the cylinder and V_{packed} is the volume of the powder after the tapping.

Both bulk and packed densities were measured three times.

Once the bulk density and packed density were measured, the flowability and cohesiveness of the powder samples were calculated through the Hausner ratio (HR) and the Carr Index (CI), respectively, according to the following equations [34].

$$\text{Hausner ratio (HR)} = \frac{\text{packed density}}{\text{bulk density}} \ (>1.25, \text{poor flowability}),$$
$$\text{Carr index (\%CI)} = \frac{\text{packed density} - \text{bulk density}}{\text{pakced density}} \times 100 \ (>25, \text{poor flowability}). \tag{4}$$

2.4.6. Encapsulation Efficiency. The total oil content of microcapsules was measured with the Soxhlet extraction method using 50% *v*/*v* hexane+50% *v*/*v* ethyl ether as the solvent for 12 hours (AOAC Method 920.39, [35]). The total oil loading was expressed as a percentage of oil based on the

weight of the microcapsules. According to the experimental design, the theoretical loading of oil is 15%, which is consistent across the samples.

The surface oil content (nonencapsulated oil fraction) of microcapsules was determined according to the previous studies [36, 37] with some modifications. Briefly, around 1 g ± 0.1 g of powder was mixed with 15 mL of hexane in a 50 mL Falcon tube and vortexed for 2 minutes. The mixture was centrifuged (model IEC CENTRA CL2, Thermo Scientific Inc., Waltham, MA. U.S.A.) for 15 min at $2310 \times g$. After centrifuging, the mixture was filtered using a no. 42 0.25 μm filter paper (Whatman plc, Maidstone, Kent, U.K.) into a glass vial. The residue on the filter paper was washed with 20 mL of hexane three times. The filtered solvent in the glass vial was evaporated in the hood overnight until a constant weight was reached. The surface oil extracted was weighed and expressed as a percentage of oil based on the weight of the microcapsules.

Encapsulation efficiency was determined by calculating the ratio of total oil contained in the microcapsules and the free oil on the surface of the capsules, according to

$$\text{Encapsulation efficiency } (\%) = \frac{\text{total oil} - \text{surface oil}}{\text{total oil}} \times 100\%. \quad (5)$$

Effective encapsulation efficiency based on the theoretical loading of oil was calculated as

$$\text{Effective encapsulation efficiency } (\%) = \frac{\text{total oil} - \text{surface oil}}{\text{theoretical loading of oil}} \times 100\%. \quad (6)$$

All determinations were performed at least in triplicate.

2.5. Comparison between Spray Drying with Three-Fluid Nozzle and Two-Fluid Nozzle. Based on the results, in Table 4, from the experimental design, laid out in Table 3, a formulation of 1% *w*/*w* sugar beet pectin and 20% *w*/*w* maltodextrin was selected to compare the properties of microcapsules prepared with a 3FN and a 2FN. Discussion of these results is expanded on in Section 3.2.1.

For the 3FN sample, the preparation was the same as stated earlier. The flow rates for the wall and core were 2 and 0.1 mL/min, respectively. For the 2FN sample, the emulsion was prepared before spray drying. Sugar beet pectin (1% *w*/*w*) and maltodextrin (20% *w*/*w*) were dissolved with DI water at room temperature and hydrated overnight. Soybean oil was dispersed into the wall material solution gradually and shear mixed with the solution at 15,000 rpm for 3 minutes using a high-speed homogenizer (IKA-ULTRA-TURAX® T-25 basic, IKA® Works Inc., Wilmington, NC, U.S.A).

The amount of soybean oil added into the solution was calculated based on the volume; the ratio of soybean oil to the solution was 1/20 *v*/*v*. After that, the emulsion was homogenized at 551 bar using a microfluidizer (M-110P, Microfluidics, Westwood, MA, U.S.A.) for enhanced emulsion stability. The emulsion was pumped into the 2FN of the Mini Spray Dryer B-290 with a flow rate of 2.1 mL/min by a syringe pump (PHD 2000 Infusion Syringe Pump, Harvard Apparatus, Cambridge, MA, U.S.A.). The inlet temperature was set as 160°C, and the outlet temperature varied from 99 to 106°C.

The particle size distribution, surface oil, and total oil of microcapsules were measured using the methods described in 2.4.4 and 2.4.6.

2.6. Statistical Analysis. The samples were made from three batches of spray drying for each formulation. All the measurements were repeated three times. Data were analyzed by analysis of variance (ANOVA) with Microsoft Excel (Microsoft Corp., Seattle, WA, U.S.A.) and OriginPro software (OriginLab Corp., Northampton, MA, U.S.A.). Mean comparison of data was determined by Fisher's least significant difference (LSD) at $P < 0.05$.

3. Results and Discussion

3.1. Rheological Properties of Wall Material Solutions. The rheological properties of the wall material solutions with different concentrations of maltodextrin and pectin fitted the power-law model (Figure 2). As expected, at the same maltodextrin concentration, the flow consistency index increased when the pectin concentration increased from 0.5 to 4% *w*/*w*. Sugar beet pectin is not capable of forming gels due to its molecular structure, so its impact on the solution viscosity change is limited compared to other pectins [38]. At the same pectin concentration, the flow consistency index also increased as maltodextrin concentration increased. As for the flow behavior index, it was close to one at the lowest pectin concentration indicating that the solution resembled the behavior of a Newtonian fluid. The solutions exhibited more shear thinning behavior at higher pectin and maltodextrin concentrations as their flow behavior index decreased below one. Sugar beet pectin solutions typically show shear thinning behavior [39]; therefore, this behavior is in line with the previous research.

3.2. Properties of Spray-Dried Microcapsules

3.2.1. Spray Drying Yield. When applying the 3FN for spray drying, the yield of the spray drying process was one of the major concerns, which is related to the efficiency of the process as well as production costs [40]. The yield of all spray-dried samples varied between 16.0 and 46.2% (Table 4). Generally, with the same pectin concentration, the lower maltodextrin concentrations in the wall material solution resulted in a significantly higher process yield; this may relate to the increasing viscosity at the higher maltodextrin concentrations. The higher ratio of the pectin in the wall material solution may have provided a higher emulsification capacity. To obtain a better solid recovery during spray drying, the optimum viscosity of feed is required, as viscosity relates to the glass transition temperature or stickiness temperature of droplets. Higher viscosity over a critical value causes the agglomeration of the particles due to the formation of viscous bridges [41]. The sample 4 pectin-20 MD

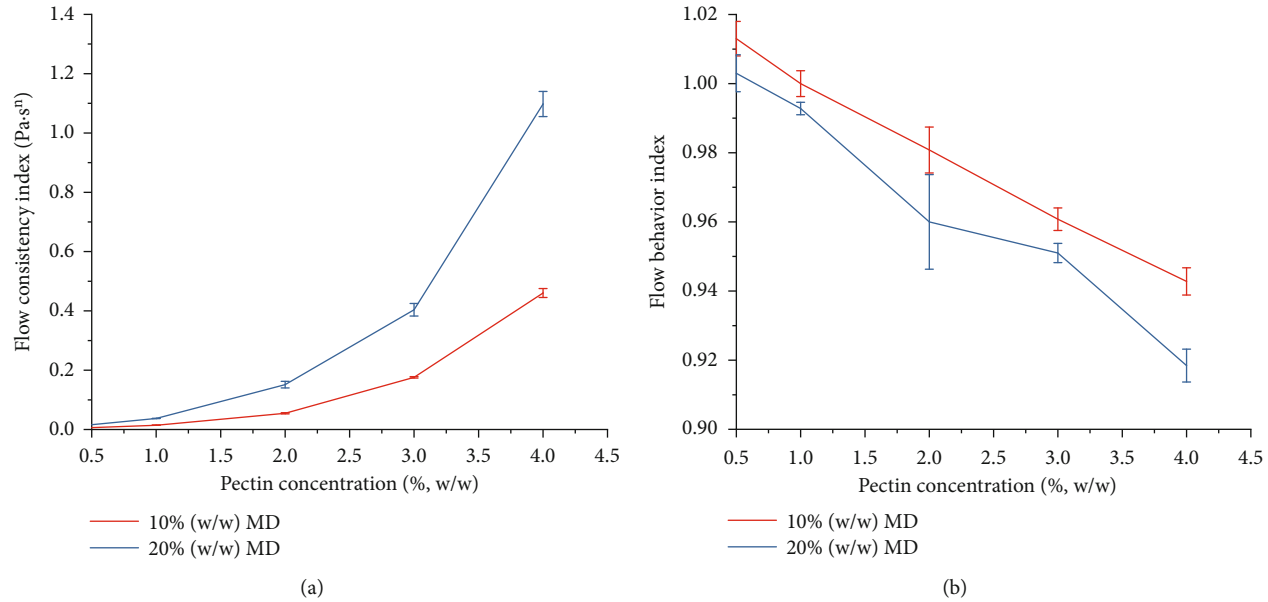

Figure 2: Rheological of wall material with different concentrations of maltodextrin and pectin before spray drying properties: (a) flow consistency index; (b) flow behavior index. Abbreviation: MD: maltodextrin. One-way ANOVA, Fisher's LSD ($P < 0.05$). Statistically different treatments have nonoverlapping error bars.

had the lowest average process yield of 21.3% and 2 pectin-10 MD had the highest average process yield of 40.6% (Table 4).

3.2.2. Morphology and Internal Structure. The surface morphology and internal structure of spray-dried samples are presented in Figures 3 and 4. The SEM images in Figure 3(a) include the samples with all formulations of wall material for spray drying. Overall, the difference in rheological properties of wall material had a limited impact on the surface morphology of spray-dried samples indicating that the drying mechanisms such as evaporation, expansion, and shrinking of the particles were similar across the studied range of viscosity and formulation. The formation of droplets was impacted by the wall material properties which will be discussed in Section 3.2.4. Most spray-dried samples had wrinkled surfaces with some microcapsules having a spherical shape with a hollow core, as shown in Figure 3(b). The CLSM images confirmed the internal structure of spray-dried samples, showing that most of the microcapsules contained hollow cores inside.

It was reported that the extent of surface wrinkling was associated with the sugar beet pectin concentration in the feed emulsions when encapsulating fish oil by spray drying with a 2FN. Increasing pectin concentrations from 1.1% to 2.2% caused a more wrinkled surface [8], but the same effect was not observed in this study. Similarly, to our findings, it was reported that the dented, wrinkled, or hollow core shape may be a function of the core/coat ratio of spray-dried materials [42], indicating that either the 3FN or the viscoelastic properties of pectin could cause a more wrinkled surface. Based on the SEM images, it was found that the particle size of microcapsules became more polydisperse with increasing the pectin concentration, as more extralarge and very fine particles existed in the 3 pectin-20 MD and 4 pectin-20 MD samples. The inhomogeneity of pressure differential at the point of atomization by a compressed gas had a more profound impact when the feed viscosity was higher, creating either very fine or large particles.

3.2.3. Color. The color of food powder is an important parameter in the food industry, which is related to the surface component content, as color reflects the sensory attractiveness of the product and influences the powder applications in different types of foods [40, 43]. Due to the color of maltodextrin and sugar beet pectin, in general, the spray-dried powders showed white with a slightly yellow color. So, the L^* value and b^* value impacted by the composition of the wall material were focused on in Figure 5. The L^* values were in the range of 87.09 to 91.85 and showed a decreasing trend as the pectin concentration increased at the same maltodextrin concentration. The b^* values were all positive (1.72 to 3.63) and increased as the pectin concentration increased, which was expected since the color of pectin powder was darker and has a yellower hue than maltodextrin [44].

3.2.4. Particle Size and Size Distribution. Particle size and size distribution are important parameters affecting the flowability, compressibility, bulk density, and stability of the particles [45]. For example, the oxidation level of powdered foods varies with particle size and decreased in smaller particles [46].

The particle size distribution shown in Figure 6 indicated that spray-dried powders were fine powders, as the median particle diameters of the samples were between 9 and 16 μm (Table 5), which were in the typical size range of powders produced from spray drying (10-50 μm) [3]. Generally,

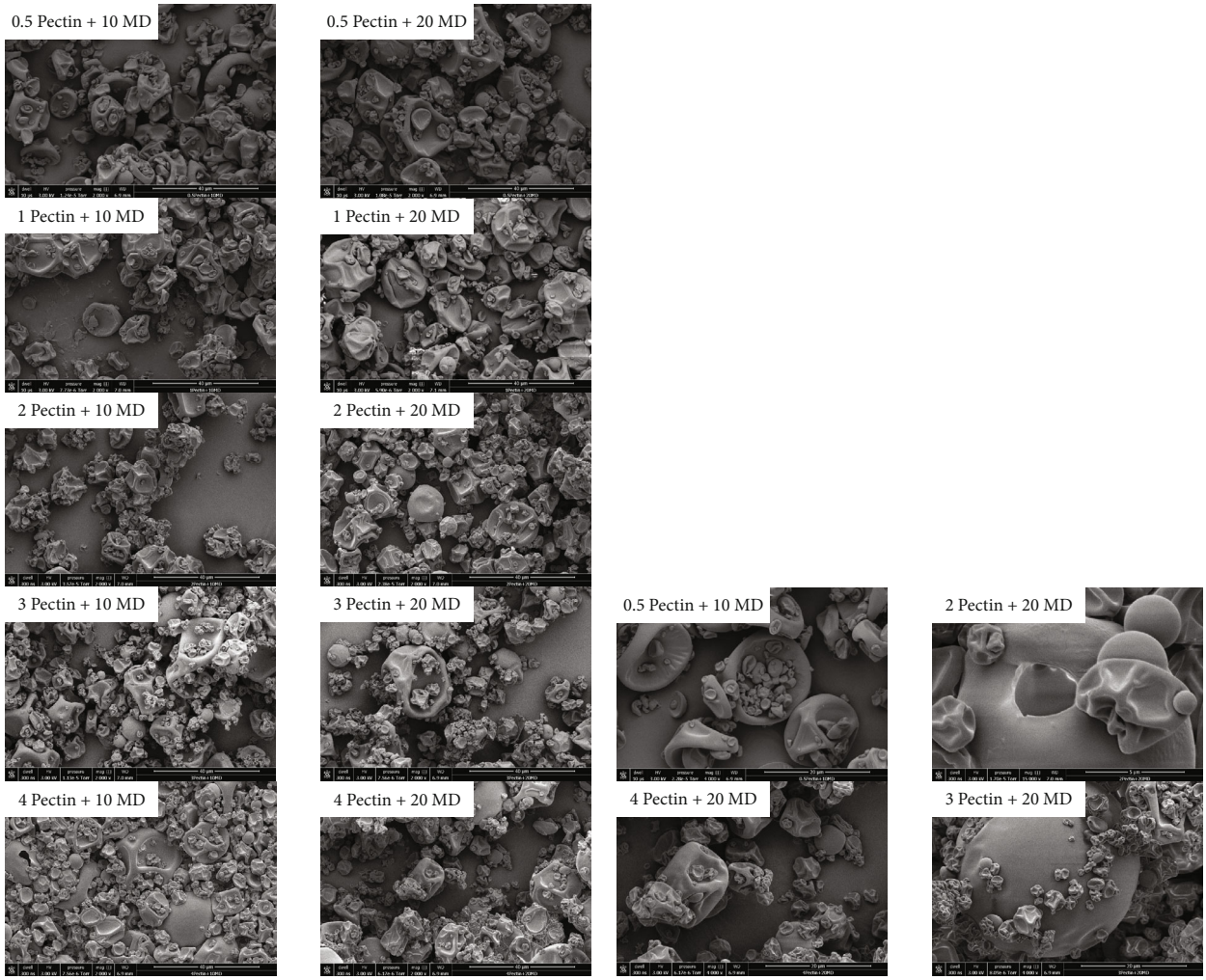

Figure 3: Scanning electron microscopy images of spray-dried microcapsules, sample information labeled in the figures: (a) images for all samples, 2,000x magnified; (b) images with 4,000x and 15,000x magnified. Sample codes represent the following: pectin concentration %, *w*/*w*-maltodextrin concentration %, *w*/*w*.

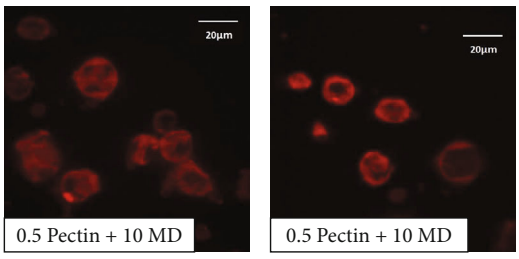

Figure 4: Confocal laser scanning microscopy images of spray-dried 0.5 pectin+10 MD microcapsules.

the particle size distribution was wider with a lowered peak of each curve as the flow consistency index increased. This trend of particle size distribution was also confirmed by the SEM images. At the low concentration of the wall material, the particles were more even in size; the particle size became more uneven at the high concentration of the wall material as there were more microcapsules of lower size. Higher viscosity and high pectin concentration may have caused an inhomogeneous break of the feed into droplets.

Previous studies have found an increase in particle size of spray-dried powders with feed solutions of higher viscosity, which may be attributed to the larger droplets formed at the nozzle tip [47–49]. In our study, this trend was not observed when comparing the median particle diameter of spray-dried samples. It might be due to the low feed flow rate compared to the compressed gas flow rate which limits the impact of the viscosity on the particle size. Further studies are necessary to confirm the impact of the flow rate.

3.2.5. Flowability. During the storage, compaction, and transportation of spray-dried powder, good flowability is necessary for easy handling [34, 49]. The Hausner ratio and Carr index were selected to represent the cohesiveness and flowability of spray-dried samples in this study. For the Hausner ratio, the classifications are "low" (HR < 1.2), "intermediate" (1.2 < HR < 1.4), and "high" (HR > 1.4); for

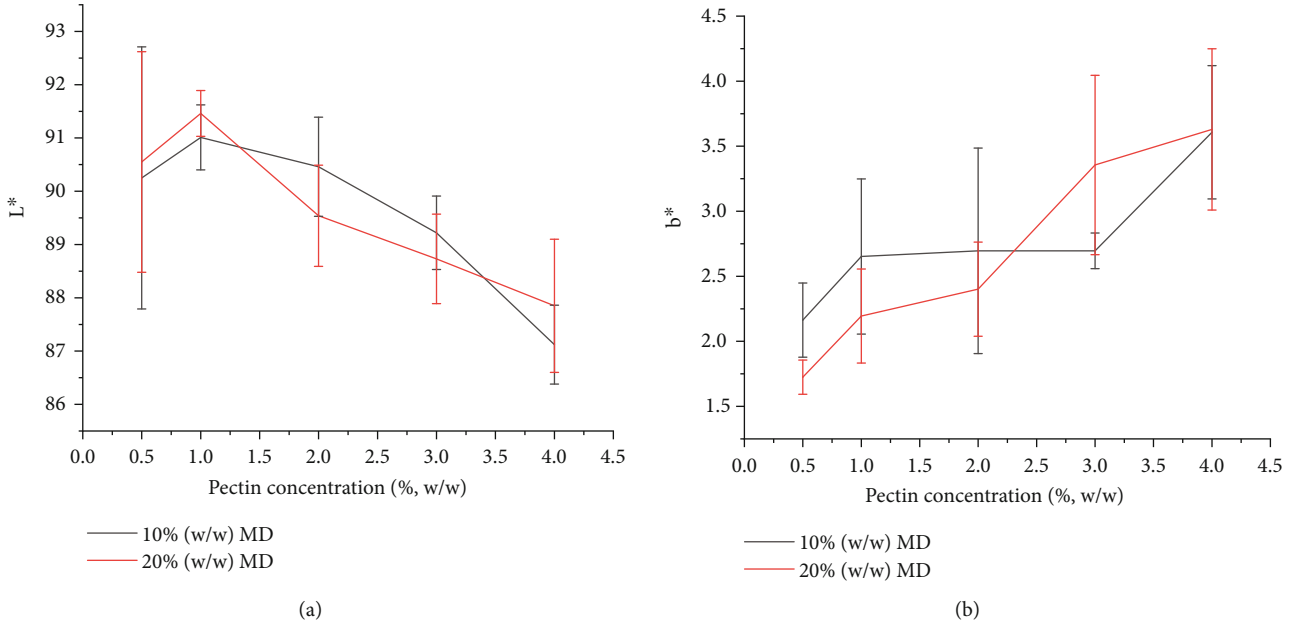

Figure 5: Color of spray-dried microcapsules with different concentrations of sugar beet pectin and maltodextrin: (a) L^*: brightness; (b) b^*: yellowness. One-way ANOVA, Fisher's LSD ($P < 0.05$). Statistically different treatments have nonoverlapping error bars. Abbreviation: MD: maltodextrin.

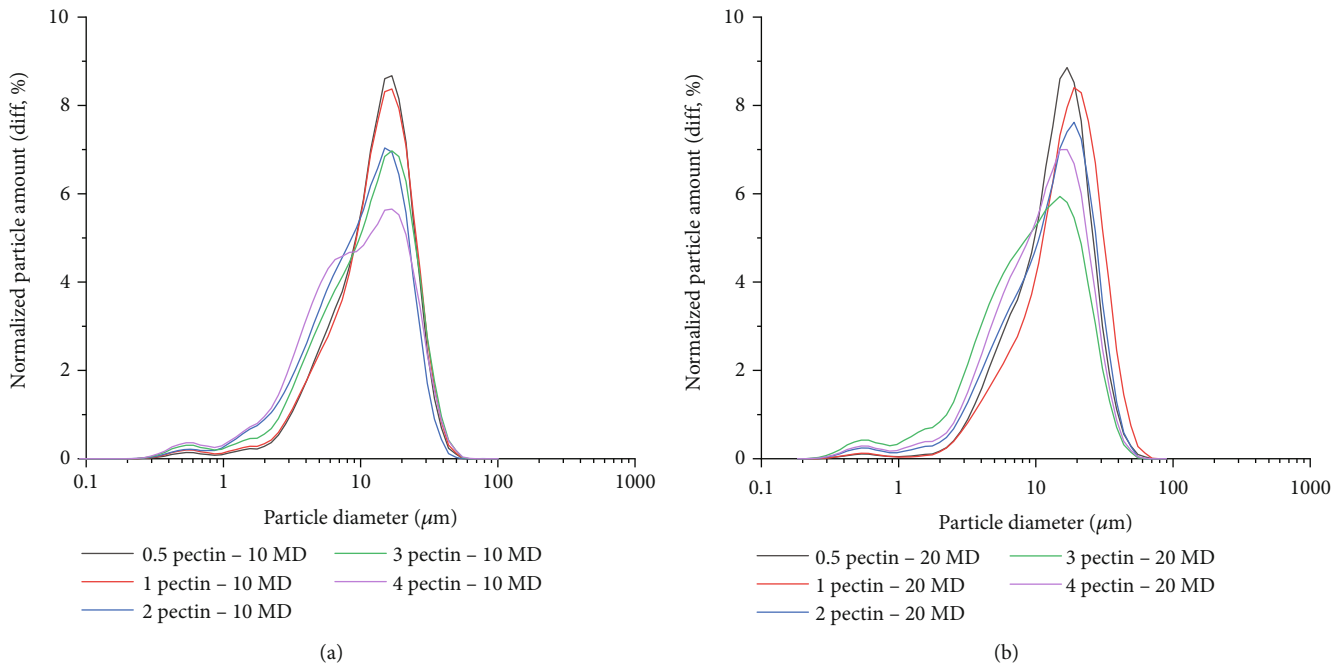

Figure 6: Particle size distribution of spray-dried microcapsules: (a) samples made with 10% w/w maltodextrin; (b) samples made with 20% w/w maltodextrin. Abbreviation: MD: maltodextrin.

the Carr index, the classifications are "very good" (CI < 15%), "good" (15 < CI < 20%), "fair" (20 < CI < 35%), "bad" (35 < CI < 45%), and "very poor" (CI > 45%) [50].

The Hausner ratio values of all spray-dried samples were higher than 1.4, and the Carr index values were between 35 and 45%, indicating "high" cohesiveness and "bad" flowability. The flowability is significantly influenced by the size and shape of powder particles [51]. Particles larger than 250 μm are usually free flowing. When particle sizes fall below 100 μm, powders become increasingly cohesive and flow problems are likely to occur. Particles that have less than 10 μm diameter size are usually extremely cohesive and resist flow under gravity [52]. Most of our samples were in the particle size range of 10 to 20 μm, which were in the typical range of the spray-dried particles without an agglomeration process and caused poor flowability.

Table 5: Median particle diameter for spray-dried samples.

Sample	Median diameter (μm)
0.5 pectin-10 MD	12.67 ± 0.70^{c}
0.5 pectin-20 MD	13.46 ± 0.46^{c}
1 pectin-10 MD	12.77 ± 1.81^{c}
1 pectin-20 MD	15.81 ± 1.28^{d}
2 pectin-10 MD	10.18 ± 2.05^{ab}
2 pectin-20 MD	13.01 ± 0.46^{c}
3 pectin-10 MD	11.50 ± 1.39^{bc}
3 pectin-20 MD	9.38 ± 0.93^{a}
4 pectin-10 MD	12.09 ± 0.24^{bc}
4 pectin-20 MD	11.24 ± 1.23^{ab}

Sample codes represent the following: pectin concentration %, *w*/*w* -maltodextrin concentration %, *w*/*w* (MD: maltodextrin). One-way ANOVA, Fisher's LSD ($P < 0.05$). Numbers sharing the same letter are not significantly different.

The Carr index and Hausner ratio values by pectin concentration are reported in Figure 7. In general, the flowability and cohesiveness increased as the flow consistency index of the wall material solution increased. This showed the same trend as the particle size distribution, as more small particles were produced when the pectin concentration increased.

3.2.6. Surface Oil and Encapsulation Efficiency. Surface oil is often defined as the oil that may be extracted with organic solvents from the surface of unbroken microcapsules [6]. It is a critical measurement for the encapsulation process as successful encapsulation of oils should result in encapsulated powders with minimum surface oil content [27]. As shown in Table 6, the surface oil content of the spray-dried samples was in the range of 1.3 to 3.4%, and the results were comparable to the previous study using the 3FN to encapsulate fish oil, in which surface oil was 4.4% *w*/*w* [6]. Within the same pectin concentration, the surface oil content of the sample with 20% *w*/*w* maltodextrin was significantly higher than the sample with 10% *w*/*w* maltodextrin. The flow consistency index of the wall material solution with 20% *w*/*w* maltodextrin was significantly higher than the solution with 10% *w*/*w* maltodextrin at the same pectin level due to the higher solid content (Figure 2). When the maltodextrin concentration was consistent, the flow consistency index showed an increasing trend as increasing the pectin concentration from 1to 4% *w*/*w*.

These results indicated that increasing the flow consistency index of wall material solution when spray drying with a 3FN would generally increase the surface oil content of a spray-dried sample while there was also an optimum concentration. The sample 1 pectin-10 MD showed the least surface oil content, which indicated that the pectin content in this sample was enough to create a stable emulsion to encapsulate the oil. With 3FN, to reach the same theoretical oil loading in the sample, the flow rate of the wall material decreased as the concentration of the wall material increased, which resulted in the oil droplet not fully covered by the wall material solution before the atomization process. When compared to the sample made with 10 and 20% *w*/*w* maltodextrin, lower surface oil content and higher encapsulation efficiency with lower maltodextrin concentration were observed, which may be due to the higher ratio of pectin in the wall at 10% *w*/*w* maltodextrin samples.

For the total oil in the spray-dried sample, the theoretical oil load was fixed at the same level (15% *w*/*w*) across treatments, and the measured total oil load in the spray-dried powder was between 12.4 and 19.6%, indicating that the spray drying with a 3FN was a promising way to create oil-loaded microcapsules. Overall, the samples with low and medium viscous wall material had higher total oil loading than the samples with high viscosity wall materials.

During the spray drying, it was also observed that the wall material solutions of high viscosity formed a film on the spray drying chamber wall, resulting in less solid retained in the dry products. It may be because the wall material, especially pectin, did not move to the surface fast enough to produce a stable emulsion due to the high viscosity, causing more oil to not be encapsulated at the tip of the nozzle. In a future study, other wall materials such as gum arabic should be explored to investigate the effect of viscosity on oil loading in 3FN as it has excellent emulsifying properties and shows good core material retention during spray drying [27].

The encapsulation efficiency was closely related to the surface oil content and total oil retention in the spray-dried powder. The encapsulation efficiency for spray-dried samples ranged between 74.7 and 91.2% (Table 6), which was comparable to the previous study using a 3FN to create microcapsules of fish oil with the reported microencapsulation efficiency of 85.8% [6]. The encapsulation efficiency, in general, was higher at 1% and 2% pectin due to low surface oil and high total oil loading in the samples. Among the samples, 1 pectin-20 MD showed the highest encapsulation efficiency at 91.2%, which was close to the previous study that encapsulated emulsion-based walnut oil by 3FN with the highest encapsulation efficiency at 94.0% [16], indicating that it is an optimum concentration of wall material for a future study.

When pectin concentration was higher than 2%, the effective encapsulation efficiency was significantly lower, which means higher viscosity of wall material is not an ideal option to encapsulate the soybean oil when spray drying with the 3FN along with the spray drying parameters used in this study.

3.3. Comparison of Microcapsules Made with Three-Fluid Nozzle and Two-Fluid Nozzle. Table 7 presents the particle size and encapsulation performance of the samples made with the 3FN and 2FN with the same formulation. 2FN samples showed a significantly smaller size than 3FN samples. The particle size of spray-dried microcapsules is greatly influenced by the droplet size formed after atomization [53], as smaller droplet sizes produced smaller microcapsules. The high shear mixing and homogenization process of 2FN samples reduced the emulsion droplet size significantly, leading to a smaller median diameter compared to the 3FN samples.

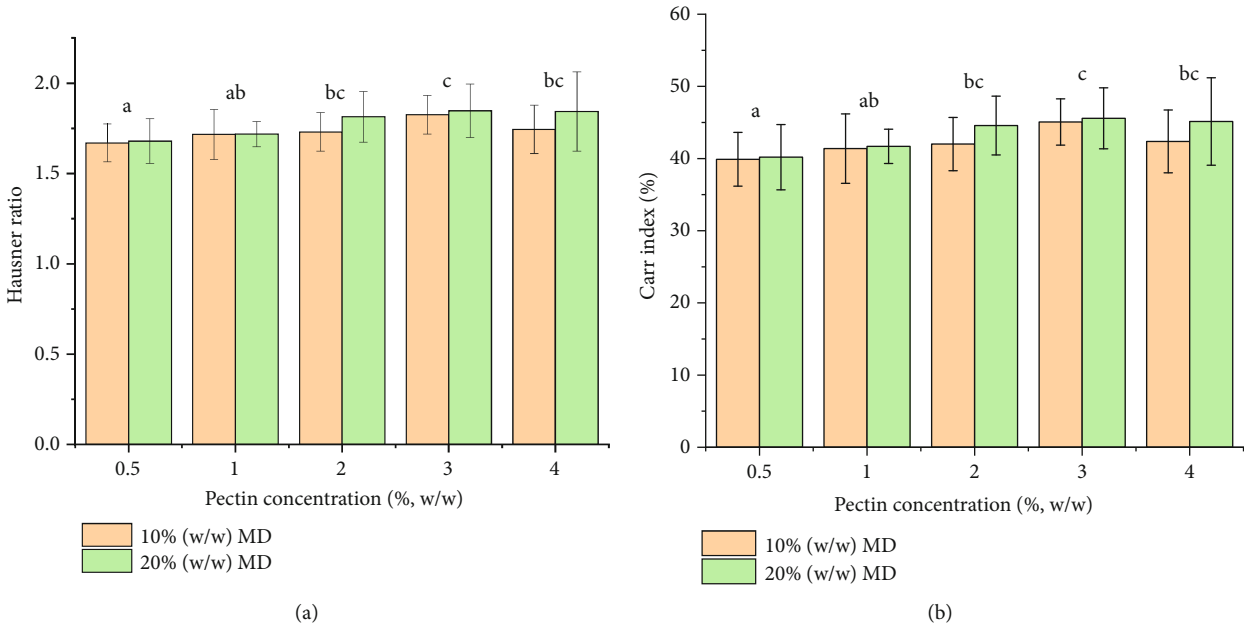

Figure 7: Flowability of spray-dried microcapsules made with different concentrations of sugar beet pectin and maltodextrin: (a) Hausner's ratio; (b) Carr's index. One-way ANOVA, Fisher's LSD ($P < 0.05$). Columns sharing the same letter are not significantly different. Abbreviation: MD: maltodextrin.

Table 6: Surface oil, total oil loading, encapsulation efficiency, and effective encapsulation efficiency of spray-dried samples.

Sample	Surface oil (%, w/w)	Total oil loading (% w/w)	Encapsulation efficiency (%)	Effective encapsulation efficiency (%)
0.5 pectin-10 MD	2.5 ± 0.88^{cd}	17.3 ± 1.56^{de}	86.0 ± 6.58^{cde}	97.9 ± 13.80^{cd}
0.5 pectin-20 MD	2.7 ± 0.80^{cde}	17.2 ± 1.38^{cd}	84.4 ± 0.90^{bcde}	96.8 ± 6.99^{bcd}
1 pectin-10 MD	1.3 ± 0.67^{a}	14.9 ± 4.06^{b}	90.4 ± 6.23^{e}	90.3 ± 31.13^{abcd}
1 pectin-20 MD	1.5 ± 0.23^{ab}	17.9 ± 2.34^{de}	91.2 ± 1.20^{e}	109.0 ± 15.33^{d}
2 pectin-10 MD	2.1 ± 1.32^{bc}	17.4 ± 2.01^{de}	87.6 ± 10.23^{de}	103.1 ± 21.29^{cd}
2 pectin-20 MD	3.1 ± 0.93^{de}	19.6 ± 1.40^{e}	86.1 ± 2.44^{cde}	110.6 ± 8.59^{d}
3 pectin-10 MD	3.0 ± 0.69^{de}	14.6 ± 1.59^{ab}	79.5 ± 4.83^{abcd}	77.9 ± 7.40^{abc}
3 pectin-20 MD	3.2 ± 0.41^{de}	12.4 ± 2.58^{a}	74.7 ± 5.03^{a}	62.1 ± 17.82^{a}
4 pectin-10 MD	3.1 ± 0.35^{de}	13.3 ± 1.37^{ab}	76.6 ± 4.71^{ab}	68.6 ± 9.56^{ab}
4 pectin-20 MD	3.4 ± 0.14^{e}	15.0 ± 2.87^{bc}	77.0 ± 4.92^{abc}	77.5 ± 18.68^{abc}

Sample codes represent the following: pectin concentration %, w/w-maltodextrin concentration %, w/w (MD: maltodextrin). One-way ANOVA, Fisher's LSD ($P < 0.05$). Numbers in the same column that share the same letter are not significantly different.

Table 7: Comparison of microcapsules made with 3FN and 2FN.

Sample	D_{50} (μm)	Surface oil (%, w/w)	Total oil loading (%, w/w)	Encapsulation efficiency (%)	Effective encapsulation efficiency (%)
1 pectin-20 MD-3FN	$10.29 \pm 2.28_{b}$	$2.2 \pm 0.20_{b}$	13.8 ± 2.06^{b}	84.0 ± 0.62^{a}	70.3 ± 4.08^{b}
1 pectin-20 MD-2FN	6.79 ± 1.50^{a}	1.6 ± 0.11^{a}	10.7 ± 0.57^{a}	85.4 ± 0.49^{a}	55.6 ± 1.85^{a}

Sample codes represent the following: %, w/w sugar beet pectin-%, w/w maltodextrin-the three-fluid nozzle/the two-fluid nozzle. The numbers in the same column sharing the same letter are not significantly different (Fisher's LSD, $P < 0.05$). D_{50}: the portions of particles with diameters smaller and larger than this value are 50%. Abbreviations: MD: maltodextrin; 3FN: three-fluid nozzle; 2FN: two-fluid nozzle.

For the surface oil, the samples made with the 2FN showed significantly lower surface oil content compared to the samples made with the 3FN, which was probably due to the formation of stable emulsions by high energy emulsification. For total oil loading, the samples made with 3FN were significantly higher than the samples made with 2FN,

resulting in comparable encapsulation efficiency and higher effective encapsulation efficiency. The cause of this difference may be related to the emulsion preparation before spray drying and droplet formation after atomization.

For the samples made with the 2FN, during the high shear mixing and homogenization, the oil may be encapsulated using most of the wall materials. For the 3FN, the amount of wall material necessary to encapsulate the oil may be smaller than the 2FN process. Sugar beet pectin provided emulsification properties by increasing emulsion stability but also raised viscosity. Higher viscosity limited the migration of crust-forming materials (e.g., maltodextrin) towards the droplet surface, which could affect the wall structure formation and oil entrapment [54]. However, for samples made with the 3FN, oil was pumped into the nozzle directly, thus limiting the oil loss before spray drying. Overall, the spray drying with the 3FN showed promising results to replace 2FN for oil encapsulation.

4. Conclusions and Prospects

As an exploration of applying 3FN during spray drying for the oil encapsulation, this study created oil-loaded microcapsules using immiscible oil and wall material solutions without forming emulsion before spray drying. The total oil loading was close to the theoretical amount, 13.8 ± 2.06 % actual compared to 15% theoretical, showing that 3FN is a potential tool to be used in the spray drying process, especially in application of oil microencapsulation without the need for high pressure homogenization, which is of key interest for industrial applications.

In this study, the rheological properties of the wall material solution did not affect the morphology of microcapsules when spray dried with a 3FN but affected other properties of the spray-dried powders. As viscosity increased, the particle size distribution of powders became wider, the cohesiveness of powders increased, the surface oil content of microcapsules increased from 2.5 ± 0.88 to 3.4 ± 0.14%, and the encapsulation efficiency decreased generally from 84.4 ± 0.90 to 77.0 ± 4.92%. There were also indications of the optimum wall material concentration or viscosity for the encapsulation efficiency. This study only investigated the combination of maltodextrin and pectin as the wall material and soybean oil as the core material. To implement this technology to broader applications, various materials should be evaluated with the 3FN during spray drying. Also, other parameters influencing spray drying such as the flow rates of the feed liquid, the composition of materials used, and the spray drying temperature should all be explored in future studies to expand the understanding of the spray drying process with a 3FN.

Data Availability

All pertinent data has been contained within the manuscript.

Additional Points

Practical Application. The findings from this study can be useful for optimizing the formulation and process conditions to produce encapsulated oil with maltodextrin and sugar beet pectin using spray drying with a 3FN, which can eliminate the need for homogenization and expand the application of 3FN in the food industry.

Disclosure

This manuscript was originally presented as one chapter of a thesis.

Conflicts of Interest

The authors declare that they have no conflicts of interest and no affiliation with any organization with a direct or indirect financial interest in the subject matter discussed in the manuscript.

Authors' Contributions

Jingwen Cai designed the study, collected test data, interpreted the results, and drafted the manuscript as part of her dissertation. Ricardo Lopez assisted in collecting samples, obtaining analytical data, and editing the manuscript. Youngsoo Lee was the principal investigator, who advised the researcher. He reviewed and revised the proposed idea of the research and the final manuscript prior to submission.

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