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**Sustainability Spotlight Statement**

Being a sustainable alternative to traditional animal-based foods, plant-based foods are increasingly used in human diets. However, inferior techno-functional properties of plant-based ingredients, which are causing inferior sensory attributes in foods, are hindering the expected growth of plant-based foods in the commercial market. The high-oil-containing powder presented in this manuscript has better functional properties, such as dispersibility and reconstitutability, making it suitable for enhancing sensory attributes of various plant-based products such as plant-based milk, cream, cream cheese, cheese, etc. The oil core present in oil globules can also be used as a vehicle for delivering hydrophobic health-promoting compounds, helping enhance the well-being of humans. We understand that this product/process would help achieve SDGs 3, 9 and 13.

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**Innovative formulations using spray-drying technology for plant-based high-oil powders: physicochemical and micro-structural analyses**

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## ABSTRACT

Plant-based ingredients, which are considered sustainable sources, are increasingly used to produce food alternatives to animal-origin products. However, despite being considered a sustainable option, the wider acceptance of plant-based alternative foods is poor. The major reasons are the lack of desirable functionalities in plant-based food ingredients and the inferior sensory attributes of prepared foods and the lack of desirable functionalities in plant-based food ingredients compared to their animal-based counterparts. To fulfil this gap, this study focuses on the production and characterization of plant-based high-fat powder with enhanced functionalities, which could serve as an alternative ingredient to the dairy-based cream powder in the food manufacturing sector. Plant-based high-oil powders containing 20% and 40% total oil were prepared from corn oil emulsion having a mean oil globule size of 0.47μm and 0.75μm by spray drying. Formulations used a water-soluble fraction of mung bean protein isolate as an emulsifier

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and maltodextrin as a wall material. The physicochemical analyses of the powders revealed that the powder prepared from corn oil emulsion with a mean fat globule size (D[4,3]) of 0.47µm and 20% oil content had a lower angle of repose, higher bulk density and lower free oil content than other high-oil powder samples. The confocal laser scanning microscopy (CLSM) and scanning electron microscopy (SEM) images also showed that powders prepared from smaller fat globules were individual, with low surface oil coverage compared to the powders prepared from larger fat globules. This study highlighted the suitability of plant-based sources for developing high-oil powders that could find potential applications in creating valuable food products.

**KEYWORDS**

Plant-based high-oil powder, emulsion, mung bean, spray drying, microstructure

## 1. Introduction

High-fat/oil powders are widely used in food product development because of their nutritional, textural, and flavor attributes. These powders, containing a high percentage of fats, from 40%, find applications in bakery products, soups, sauces, processed meats, creamy condiments, evaporated milk, infant formulas, cheese, coffee, and tea. Owing to their low water activity and moisture content, these powders have a long shelf life and are convenient for transportation, storage, and use. These powders are manufactured at the commercial scale using spray drying to ensure they possess essential instant properties such as wettability, sinkability, solubility and dispersibility. While high-fat powders have a long shelf life due to their low water activity and moisture content, they are more prone to fat oxidation and particle caking during storage. The fat oxidation can be prevented by encapsulating it in a wall material before spray drying [1-3].

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The first step for the encapsulation is to prepare the stable emulsion, which can be done through homogenization techniques such as high-pressure homogenization, ultrasonication, and microfluidization [4]. Among these, ultrasonication forms a nano emulsion with the least energy and offers better stability [5].

Different synthetic (polysorbates, acetylated mono and diglycerides, carboxymethyl cellulose etc.) and natural (caseinates, whey proteins, lecithin, plant proteins, etc.) compounds can be used as food emulsifiers [6]. Among these, proteins are the best contenders for use in emulsion preparation due to their amphiphilic nature. In the present context, the most widely used protein-based emulsifiers are caseinates and whey proteins. Several studies have been conducted on oil-in-water emulsions prepared using caseinates, whey proteins or their mixture in the last few decades [7-10]. These emulsions have been successfully used to prepare dairy-based high-fat powder, such as cream powder. However, the research on applying plant proteins as an emulsifier for preparing plant-based high-fat powder is limited[11, 12]. While soy protein is abundant and possesses excellent emulsifying properties, the regulatory concerns over allergen status have necessitated researchers to find novel alternatives [13]. Mung bean protein, particularly its water-soluble fraction, has recently gained attention for its various techno-functional properties, including foaming, solubility, emulsifying capacity, and water and oil absorption capacity [14]. In the studies conducted by Brishti, Zarei et al. [15] and Du et al.[16], the emulsifying, water absorption and gelling properties of mung bean protein isolates were more desirable compared to other plant proteins. Furthermore, the bland taste and mild odour of mung bean protein make it more preferable when compared with soy protein [17].

The second step in encapsulation involves selecting the appropriate wall material and understanding its properties [18]. The wall material can be proteins and water-soluble carbohydrates such as maltodextrin and gums. The wall material used in high-fat powder

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embeds the fat globules in its continuous network. However, if the fat content in the emulsion is increased without increasing the wall materials proportionally, a thinner protective layer is formed around the fat globules. This results in an increase in free fat content in the encapsulated material (i.e. oil phase ) [10]. The free fat content is crucial for ensuring the functionalities of food powders, such as high-fat powder. In the studies conducted by Drusch and Berg [19] and Sarkar et al.[20], there was an increase in the surface fat content and number of fat globules close to the surface of the powder particles with an increase in total fat content in the powder. The free fat content in a spray-dried powder represents the surface fat, capillary fat, and outer layer fat that are easily extractable in the solvent [19]. The amount of free fat content influences the functional properties of the powder, such as oxidative stability, flowability, wettability, and reconstitutability [21]. Higher free fat content leads to lower shelf life because of oxidative rancidity [8].

Mohammed et al.[22] reported that high water-soluble and low viscous wall materials are ideal for the preparation of spray-dried powder [22]. Maltodextrin is such an example, which is an inexpensive, flavourless starch hydrolysate that can protect the encapsulated oil phase from oxidative degradation. Furthermore, it is highly soluble in cold water, which aids in the process of emulsification[22]. In a study conducted by Munin and Edwards-Lévy [23], high encapsulation efficiency of oil was achieved when a blend of maltodextrin and sodium caseinate was used as wall material. The ability of the wall material to keep the core material intact determines the encapsulation efficiency of the material. In general, the proportion of fat that cannot be extracted as free fat gives the encapsulation efficiency of the wall material in fat powders [24].

Powder properties are also affected by the melting point of the fat in the core. Fat powder containing encapsulated fat with a low or high melting point tends to have lower free fat content compared to fat with an intermediate melting point. The reason behind this is the

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presence of partially crystalline fat in the liquid oil phase. The partial crystalline phase tends to break the protective film that surrounds the partially liquid droplets, leading to fat leakage and poor encapsulation [10, 25, 26]. Furthermore, the encapsulation efficiency is also affected by the droplet size of the emulsion. Smaller droplet size in the emulsion generally leads to less free fat content in the powder or greater encapsulation efficiency. Upon decreasing the mean droplet size from 1.2 µm to 0.5 µm, a seven-fold decrease in surface fat content in the fat powder was observed by Danviriyakul et al.[27]. Similar results were reported by Jafari et al.[9] in the study of fish oil powders. The author reported a decrease in free oil content from 1.27% to 0.69% when the average fish oil droplet size in the emulsion was reduced to 0.28µm from 5.9µm [9].

While the market for plant-based food products is continuously increasing, there is a need for the development of plant-based high-fat powder that can replace dairy cream powder. Commercial dairy cream powder is known for better functionalities such as reconstitutibility and dispersibility. Numerous studies have been carried out on powdered emulsions; however, the materials (i.e. oil phase, emulsifiers, wall material) are either entirely animal-based or a combination of animal and plant-based, or plant-based with limited solubility or lower fat content [28],[29]. To the best of our knowledge, very limited studies have been done to date that produced plant-based spray-dried high-fat (>40% w/w) yet highly soluble powder. The product in this study is aimed to be an equivalent alternative to dairy cream powder. Therefore, this study investigated the various physicochemical properties of the high-fat powder prepared using only plant-based materials (i.e. oil phase, wall material and emulsifier).

## 2. Materials and methods

### *2.1. Materials*

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Corn oil (Reinna Brand) was purchased from a local supermarket in Perth, WA, Australia. Mung bean protein isolates were purchased from Bulk Powders (Australia), and maltodextrin (unflavored) was purchased from nutricost.com, USA. Analytical grade petroleum ether (BP= 40-60 °C) was purchased from Sigma-Aldrich, Bayswater, Victoria, Australia.

### *2.2. Methods*

#### *2.2.1 Experimental design*

Table 1 shows the formulation used for preparing oil powder containing 40% and 20% oil, and Table 2 shows the oil percent and the oil globule size for the four different samples prepared in this study.

**Table 1** Formulations used for the preparation of oil powder containing 40% and 20% oil.

| Ingredients | Weight (g) | Weight (g) |
|---|---|---|
| Corn Oil | 44 | 22 |
| Supernatant (containing 0.3% protein) | 250 | 250 |
| Maltodextrin | 67 | 89 |
| Addition of water to dissolve wall material (maltodextrin) | 150 | 150 |
| Final Fat percentage (%) in spray-dried powder | 40 | 20 |
| Total | 511 | 511 |
| Solids in total mix, including oil phase (%) | 23 | 23 |

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**Table 2**

Description of the spray-dried powder samples

| Total oil content (%) in spray-dried powder | Size of the fat globule in emulsion before spray drying (μm) | Sample Code |
|---|---|---|
| 20 | 0.47 | PS0.47-20 |
| 40 | 0.75 | PS0.75-40 |
| 20 | 0.75 | PS0.75-20 |
| 40 | 0.47 | PS0.47-20 |

Note: The average size of the oil droplets of the emulsion collected after atomization remained the same as before atomization.

*2.2.2. Sample preparation.*

*2.2.2.1. Mung bean protein isolate as emulsifier*

The emulsifier used in the study was prepared by modifying the method used by Wei et al.[30]. Four g of mung bean protein isolate (protein content, 85.35g/100g powder) was soaked in 296 g of deionized water. The soaked mung bean protein isolate was kept overnight in a refrigerator. It was then centrifuged at 4700 rpm (Eppendorf 5810 R, Germany) for 30 min, and the supernatant was carefully transferred into a beaker. It was analyzed for protein content using the standard Kjeldahl method using a 6.25 multiplication factor. Further details of protein content determination are not elaborated in this manuscript. The protein present in the supernatant was used to prepare the emulsion. The aim of this work was to study the effect of emulsion droplet size on the physicochemical properties of the high-fat powder. Only supernatant (completely soluble fraction) was used to nullify the effect of undissolved protein particles on the average size of the emulsion droplet while measuring via particle size analyzer. In our preliminary trials, we observed that it was impossible to completely dissolve the mung-

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bean protein powder particles, which were reflected in the particle size results.

*2.2.2.2. Emulsion preparation*

For emulsion preparation, the formulation in Table 1 was used. To prepare an oil-in-water emulsion (E1) for spray-dried powder containing 40 % (w/w) oil, 44 g of oil was mixed with 250 g of supernatant (0.3 %w/w protein content). The mixture was pre-emulsified for 5 min at 17500 rpm using an Ultra-turrax homogenizer (IKA T18 basic, Germany). Then, it was immediately homogenized using an ultrasonicator (VCX750, Sonics & Materials Inc., Newtown, USA) for 8 min and 16 min at 27°C to obtain two different fat droplet sizes (i.e. D[4,3] of 0.75 µm and 0.47 µm, respectively). Similarly, to prepare emulsion (E2) for spray-dried powder containing 20% (w/w) oil, 22 g of oil was mixed with 250 g of supernatant. Then Ultra-turrax homogenizer was used for 5 min at 17500 rpm and then ultrasonicated for 4 min and 12 min at 27°C to obtain particle sizes D [4,3] of 0.47 µm and 0.75 µm, respectively. Ultrasonication was carried out using the frequency of 20kHz, 600 W power, and 50% amplitude in continuous mode (without pulse) to prepare E1 and E2. The pre-emulsified sample was kept in an ice bath during the time of ultrasonication to prevent an excessive rise in temperature. The prepared emulsions were analysed for oil globule size and stored in refrigerated conditions.

*2.2.2.3. Wall material*

The wall material solutions were prepared by dissolving 67 g of maltodextrin in 150 g of deionized water for spray-dried powder containing 40% (w/w) oil and by dissolving 89 g of maltodextrin in 150 g of deionized water for spray-dried powder containing 20% (w/w) oil (Table 1).

*2.2.2.4. Spray drying of the emulsion*

The emulsion and wall material were mixed using an overhead stirrer at 150 rpm for 3 min. Then, it was spray dried using a spray dryer (Buchi Mini Spray Dryer B-290, Buchi Co.

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Switzerland). Feeding of the emulsion was done at 45 °C with a feeding rate of 5 mL/min through a 0.7 mm diameter two-fluid nozzle. The compressed air pressure going to the atomiser was set at 0.6 MPa. The inlet and outlet temperatures of the drying air were maintained at 170 °C and 70 °C, respectively. The powdered samples were collected from the cyclone separator and the collecting vessel by using a soft brush into airtight 70 mL sterile containers. The powder from the drying chamber was not used for analysis. The sample containers were kept in resealable bags and stored at refrigeration temperature until further analysis. Each batch of freshly prepared powdered samples was analysed within a week.

### *2.3. Product characterization*

#### *2.3.1. Size measurement*

The measurement of average oil globule size in the emulsion before spray drying was carried out using a particle size analyzer (Malvern Mastersizer 2000, Malvern Instruments Ltd. Worcestershire, UK). Deionized water was used as a dispersant, and the emulsion was added dropwise to the deionized water using a dropper until a laser obscuration of 10-10.5% was obtained. The refractive index values used for deionized water (dispersant) and oil (dispersing material) were 1.33 and 1.47, respectively. The absorption index was set at 0.01.

#### *2.3.2 Moisture content and water activity*

The moisture content of the oil powder was measured using the hot air oven method. The oil powder was heated at 105 °C in a hot air oven until a constant weight was reached. Water activity was measured using a water activity meter (AQUALAB 4, METER Group Inc., USA) using approximately 3 g of sample to fill the base of the sample holding cup.

#### *2.3.3 Free oil content*

Free oil content of the oil powder was determined by modifying the process

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described by Schuck et al.[31]. Briefly, 5±0.5 g of the oil powder was weighed in a conical flask, and 100 mL of petroleum spirit was added to the flask. The conical flask was then kept in an electric shaker for 15 min with gentle shaking, enough to ensure adequate mixing without creating high turbulence. The solution was then filtered into a pre-weighed dry round-bottom flask using Whatman filter paper 41. Then, 50 mL of petroleum spirit was poured into the residue in the conical flask and was filtered into the round-bottom flask. The solvent was evaporated off using the rotary vacuum evaporator at 60°C. Then, the round-bottom flask was dried in the hot air oven at 105°C for 1 h. The flask was then kept in a desiccator to cool down, and the dried weight was measured as extractable fat. The following equation was used to calculate the percentage of free oil content:

Free oil content (%) = (Extractable oil (g)/Total oil in powder (g)) ×100.

The residue was left to dry in the fume hood overnight and collected into an airtight container for the analysis of solvent-washed high-oil powder.

*2.4.4. Oil globule size of reconstituted powder*

The reconstitution of the spray-dried fat powder and the solvent-washed fat powder was carried out as described by Hogan et al.[32]. For the reconstitution, 0.5 g of the powder was dissolved in 150 mL of deionized water. The solution was gently stirred using an overhead stirrer for 30 min at room temperature (25-28 °C). Then the oil globule size was measured using Mastersizer as described in section 2.3.1. The oil globule size of the reconstituted spray-dried powder and solvent-washed reconstituted powder was compared with the mean oil globule size of the parent emulsion. Parent emulsion indicates the emulsion before spray drying.

*2.4.5. Angle of repose*

The angle of repose of the spray-dried powder and the solvent-washed powder was

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measured as described by Kim et al.[33]. The powder was poured through a funnel into a petri plate (diameter: 80 mm, radius: 40mm). The maximum height (mm) of the powder was recorded after it fully covered the base. It was then calculated using the following equation:

$$\theta = \tan^{-1} \frac{radius\ of\ Petri\ plate}{height}$$

*2.4.6. Bulk density*

Briefly, the fat powder was filled into a 10 mL measuring cylinder. The weight of the powder was then divided by the volume of the cylinder to calculate the bulk density [18].

*2.4.7. Confocal laser scanning microscopy (CLSM)*

The microstructure of the oil powder was visualized using a confocal laser scanning microscope (Nikon A1+, US). The oil powders were dyed with Rhodamine B for protein and Nile red for fat, 10 min before imaging. The dyes were prepared by dissolving 5 mg of each dye powder in 50 mL of polyethylene glycol 400. To stain the powder, 20 mg of each of the dyes was added to 0.1 g of powder and gently mixed. A thin layer of stained powder was placed on a slide and gently pressed with a coverslip. The powders were then observed with a 63X water-immersion objective. For the excitation of Nile Red and Rhodamine B, lasers with wavelengths 488 nm and 555 nm were used, respectively [10].

*2.4.8. Scanning electron microscopy (SEM)*

The surface morphology of the oil powders was studied using SEM (VEGA3 TESCAN, Brno, Czech Republic). To visualize the powder morphology, the samples were mounted on the SEM stubs, and a sputter was used to coat them with gold. To ensure the samples are placed securely in the stubs, double-sided adhesive tape was used as described by Shivakumar et al.[18]. The samples were then examined with 5000x

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magnification.

*2.5 Statistical Analysis*

Two-way analysis of variance with no blocking was carried out using GenStat (12th Edition, VSN International Ltd.); the significant difference was considered at a 5% level of significance. To determine whether the sample means were significantly different or not, LSD and interaction effects were obtained. All the analysis was done in triplicate.

## 3. Results and discussion

*3.1 Water activity and moisture content*

The mean water activity of the spray-dried oil powders was measured to be 0.13±0.01, 0.14±0.01, 0.15±0.01 and 0.16 ±0.01 for PS0.75-40, PS0.4-20, PS0.75-20 and PS0.47-40, respectively. There was no significant difference ($p>0.05$) in the values of water activity among the powders. Likewise, the mean moisture content of the powders was 4.48±0.24, 4.46±0.21, 4.82±0.22 and 4.52±0.01 % for PS0.75-40, PS0.47-40, PS0.75-20 and PS0.47-20, respectively. There was no significant difference ($p>0.05$) between the mean moisture content of the powdered samples. A similar result was also obtained by Dhungana et al. [10] in the spray-dried high-fat powder that was prepared from a cream emulsion. The authors also reported no significant difference in the mean moisture content and water activity of the powdered samples having a particle size ranging from 0.21 µm to 1.42 µm with fat content ranging from 35% to 75 %. Similarly, no difference in moisture content and water activity, even when the inlet temperature of the spray dryer varied between 150, 170 and 190 °C was observed by Himmetagaoglu and Erbay [34] in their study of fat powders. The drying conditions used for all the powders (i.e. PS0.47-20, PS0.47-40, PS0.75-20 and PS0.75-40) were the same, which could be the reason for no significant difference in the mean values of moisture content and water activity.

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*3.2 Bulk density*

The average bulk density of the oil powders was 0.236±0.004, 0.239±0.006, 0.246±0.005 and 0.252±0.005 g/cm$^3$ for oil powders PS0.75-40, PS0.47-40, PS0.75-20 and PS0.47-20, respectively. A significant effect ($p<0.05$) of oil content on the bulk density of the fat powder was observed when two-way ANOVA was carried out. There was an increase in the bulk density of the fat powder with a decrease in total fat content in the powder. A similar result was obtained by Zhang et al.[35], where the bulk density of soya bean oil powder increased from 0.769 g/cm$^3$ to 0.916 g/cm$^3$ when the oil content in the powder was decreased to 10% from 20%. Likewise, Domian et al.[36] also reported an increase in bulk density (0.282 g/cm$^3$ to 0.354 g/cm$^3$ ) of spray-dried emulsion when the fat content dropped (55% to 40%). This trend is primarily attributed to the higher density of maltodextrin compared to oil. Besides, the powders with higher fat content had characteristic dents, which resulted in lower bulk density [36]. The results obtained showed a direct relationship between the morphology of powders and different wall thicknesses. High-oil powders with low maltodextrin content led to thinner shell-wall formation around the oil droplets, resulting in powder particles with irregular shapes. These irregularly shaped powder particles required more space, leading to a lowered bulk density of the powder. On the other hand, oil powders with thicker shell-walls have relatively small and uniform sizes, which resulted in higher bulk density [35].

*3.3 Free fat content*

Based on total oil weight in powder, the powdered corn oil samples PS0.75-40, PS0.47-40, PS0.75-20 and PS0.47-20 had a mean free oil content of 29.51±0.35, 23.32±0.28, 22.33±0.37 and 19.60±0.20 %, respectively (Fig. 1). The results of ANOVA indicated a significant effect ($p<0.05$) of total oil content, oil globule size and their interaction term (oil globule size × total oil content) on the mean free oil content of the

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powder. There was a significant ($p<0.05$) decrease in average free oil content with the decrease in mean oil globule size of the initial emulsion and a drop in total oil content in the final powder. Similar results were obtained by Dhungana et al.[10] in spray-dried cream emulsions. The authors reported an increase in free fat content from 1.8% to 75.6% when the fat globule size of the parent cream emulsion was increased from 0.21 µm to 1.41 µm, and the total fat content in the powder was increased from 35% to 70%, respectively.

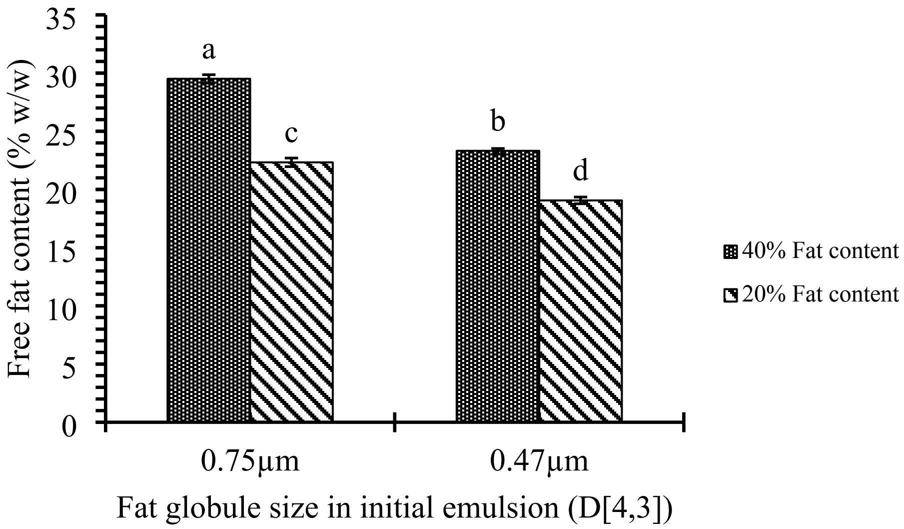


**Fig. 1.** Free fat content (g/100 g oil in powder or %w/w) of the spray-dried corn oil powder prepared from parent emulsion size with oil globule size of 0.75 µm and 0.47 µm.

The increase in surface oil content from 45.3% to 48.9% was also reported by Hogan et al.[32] when the mean droplet size of the oil emulsion was increased from 0.41 µm to 1.41 µm. The oil-to-sodium caseinate ratio in their experiment was 1. Similarly, in the study of spray-dried encapsulated fish oil powders in which whey protein concentrate was used as a stabilizer, Jafari et al.[9] observed an increase in free fat content from 690 mg/100g to 1270 mg/100 g when the mean droplet size of the fish oil emulsion was

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increased from 0.28 µm to 5.9 µm. Furthermore, an increase in free fat content with an increase in droplet size in the emulsion was also reported by Danviriyakul et al. [27]. The free fat content of the spray-dried milk fat prepared from the emulsion with a mean droplet size of 0.5 µm was 2%, whereas the milk fat powder prepared from the emulsion with a mean droplet size of 1.2 µm was 13.2%. The reason behind this could be because of the ability of the smaller fat globules to disperse more uniformly within the wall matrix during the process of spray drying. An even dispersion of fat globules within the wall matrix decreases the chances of being washed away during the process of solvent washing. Furthermore, the migration of larger fat globules to the outer part of the sprayed droplets is comparatively faster than that of smaller fat globules. Therefore, bigger fat globules tend to remain on the surface of the powder particles, which increases their chances of being easily washed away by the solvent [9]. Moreover, the larger fat globules are highly sensitive to mechanical stress and can easily break down during the process of spray drying. These ruptured fat globules remain on the surface of the powder and can easily get washed away with the solvent, resulting in higher free fat content [10].

The increase in free fat content with the increase in total fat in the final powder was also reported by Hogan et al.[32]. A drop in the surface oil content to 10.85% from 81.2% was reported by the authors when the ratio of oil to sodium caseinate was decreased from 3 to 0.25. Since, with an increase in total fat content, there is a decrease in the amount of wall material used (Table 1). This can lead to the formation of thinner interfacial membranes around the fat globules. This results in a higher chance of the fat globules being broken down and subsequently washed away during solvent extraction. Furthermore, an increase in the amount of wall materials helps keep the fat globules far away from each other, which ultimately decreases the chances of recoalescence and fusion of the fat globules. This can lead to lower free fat content since the fat globules

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are embedded and dispersed evenly within the wall matrix [10, 32].

*3.4 Angle of repose*

The angle of repose of the high-oil powders, PS0.75-40, PS0.47-40, PS0.75-20 and PS0.47-20 was 52.23±0.89°, 48.01±0.73°, 45.73±0.79° and 39.04±0.86° respectively (Fig. 2). Two-way analysis of variance indicated a significant effect ($p<0.05$) of total fat in powder, mean oil globule size in parent emulsion and their interaction (total fat × content fat globule size) on the angle of repose of the powder. An increase in mean oil globule size in the emulsion and an increase in total oil content in the powder resulted in an increase in the angle of repose of the high-oil powder. Similar results were obtained by Kim et al.[33] in dairy powders. The authors reported an increase in the angle of repose of the dairy powders (skim milk, whole milk, and cream) with different fat content. Furthermore, the authors also reported an increase in the angle of repose for each sample when the fat globule size was varied for each level of fat content. The angle of repose is an indication of the flow properties of the powders. A lower angle of repose indicates higher flowability of the powder and vice versa. The increase in the angle of repose could be due to an increase in the free oil content, which increased with an increase in oil globule size and oil content in the powder, that acts as a connecting bridge between the powder particles, leading to the clumping of the powder particles and decreasing their flowability [33].

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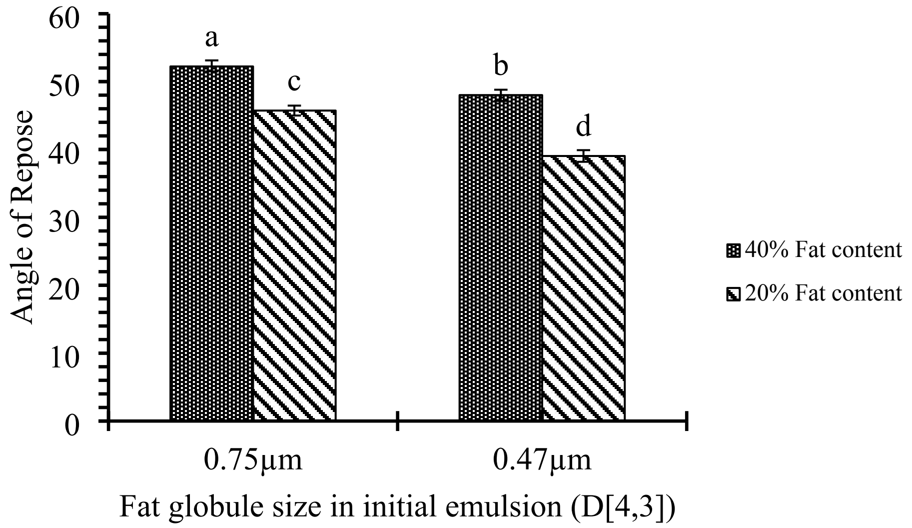


**Fig. 2.** Angle of repose of the spray-dried high-oil powders having two different fat contents (i.e. 40% and 20%) and two different oil droplet sizes (i.e. 0.75 µm and 0.47 µm) in the parent emulsion.

*3.5 Oil globule size of the reconstituted high-oil powder and solvent-washed reconstituted powder.*

The size distribution plots of the initial emulsion, after the addition of maltodextrin and after reconstitution of spray-dried powder and of the reconstituted emulsion prepared from solvent-washed powder are given in Fig.3. In all cases, the mean oil globule size of the corn oil emulsion remained almost the same after the addition of the maltodextrin. Maltodextrin was added after its solubilization. However, there was an increase in the mean oil globule size of the reconstituted powder samples. Similar results were reported by Ixtaina et al.[24]. The authors reported an increase in the mean fat globule size of the powder compared to the parent emulsion. Among the powders prepared from the same-sized parent emulsions, the extent of oil globule size increment in their reconstituted solutions was higher for the powder with higher oil content. This could be due to higher free oil on the powder with higher oil content, which was noticeable during size measurement as bigger oil globules

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(coalesced) upon reconstitution.

In the case of reconstituted emulsion prepared from solvent-washed powder, the mean droplet size for the oil powders prepared from the parent emulsion with a mean size of 0.47 µm, there was only a slight drop in the mean oil globule size (Fig. 3a & 3b). However, the mean oil globule size was lower for the reconstituted emulsion prepared from the parent emulsion with a mean size of 0.75 µm (Fig. 3c & 3d). A similar drop in the average fat globule size compared to reconstituted emulsion was also reported by Dhungana et al.[10]. This could be due to the presence of less stable free oil droplets on the surface of the powder prepared from larger fat globules in the parent emulsion.

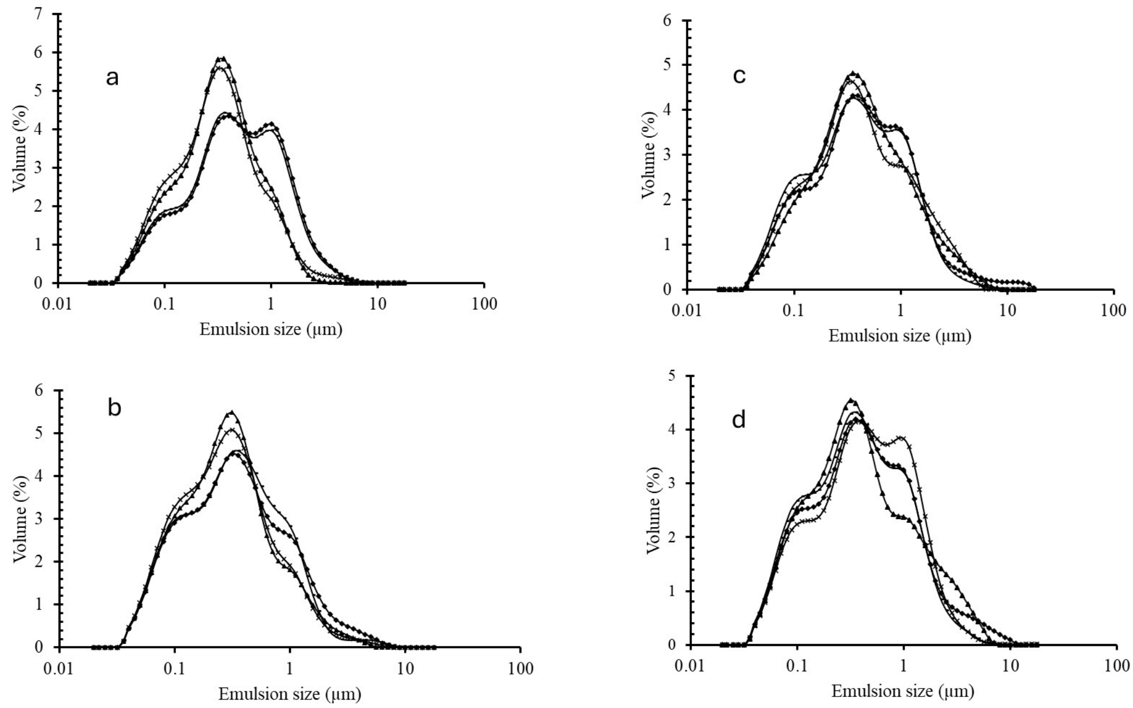


**Figure 3**. a) Oil globule size distribution plot (0.47µm and 40% oil); b) Oil globule size distribution plot (0.47µm and 20% oil); c) Oil globule size distribution plot (0.75µm and 40% oil); d) Oil globule size distribution plot (0.75µm and 20% oil). Initial emulsion ( ), emulsion after addition of maltodextrin ( ), reconstituted emulsion from high-oil powder ( ), reconstituted emulsion from solvent-washed powder ( )

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The plots in Fig. 3 show the change of oil globule size distribution from unimodal in the initial emulsion to multimodal or bimodal in the reconstituted powder, which could be due to coalescence that might have occurred during reconstitution [24, 32]. However, the oil globule size distribution of reconstituted powder is not significantly skewed to the larger globule size zone, as is visible in the plots. This indicates that the high-fat powder prepared for this study has very good reconstitutability. Ideally, if the powder surface is free of surface oil or if there is no breakage of oil globules during spray drying and storage, the reconstituted emulsion should have a similar oil globule size and distribution as the parent emulsion. This is the main achievement of this study, to produce a high-oil plant-based powder with better reconstitutability. The reconstitutability is one of the crucial powder properties that governs its functionality in product development. Similar fat globule size distribution plots were also obtained by Dhungana et al.[10] in the study of spray-dried high-fat powder prepared from the dairy cream emulsion.

### *3.6. Confocal laser scanning microscopy (CLSM) of the corn oil powders*

The CLSM images of high-oil powders and solvent-washed high-oil powders are given in Fig. 4. The green color in these images denotes oil. The images show a higher proportion of oil on the surface of the powder with the increase in the average fat globule size of the corn oil emulsion. Furthermore, on lowering the total oil content in the powder prepared from corn oil emulsion having the same mean oil globule size (0.47 μm and 0.75 μm), a decrease in the proportion of oil present on the surface of the oil powder was observed. The images obtained are comparable to those obtained by Dhungana et al.[10] in the study of cream powders and Kosasih et al.[37] in the study of spray-dried whole milk powders. The observations made through images are also supported by the values obtained for free oil content (i.e. the oil powder with the highest amount of free fat

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content has the highest proportion of surface oil in the image and vice versa) as presented in Fig. 1. The CLSM images (Fig 4e, 4f, 4g &4h) of the solvent-washed powder showed a decrease in the proportion of oil compared to their parent oil powders, which was due to the removal of free oil from the surface by the petroleum spirit. Moreover, the images of the solvent-washed powders showed an increase in the proportion of oil globules on the powder with the increase in total oil content in the powder (i.e. 40% fat). Such correlation between the CLSM image of high-oil powder and surface fat content indicates that CLSM imaging techniques can be utilized as a rapid quality control tool during the production of plant-based high-oil powder.

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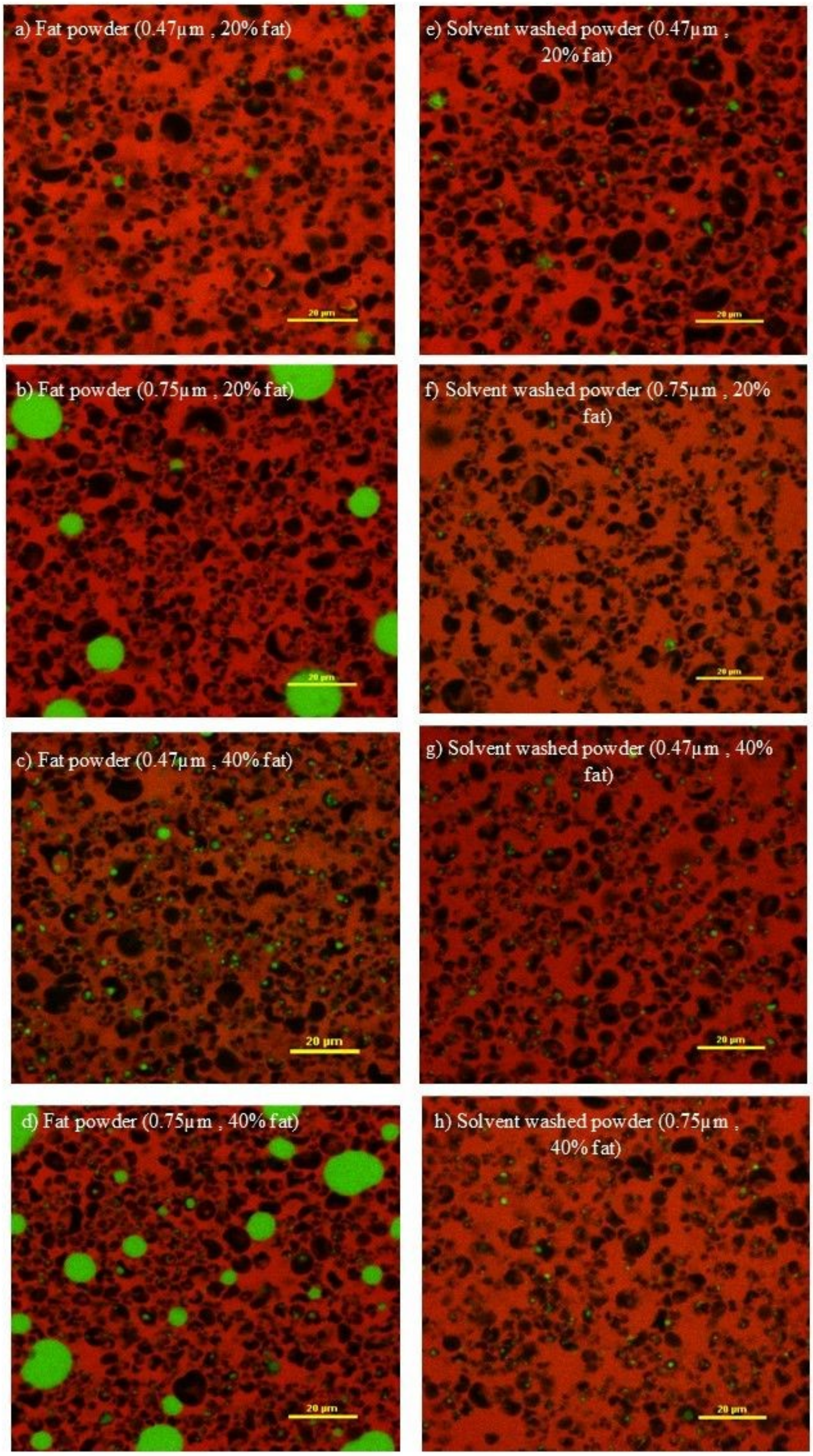


434

435 **Figure 4**. CLSM images of oil powders and solvent-washed high-oil powders. a) 20% fat
436 content powder from 0.47 µm parent emulsion, b) 20% fat content powder from 0.75 µm
437 parent emulsion, c) 40% fat content powder from 0.47 µm parent emulsion, d) 40% fat

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content powder from 0.75 µm parent emulsion, e) Solvent washed 20% fat content powder from 0.47 µm parent emulsion, f) Solvent washed 20% fat content powder from 0.75 µm parent emulsion, g) Solvent washed 40% fat content powder from 0.47 µm parent emulsion, h) Solvent washed 40% fat content powder from 0.75 µm parent emulsion. Green dots represent oil globules present on the powder surface.

### *3.7. Scanning electron microscopy (SEM) of corn oil powders*

The surface structure of the oil-encapsulated powders was studied using scanning electron microscopy (Fig. 5). The total field captured in each image is 50 µm. Overall, it is evident from the images (Fig. 5, a, b, c, &d) of samples without surface washing that there are lesser number of particle clusters. However,   Cluster formation in oil/fat encapsulated powder is primarily influenced by surface fat content in the powder [33]. In this present study,  based on powder weight ( in the section 3.3, free oil is expressed based on total oil weight in powder), the free oil content of the powder samples, PS0.75-40, PS0.47-40, PS0.75-20 and PS0.47-20 were 11.80±0.14, 9.32±0.12, 4.44±0.10 and 3.92±0.04 %, respectively. These free oil content values are relatively small and are reflected in the prevalence of a large proportion of individual particles (Fig. 5, a, b, c, &d) in bulk powder, which can further be correlated to the oil globule size distribution of emulsions before spray drying and after reconstitution of powder ( in section 3.5). Therefore, it is very clear that the preparation of plant-based high-oil containing powder with superior dispersibility and recosititutability is possible with the method reported in this study. There are very limited reports on production of such powder.

Further, the particles in high-oil powder having 20% oil content prepared from a parent emulsion size 0.47µm, had a comparatively rounder surface structure (Fig. 5a) than the other high-oil powders (Fig. 5 b, c & d), which is reflected as better flow properties or

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lower angle of repose. A rounder particle structure results in better flowability of the powders[38]. The flowability of powder significantly affects the ease of handling during industrial processes such as filling packs. In those images, it is evident that there is an increase in the number of dimpled powder particles with an increase in the average oil globule size of the parent emulsion. During spraying, the bigger oil globules tend to migrate to the surface of the spray droplets. Besides, the oil globules on the surface experience more physical stress during drying. As the bigger oil globules are sensitive to physical stress, they either buckle or rupture during drying. Furthermore, the increase in total oil content in the powder increased the number of buckled and dimpled particles. An increase in oil phase decreases the wall material-to-oil ratio, resulting in a thinner layer of wall material surrounding the oil globules. Such a condition also leads to collapsing, buckling and dimple formation, especially in larger oil droplets. Similar images were also obtained by Kim et al.[39] and Jones et al.[40] in spray-dried powders from dairy and soybean oil emulsion, respectively.

In addition, solvent washing of the plant-based high-oil powders disintegrated powder clusters to a greater extent in each formulation, an expected phenomenon. The disintegration of powder particles is because of the removal of surface oil that was acting as a connector in powder particle clusters. Removal of surface oil exposed buckled oil globules as well as dimples present in oil globules. These are visible in the SEM images of the solvent-washed powders in Fig. 5e, f, g, & h)

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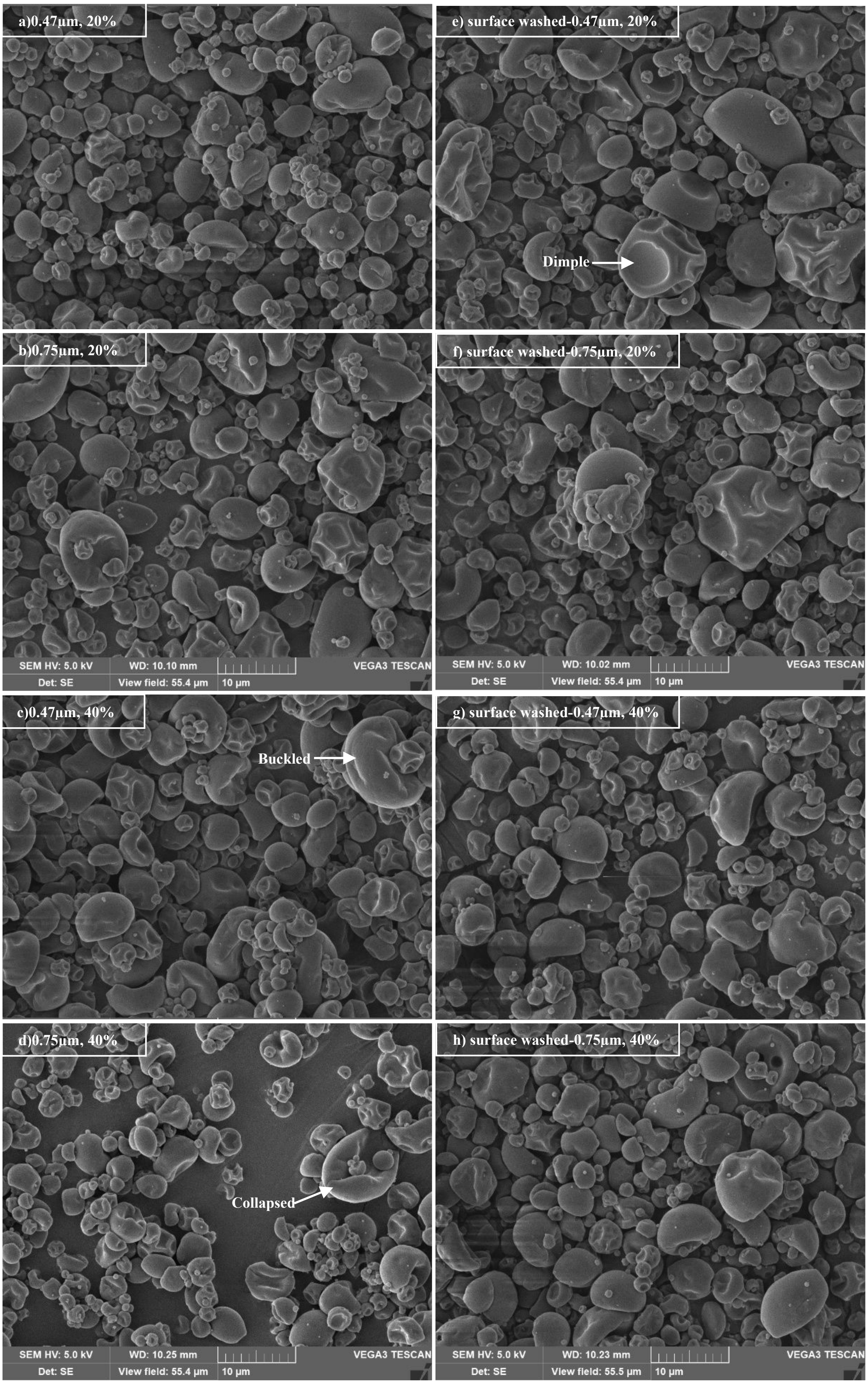
483 


484 **Figure 5.** SEM images of fat powders and solvent-washed fat powders. a) 20% fat content
485 powder from 0.47 µm parent emulsion, b) 20% fat content powder from 0.75 µm parent

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emulsion, c) 40% fat content powder from 0.47 μm parent emulsion, d) 40% fat content powder from 0.75 μm parent emulsion, e) Solvent washed 20% fat content powder from 0.47 μm parent emulsion, f) Solvent washed 20% fat content powder from 0.75 μm parent emulsion, g) Solvent washed 40% fat content powder from 0.47 μm parent emulsion, h) Solvent washed 40% fat content powder from 0.75 μm parent emulsion.

## 4. Conclusion

The present study demonstrated that a plant-based high fat powder can be prepared using all plant-based materials formulations. In this study, the soluble fraction of mung bean protein isolate (supernatant; 0.3% w/w protein) was used as the emulsifier. Corn oil was used as the oil phase, and maltodextrin was used as the wall material. The fat powders with an average fat globule size (D[4,3]) of 0.47 μm and 0.75μm and with a total fat content of 20% and 40% were prepared. Fat powder prepared from the corn oil emulsion with the lowest mean fat globule size i.e. (D[4,3]) of 0.47 μm and 20% total fat, was found to have better physicochemical properties compared to other formulations. This fat powder had the lowest angle of repose, better reconstitutability, higher bulk density, and lower free fat content. Furthermore, an increase in total fat content in the powder and mean fat globule size in the initial emulsion led to an increase in free fat content and angle of repose of the fat powders. These findings were further supported by the CLSM and SEM images. The results from the present study are useful in the development of plant-based food products as well as in our transition towards a sustainable future.

Although this study demonstrated that mung bean protein isolate/corn oil/maltodextrin could be used to prepare plant-based high-fat powders, further research on the other plant proteins, oils and wall materials can be explored.

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**Conflict of interest**

All authors declare no conflict of interest associated with this submission.

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**All the relevant data is already present in the manuscript. However, other supplementary data can be made available upon a valid request.**