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Microencapsulation of flaxseed oil in flaxseed protein and flaxseed gum complex coacervates

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ABSTRACT

Flaxseed oil, a rich source of omega-3 fatty acids, was microencapsulated in a novel matrix formed by complex coacervation between flaxseed protein isolate (FPI) and flaxseed gum (FG). This matrix was crosslinking with glutaraldehyde. Liquid microcapsules with three core (oil)-to-wall ratios (1:2, 1:3 and 1:4) were prepared and spray-dried or freeze-dried to produce powders. The microencapsulation efficiency, surface oil, morphology and oxidative stability of these microcapsules were determined. The spray-dried solid microcapsules had higher oil microencapsulation efficiency, lower surface oil content, smoother surface morphology and higher oxidation stability than the freeze-dried microcapsules. The highest microencapsulation efficiency obtained in spray-dried microcapsules was 87% with a surface oil of 2.78% at core-to-wall ratio 1:4 and oil load 20%. The oxidation stability obtained from spray-dried microcapsules at core-to-wall ratio of 1:4 was nearly double that of the unencapsulated flaxseed oil.

1. Introduction

Flaxseed oil (FO) has a higher percentage of alpha linolenic acid (ALA) than any other plant or marine oil, with ALA about 57% of total fatty acids (Carneiro, Tonon, Grosso, & Hubinger, 2013). However, like fish oil, its unsaturated nature renders it prone to oxidation, with subsequent loss of biological functionality. Microencapsulation has been applied by researchers to address this issue (Carneiro et al., 2013; Heinzelmann, Franke, Jensen, & Haahr, 2000; Liu, Low, & Nickerson, 2010). The process of complex coacervation followed by spray drying is recognized as one of the most promising technologies for stabilization of omega-3 oils by microencapsulation, while delivering a high payload (40–60%) (Barrow, Nolan, & Jin, 2007).

However, most of the microencapsulated products formed through complex coacervation use gelatin as the protein part of the wall material (Liu et al., 2010) rendering it unacceptable to the vegetarian population (Kralovec, Zhang, Zhang, & Barrow, 2012). There are also safety concerns associated with some gelatins, in particular beef gelatine, due to the potential for prion diseases (Morrison, Clark, Chen, Talashek, & Sworn, 1999). Moreover, there is increasing interest in industry for finding plant-based ingredients as encapsulating shell materials, due to their healthy image. Hence, it is necessary to find alternatives to gelatin as a shell material for microencapsulation.

In this context, the complex coacervates of plant-based proteins such as soy protein (Jun-xia, Hai-yan, & Jian, 2011), pea protein and cereal protein (Ducel, Richard, Saulnier, Popineau, & Boury, 2004), flaxseed protein (Wang, Wang, Li, Adhikari, & Shi, 2011) with gum Arabic have been explored for different active ingredients. Specifically for microencapsulation of FO, different wall materials used to date include whey protein isolate (Partanen et al., 2008), zein protein (Quispe-Condori, Saldaña, & Temelli, 2011), gum Arabic (Tonon, Grosso, & Hubinger, 2011), and different combinations of maltodextrin, whey protein concentrate and modified starch (Carneiro et al., 2013; Omar, Shan, Zou, Song, & Wang, 2009). Maltodextrins or starches act as good microencapsulating agents because they exhibit low viscosities at high solids contents and aid in spray drying of sticky materials (Adhikari, Howes, Bhandari, & Troung, 2004; Gharsallaoui, Roudaut, Chambin, Voilley, & Saurel, 2007). However, due to the lack of interfacial properties, maltodextrin and starches are generally associated with other microencapsulating materials such as proteins or gums (Gharsallaoui et al., 2007).

None of the published studies on the microencapsulation of FO have used flaxseed protein isolate (FPI) and flaxseed gum (FG) as wall materials, even though such an association should be considered natural. There are also some demonstrated health benefits, such as lowering of blood sugar, blood pressure and blood cholesterol, associated with the dietary consumption of FPI and FG (Doyen et al., 2014; Thakur, Mitra, Pal, & Rousseau, 2009). Hence, FPI and FG, which are emerging as potential emulsifiers (Oomah, 2001; Wang et al., 2011), should be considered for their efficacy in terms of payload, preventing oxidation as well as the structural strength of the coacervates formed.

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Considering the above points, the current study was undertaken with the following experimental steps: Firstly, FO was microencapsulated in crosslinked FPI–FG complex coacervates consolidated by maltodextrin at varying core-to-wall ratios. Secondly, FO liquid microcapsules were subjected to spray-drying or freeze-drying and finally, the dried microcapsules were characterized in terms of oxidative stability, microencapsulation efficiency, surface oil content and morphology.

2. Materials and methods

2.1. Materials

The golden flaxseeds (*Linum usitatissimum*) and flaxseed oil (FO) were received from Stoney Creek Oil Products Pty Ltd. (Talbot, VIC, Australia). Flaxseed gum (FG) and flaxseed protein isolate (FPI) were extracted in the laboratory as described in Section 2.2. All other chemicals were purchased from Sigma–Aldrich Australia (New South Wales, Australia) and were of analytical grade.

2.2. Proximate analysis

Proximate composition analyses for extracted FPI and FG were conducted according to the AOAC (Association of Official Analytical Chemists) official Methods 925.10 (moisture), 923.03 (ash), 920.87 (crude protein, by using %N × 6.25) and 920.85 (lipid) (AOAC, 2003). Carbohydrate content was determined on the basis of percent difference from 100%.

2.3. Extraction of FPI and FG

Extraction of FPI and FG was carried out using the method described in our previous work (Kaushik, Dowling, Barrow, & Adhikari, 2015). Briefly, flaxseed was soaked in Milli-Q water at a flaxseed-to-water ratio of 1:18 at 50 °C with continuous gentle stirring for 2 h. Subsequently, the seeds were filtered and the water containing the dissolved gum was treated with three volumes of 95% ethanol to precipitate the gum. The precipitated gum was collected by centrifugation at 4000 ×*g* for 10 min. The precipitated gum was vacuum dried at 50 °C and stored at 4 °C until further use.

Flaxseed protein was extracted from demucilaged flaxseed. The demucilaged seeds were dried in a hot air oven at 50 °C for 24 h and ground using a coffee grinder (EM0415, Sunbeam Corporation Ltd. NSW, Australia). The crushed meal was defatted for 3 h using hexane at a ratio of 1:6. The hull was separated from the kernel by screening the tailings using a 0.15 mm sieve to further reduce the interference of the mucilage during protein extraction. This defatted powder was subsequently soaked in 0.1 M tris buffer (pH 8.6 with 0.1 M NaCl) at a seed-to-buffer ratio of 1:16 for 24 h. The large residues were then separated from the protein extract using double layered cheesecloth. The filtered sample was centrifuged at 9000 ×*g* for 20 min using an ultracentrifuge (Sorvall Instruments, Wilmington, DE). The supernatant was collected and the pH was adjusted to 4.2 using 0.1 M HCl to precipitate the flaxseed protein. The extract was then stored at 4 °C for 16 h in order to provide sufficient time for protein to precipitate completely. The precipitated protein was recovered by centrifuging at 12,000 ×g for 20 min. The recovered solid mass was redispersed in Milli-Q water and was neutralized using 0.1 M NaOH. Finally, the FPI was obtained by freeze drying the sample at −50 °C condenser temperature and 0.04 mbar vacuum pressure using a freeze drier (DYNAVAC, Dynavac Engineering, Australia). The freeze-dried FPI was ground, vacuum sealed and stored at 4 °C.

2.4. Complex coacervation of FPI and FG for the microencapsulation of FO

Complex coacervation between FPI and FG was optimized at pH 3.1 and an FPI-to-FG ratio of 3:1 (Kaushik et al., 2015). Firstly, 250 g FPI solution (6%, w/w) and 250 g of FG solution (2% w/w) were prepared by dissolving the FPI and FG in distilled water at 50 °C for first two hours and then this was left to hydrate overnight at ambient temperature while stirring. Different amounts of FO (10.0 g, 6.66 g, and 5 g) were dispersed in the FPI solution to maintain a core-to-wall ratios of 1:2, 1:3, and 1:4, respectively. Subsequently, the first homogenization was carried out using an Ultra Turrax (RW 20, IKA GmbH Co., Bitterfeld-Wolfen, Germany) at 12,000 rpm for 5 min to produce an O/W emulsion. After that 250 g FG solution (2%, w/w) was added drop wise into this O/W emulsion with continuous stirring. A second homogenization was carried out at 18,000 rpm for 15 min. Since, FPI and FG were both extracted in the lab, the particle size was not as small as for commercial samples. Our preliminary experiments showed that for proper dissolution and mixing of FPI and FG, homogenization at each stage (one after adding protein and other one after addition of FG) works better. In addition, it reduces the size of emulsion droplets, as measured by a Zetasizer (data not presented here). The pH of this emulsion was adjusted to 3.1 by adding 0.1 M HCl drop wise to induce interaction between the FPI and FG. A microscope (Eclipse 80 i, Nikon, Japan) was used to obtain optical images of the coacervate microcapsules. The coacervation procedure was carried out at 50 °C for 1 h, followed by the crosslinking step. Crosslinking of the wall materials was carried out by addition of 4 g of 70% (w/w) glutaraldehyde solution (Devi & Maji, 2011). This comes out to be 2.8 mM of glutaraldehyde per g of wall material (FPI and FG). The crosslinking step was carried out after the coacervation step, at 50 °C for proper mixing of the glutaraldehyde in the dispersion. The liquid microcapsules thus formed were cooled to 5 °C at a slow rate of 5 °C/h using a programmable water bath (PolyScience, Niles, Illinois, USA). After maintaining, the sample at 5 °C for 4 h, 7.5 g maltodextrin (10 DE) was added to the liquid microcapsules before drying. The flaxseed protein (6% w/w) and gum solutions (2% w/w) are very viscous at the given concentrations and difficult to spray dry. Therefore, maltodextrin that has high solubility and low viscosity was added to the emulsion to help the spray drying of microcapsules (Adhikari et al., 2004; Gharsallaoui et al., 2007). The amount of maltodextrin (27% (w/w) of total wall material) was standardised through pre-experiments. Finally, the microcapsules were dried (Section 2.5) to produce solid microcapsules.

2.5. Drying of microencapsulated flaxseed oil

2.5.1. Spray drying

Microencapsulated FO produced as per Section 2.4 was spray-dried (Mini spray dryer B-290, BUCHI Labortechnik, Switzerland) using two a fluid nozzle, maintaining inlet and outlet temperatures of 190 °C and 95 ± 2 °C, respectively, with pump rate adjusted between 25 and 30% and air pressure at 35 m^3/h. These spray-dried microcapsules were collected and stored at 4 °C for further characterization.

2.5.2. Freeze drying

The freeze drying of the liquid microcapsules was carried out by first freezing them at −80 °C overnight. Frozen samples (<0.5 cm in thickness) were then freeze-dried (Christ Alpha 2-4LD, Osterode, Germany). The temperature of the ice condenser was set at −50 °C and the vacuum pressure was set to 0.04 mbar. The frozen samples were dried for approximately 36 h and the dried product was collected and stored at 4 °C for further characterization.

2.5.3. Solid yield

The solid yield was calculated to determine the losses incurred during spray drying and freeze drying of microcapsules. The solid yield was calculated as the ratio of the powder mass collected after every drying