International Food Research Journal 27(1): 78 - 87 (February 2020)
Journal homepage: *http://www.ifrj.upm.edu.my*

Effect of polymer and polymer blends on encapsulation efficiency of spray-dried microencapsulated flaxseed oil

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Article history

Received: 3 May 2019
Received in revised form: 19 December 2019
Accepted: 23 December 2019

Keywords

spray-dried microencapsulated flaxseed oil, inulin, SEM, XRD, thermal stability

Abstract

Flaxseed oil emulsions were prepared by homogenising flaxseed oil, gum Arabic (GA), maltodextrin (MD), and inulin (IN) in water (polymers were used alone and in combinations). Emulsions were analysed for their stability and viscosity. Results indicated that emulsions containing IN showed some layer separation while most of the emulsions were stable before spray-drying. The microparticles obtained after spray-drying were analysed for encapsulation efficiency, surface oil, density, flowing properties, moisture, water activity, colour parameters (L^*, $a^*$and b^*), particle size, morphology, dissolution behaviour, X-ray diffraction (XRD) pattern, and differential scanning colorimetery (DSC). All the encapsulated flaxseed oil powders (EFOPs) showed a bimodal particle size distribution with average particle size in the range of 1.18 to 9.80 µm. GA:MD (1:1) showed the highest encapsulation efficiency (92%) and lower surface oil. Scanning electron micrographs depicted spherical particles with no apparent cracks. IN yielded smooth-surfaced microparticles. DSC thermograms depicted that microcapsules were thermally stable. XRD analysis showed that most of the EFOPs were amorphous and spray-drying did not change the structure of wall materials. Dissolution behavior showed that presence of IN increased the solubility of EFOPs in water at room temperature. Hence, it is concluded that IN and MD effectively support GA in encapsulating flaxseed oil.

Introduction

Flaxseed (*Linum usitatissimum* L.) is a rich source of omega-3 (ω-3) fatty acids; 57% of its fatty acids is α-linolenic acid (ALA). This feature advocates its nutritional and health benefits (Naz et al., 2019). However, the incorporation of flaxseed oil into foods is a great challenge as they are prone to oxidative deterioration resulting in the development of flavours that decreases the quality of these oils (Augustin *et al.,* 2006) as well as certain volatile compounds and free radicals that pose toxic effects to the consumers.

Microencapsulation technology has several roles in food industry such as controlled delivery of lipophilic functional food ingredients and to supplement polyunsaturated fatty acids in foods (Drusch *et al.,* 2007). In this technique, the more sensitive component, i.e. the active ingredient, is contained within the micron-sized carrier matrices, which protect the active ingredient against the harsh external conditions (Reineccius, 2004). One of its important benefits is to protect sensitive oils against oxidation. Initially, an oil-in-water emulsion is prepared using any wall material and then dried to powder form. Spay-drying is the method used the most to make powders out of these oil emulsions (Desai and Park, 2005).

There are several substances used as encapsulants studied so far; however, gum Arabic (GA), due to its emulsifying properties, is the most popular and commonly used wall material for encapsulation by spray-drying (Jafari *et al.,* 2008). The presence of protein moiety in GA structure is responsible for its emulsifying properties. Due to the amphiphilic nature of protein, it gives better emulsifying and stabilisation to the emulsions. On the other hand, it has a limitation being a high cost wall material with limited availability. It is recommended to use GA with other materials or completely replace it with other ingredients for encapsulating oils or other substances. Maltodextrin (MD) has the advantage of

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being relatively inexpensive and provides good protection of the encapsulated materials. However, it cannot be used alone as a wall material for encapsulation since it is void of emulsifying characteristics. At the same time, it can be used as a wall material in blend with other materials to enhance the drying of microcapsules produced. Another interesting carbohydrate for encapsulation is inulin (IN), a polysaccharide composed of fructose units linked by β-(2,1) bonds and containing a glucose unit. IN can be commercially obtained from chicory and has prebiotic effects and dietary fibre actions, among other health related benefits (Ranawana, 2010).

In the present work, flaxseed oil was encapsulated using three different wall materials, alone and in combinations. The emulsions were characterised for their stability and viscosities followed by spray-drying of the emulsions. The obtained encapsulated flaxseed oil powders (EFOPs) were characterised for various analysis i.e. encapsulation efficiency, surface oil, bulk and tapped density, flowing properties, moisture, water activity, particle size and morphology, dissolution behaviour, X-ray diffraction (XRD) pattern, and differential scanning colorimetery (DSC).

Materials and methods

Materials

Flaxseed oil was purchased from a local market in Lincoln, NE, USA. MD (with 10 DE), GA, and IN was purchased from TIC Gums Inc. (White Marsh, MD, USA). Tween 80 was purchased from Sigma-Aldrich Co. (St. Louis, MO, USA). The composition of treatments is shown in Table 1.

Emulsion preparation and characterization

Solutions (30%, w/v) of the blends of GA, MD, and IN were dispersed in distilled water (25°C) and made up to 100 mL. About 6 mL (20% of the wall material used) of flaxseed oil was added to each mixture and emulsified in a shear homogeniser (Model AD500S-P, FoShan, GuangDong, China) for 5 min at 14,000 rpm to a complete dispersion. Two drops of Tween 80 were added during mixing to aid emulsification. The composition of different emulsions is given in Table 1.

Emulsion stability

To measure emulsion stability, the method of Kuhn and Cunha (2012) was adopted. Immediately after preparing emulsions, 10 mL of emulsion was poured into a graduated cylinder, sealed with parafilm and stored at room temperature. The height of the upper phase was measured after 24 h. The measurements were made in triplicate. Eq. 1 was used to measure the stability as percentage separation:

$$\% \text{ Separation} = (H_1/H_0) \times 100 \quad \text{(Eq. 1)}$$

where, H_0 = emulsion initial height, and H_1 = upper phase height.

Table 1. The composition of treatments.

Treatment	Inulin	Gum Arabic	Maltodextrin
EFOP1	100%	-	-
EFOP2	-	100%	-
EFOP3	-	-	100%
EFOP3	50%	50%	-
EFOP5	-	50%	50%
EFOP6	50%	-	50%
EFOP7	50%	25%	25%
EFOP8	25%	50%	25%
EFOP9	25%	25%	50%

Emulsion viscosity

The viscosity of emulsions was determined following İbanoğlu (2002) using a Brookfield Viscometer (RV DVII+Pro, Brookfield Engineering Laboratories, Middleboro, MA USA.). The viscosity of the emulsions (350 mL) was measured at 50 rpm using a spindle number 5 at room temperature after 1 min of shearing.

Spray-drying

The emulsion formulations were spray-dried using a laboratory scale two-fluid nozzle atomiser spray-dryer (Mini Spray Dryer B-290, BÜCHI Labortechnik AG, Flawil, Switzerland). Standard nozzle diameter of 0.7 mm was used with the main spray chamber of 500 × 215 mm. Emulsions were fed to the main spray chamber using a peristaltic pump while pump rotation speed controlled the feed flow rate. Feed flow rate was 12 mL/min. Drying air flow rate was 73 m3/h and the compressed air pressure was 0.06 MPa. Inlet and outlet temperature during the drying were 180 and 110°C, respectively.

Moisture content and water activity

The moisture content of EFOPs was measured by a Halogen moisture analyser (HB43-S, Mettler Toledo, Columbus, OH, USA). Water activity was measured using water activity analyser (AquaLab 4TE, METER Food, Pullman, WA, USA) at 35°C. All measurements were performed in triplicates.

Bulk density and tapped density

The samples (10 mg) were placed in a 50 mL graduated cylinder. The mass of powder was measured over a balance. The volume was noted directly from the cylinder. Then bulk density was calculated using the formula of density i.e. mass/volume. Tapped density was calculated by taking 5 mg of the sample in a 25 mL graduated cylinder. It was then gently tapped on the bench by raising almost 10 cm height. The volume was noted when there was no difference in the height of powder after successive tapping. This volume was then used to calculate tapped density by using the ratio of mass and volume.

Colour analysis

The L^*, a^*, b^* values of EFOPs were measured using a colorimeter, (Chroma Meter CR-400, Konica Minolta, Ramsey, NJ, USA). L^* is the lightness, while a^* and b^* represent the colours where $+a$ is redness, $-a$ is greenness, $+b$ is yellowness, and -b is blueness. Initially, it was standardised using pure white tile. The colour of each powder was measured in triplicates.

Particle size distribution

The particle size and size distribution of EFOPs were determined by a laser light diffraction instrument, Mastersizer 3000 (Malvern Instruments Ltd., Worcestershire, UK). Approximately 30 mg of the particles were mixed with 25 mL distilled water, followed by addition of 0.4% (w/w) polyoxyethylene sorbitan monooleate (Tween 80). The mixture was then placed for 30 min in an ultrasonic water bath (3510 R-MTH, Branson Ultrasonics Corporation, Danbury, CT, USA). The particle size distribution was monitored during each measurement until successive readings became constant.

Scanning electron microscopy

The microstructure of EFOPs was analysed by Scanning Electron Microscopy (SEM). For sample preparation, a small amount of each powder was placed on a double-sided sticky carbon tape adhered over SEM aluminium studs. Then, a blower was used to remove the excess powder, resulting in the formation of an even thinner layer. It was followed by sputter coating in argon atmosphere with chromium using HiPace 80 (Pfeiffer Vacuum, Germany). The morphology of microparticles was studied under Field Emission - Scanning Electron Microscope (FE-SEM) (S4700, Hitachi High-Technologies Corporation, 157 Japan).

Differential scanning calorimetry

The thermal characteristics of EFOPs were analysed by Differential Scanning Calorimetry (DSC) (Diamond DSC, Perkin Elmer, Waltham, Massachusetts, USA). The samples (~7 mg) were placed directly in the aluminium pans. An identical empty sealed pan was used as reference. Nitrogen was used as a carrier with a flow rate of 20 mL/min. Pans were heated at 25°C for 1 min and then samples were heated from 20 to 150°C at 5°C/min.

Dissolution behavior

The dissolution behavior of EFOPs was measured using a UV-VIS spectrophotometer (Evolution 201, Thermo Fisher Scientific, Shanghai, P.R. China) following the protocol of Millqvist-Fureby et al. (2001). 30 mg of microparticles was placed on the surface of 3 mL (approximately) water in a cuvette. After every 1 min, the absorbance was recorded at 620 nm until subsequent readings were constant.

XRD analysis

X-ray Diffraction (XRD) of microcapsules was performed using PANalytical Empyrean 187 Diffractometer (XRD) unit (Empyrean, PANalytical, Westborough, MA, USA). The powders were put in the sample slot and firmly pressed with glass cover. The excess powder was scraped off so that the powder surface was in plane with the glass shelf surface. The samples were placed on the instrument with a continuous scan speed of 0.02°/min, the patterns recorded over 2θ at angles 4 to 40° as described by Botrel *et al.* (2016).

Results and discussion

Emulsion stability

If an emulsion is stable, it will result in higher encapsulation efficiency and better protection to the core material. The emulsions were fed to spray-drying after 24 h; so, the stability of emulsions was critical during this period. In the present work, most of the emulsions prepared were stable when stored for 24 h at room temperature. There was no layer or separation seen in emulsions except for a few. However, it was observed that emulsions containing IN as a wall material were not completely stable as compared to GA and MD emulsions. Emulsions for EFOP1, EFOP4, and EFOP6 showed 10, 4 and 3% separation, respectively. There was either a phase separation or tiny oil droplets were observed on the surface of emulsion after storage period. It has been reported by several researches that these polymers had good emulsifying properties. As a result,

the emulsions prepared by using these polymers were stable emulsion systems. Turchiuli *et al.* (2014) reported that MD and IN were support materials. They are poor emulsifier agents, and would obtain stable fine emulsions required using GA for its emulsifying properties.

Emulsion viscosity

Viscosity is one of the most critical properties of any emulsion. It is directly proportional to total solids and inversely proportional to particle size. The emulsions showed a slightly pseudoplastic shear thinning behavior but still different depending on the polymers used in the aqueous phase. The highest value for emulsion viscosity was shown by EFOP2 which contained GA. GA finds its role in the food industry as a thickening agent. It shows a ramified structure with long chains and this property is responsible for the higher viscosity of emulsions containing GA. MD is a low cost hydrolysed starch. It has low viscosity even at higher concentrations and known for providing protection against oxidation, but it has a drawback of low emulsifying capacity. Preferentially, it is recommended to use in combination with other carrier agents to enhance its encapsulation properties (Bule e*t al.,* 2010). Turchiuli *et al.* (2014) reported that GA increased the viscosity of the emulsion whereas IN had a counter effect and decreased the viscosity.

Encapsulation efficiency and surface oil

Encapsulation efficiency (EE%) is one of the vital parameters for successful encapsulation of oil. The EE% and surface oil content of EFOPs varied from 45.06 to 92.85% and 0.031 to 0.073 mg/100 g powders, respectively (Figure 1). EE% was higher for microcapsules with GA (EFOP2: 88.85%) as compared to those which contained MD (EFOP3: 78.57%) and IN (EFOP1: 45%). The reason for the higher EE% of the GA microcapsules may be due to higher emulsion viscosity of this treatment, which promoted faster drying. Due to faster drying, the core material will not find enough time to escape from the encapsulating material and hence retained in the microparticles leading to higher EE%.

Generally, stable emulsion has better EE% (Barbosa *et al.,* 2005), which means the particle surface of the microcapsules had a lower proportion of non-encapsulated material. For EFOP6 (that contained IN:MD at 1:1), lowered EE% (48.7%) was observed as compared to EFOP3 (78.6%) that contained MD alone, which may be due to poor stability of the emulsions containing IN. The emulsion stability for treatments containing MD and IN was observed comparatively inferior, hence EE% can be correlated with emulsion stability. Similar findings were reported by other scientists. Fernandes *et al.* (2016) microencapsulated ginger essential oil through spray-drying and used GA, MD, and IN as wall materials. The EE% ranged between 48.0% (EFOP4) and 93.0% (EFOP5). Carneiro *et al.* (2013) found that the EE% for flaxseed oil was highest when GA:MD (80%) was used as the wall material instead of other treatments containing MD, whey protein (WP), and starches.

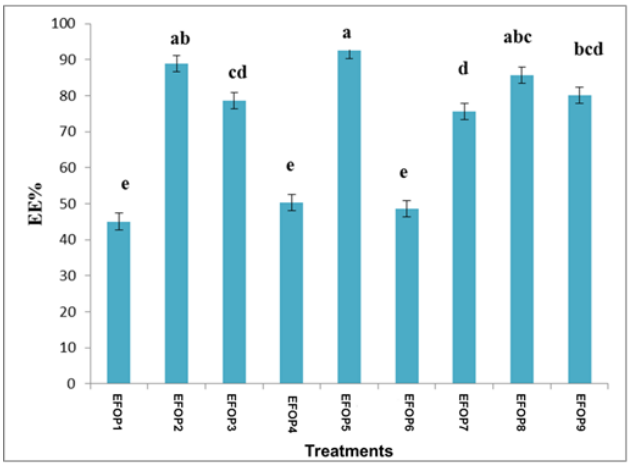

Figure 1. The encapsulation efficiency (EE%) of microparticles produced with different wall materials. Different letters indicate significant difference

Sansone *et al.* (2011) reported that MD lacks emulsifying property and so cannot be used alone as a wall material. But if some portion of carbohydrate is combined with MD, it can greatly enhance the drying of particles by readily forming a dry crust around the microparticles, filling the empty spaces and protecting against oxidation. The results of the present work showed that the treatments containing IN presented a lower EE% than the others, yet the presence of IN would be beneficial because of its prebiotic properties and attract the consumers seeking food with additional health benefits.

Bulk (ρB) and Tapped (ρT) density of EFOPs

Physical parameters such as bulk density, tapped density, and compressibility affect the powder's flowability and storage stability. Microcapsules showed significant variation in bulk density, based on the type of wall material used. Bulk density values ranged from 0.293 (EFOP1) to 0.487 g/cm^3 (EFOP2). It was noted that GA was the material that resulted in the densest particles (0.487 g/cm^3). Carneiro *et al.* (2013) reported that the bulk density ranged between 0.28 and 0.40 g/cm^3 for microencap

sulated flaxseed oil using GA, MD, modified starches, and whey protein isolates. Similarly; Tonon *et al.* (2012) observed the bulk density of microencapsulated flaxseed oil powder in the range of 0.174 to 0.350 g/cm^3 and 0.289 to 0.458 g/cm^3, respectively.

A high-density dry product can be stored in a smaller container, compared with a low-density product (Quispe-Condori *et al.,* 2011). The statistical data for tapped density on different EFOPs showed that it was significantly affected ($p < 0.05$) by the treatments. The tapped density was in the range of 0.642 to 0.433 g/cm^3. It was the highest for EFOP2 (0.642 g/cm^3) in which the wall material was GA alone while the lowest tapped density was observed for EFOP1 (0.433 g/cm^3) which was IN encapsulated flaxseed oil. After EFOP2, the higher density was observed in EFOP5 (0.616 g/cm^3), followed by EFOP3 (0.573 g/cm^3), EFOP8 (0.0.550 g/cm^3), EFOP9 (0.526 g/cm^3), EFOP4 (0.504 g/cm^3), EFOP7 (0.481 g/cm^3) and EFOP6 (0.475 g/cm^3). The treatments showing high tapped density gave an idea that a higher quantity of these powders can be stored in equal volume containers as compared to those powders which have low values for tapped density.

Flowing properties

Flowing characteristics of dry powders are generally assessed by Carr's index (% compressibility) and the Hausner ratio (HR) (Fitzpatrick, 2005). The higher the Carr's index, the more compressible the sample is, and the less free flowing the powder is. The borderline between free-flowing (granular) and non-free-flowing (powder) is about 20 - 25% compressibility (Quispe-Condori *et al.,* 2011). The statistical data for Carr's index on different EFOPs showed that it was significantly affected ($p < 0.05$) by the treatments. The Carr's index for different EFOPs preparations was in the range of 24.14 to 32.33, which is corresponding to the reference values of 21 - 25, 26 - 31 and 32 – 37, thus indicating the flow ability of the developed EFOPs ranged from "passable" to "very poor". The highest value of Carr's index was observed in EFOP1 (32.33) followed by EFOP6 (27.37) while the lowest value was observed in EFOP2 (24.14).

A higher HR indicates that the powder is more cohesive and less able to flow freely. A HR of more than 1.34 generally indicates the poor flowing characteristics of the dry powders (Turchiuli *et al.,* 2005). It is evident that the values of HR for different flaxseed oil powder were in the range of 1.32 to 1.48. Only EFOP1 showed "very poor" flowability and two treatments (EFOP6 and EFOP3) were "poor". Mostly the treatments (i.e. EFOP2, EFOP4, EFOP5, EFOP7, EFOP8, and EFOP9) were "passable" in terms of flowability. Our results agreed with Fernandes *et al.* (2013), who reported poor flowing characteristics of rosemary essential oil powders with high value of HR (1.30 - 1.67).

Moisture content

The moisture content of powder products is a crucial factor in determining their shelf life. Normally a moisture content of 3 to 4% is permissible in the food industry for powders (Gallardo *et al.,* 2013). In the present work, the moisture content varied from 2 to 4.5% for different formulations. EFOP2 that contained 100% GA showed the highest value (i.e. 4.5%). This is probably because GA contains more hydrophilic regions, which increases its moisture content. Those treatments which had a lesser share of GA in their wall material composition had less moisture when compared with EFOP2. The moisture content for different EFOPs was in the order of EFOP2 (4.5%) > EFOP5 (4.0%) EFOP3 (3.5%) > EFOP4 (2.8%) > EFOP7 (2.6%) > EFOP6 (2.4%) > EFOP8 (2.3%) > EFOP9 (2.0%) > EFOP1 (1.4%). Aghbashlo *et al.* (2013) also reported similar values for moisture content i.e. 1.41 to 4.36% in fish oil encapsulated particles. Similarly, Quispe-Condori *et al.* (2011) reported 3.88 to 5.06% moisture content in microencapsulated flaxseed oil. However, the moisture content was within the range that is considered as safe against microbial attack and the development of product alteration.

Water activity (a_w)

Generally, the water activity (aw) in dried foods is between 0.2 and 0.3, which is associated with the low levels of lipid oxidation (Polavarapu *et al.,* 2011). So, dried food products/powders should have aw within this range, otherwise there will be risks of auto-oxidation of lipids present in these powders. In the present work, aw was in the range of 0.19 to 0.32 for different EFOP treatments. The lowest value was recorded for EFOP1 (100% IN) (i.e. 0.19). The aw for different EFOPs was in the order of EFOP2 (0.32) > EFOP8 (0.31) EFOP5 (0.29) > EFOP9 (0.25) > EFOP4 (0.22) > EFOP7 (0.21) > EFOP6 (0.20) EFOP3 (0.20) > EFOP1 (0.19). The lowest values observed in EFOP1 might be due to many fructose subunits with hydroxyl groups (–OH) present in the IN structure as they are able to bind the water within the foods. Generally, those food systems which contain carbohydrates have low a_w. They can control a_w due to many –OH forming hydrogen bonds with the water. This binding

capacity is influenced by the structure of a compound, thus there was a variation in the a_w of powders. Bustos-Garza *et al.* (2013) encapsulated astaxanthin oleoresin by spray-drying using GA, MD, IN, and WP as wall materials. They reported a_w between 0.27 and 0.35. Tontul and Topuz (2014) reported a_w in the range of 0.088 to 0.213 in flaxseed oil microencapsulated powders.

Colour (L, a*, and b* values) of EFOPs*

The nature of ingredients in any formulation has a direct effect on the colour of the food product. The value of *L** varied between 93.11 and 90.57. The highest value was shown by EFOP3 (93.11). In this treatment, the wall material was only MD. The lowest *L** was observed for EFOP4 in which the wall material was IN/GA (1:1). Our results were slightly different from the observations reported by Karaca *et al*. (2013), who found *L** values ranging from 87.3 to 90.6, *a** values from -0.5 to 0.3 and *b** values from 11.2 to 20.3 for microencapsulated flaxseed oil employing chickpea protein isolates. The possible justification may be the different wall materials used in these studies. The results were in accordance with findings of Mohd Nawi *et al.* (2015). They encapsulated the anthocyanin in MD, GA, and their mixture and reported that the highest value of lightness was in the treatment containing MD as wall material, followed by their mixture. The *b** value showed the yellowness and was used as an indicator of the flaxseed surface oil. The highest *b** value was found in EFOP7 at 10.42. This treatment contained IN/GA/MD (2/1/1) as wall material. IN had lower encapsulation efficiency (45.1%) as compared to GA and MD, probably due to poor emulsifying properties, thus resulting in more surface oil. Therefore, higher *b** was recorded in this treatment.

Particle size distribution and average particle size of EFOP

Particle size of microcapsules can affect the textural and sensory properties of food products in which they are used. There was nearly a bimodal and almost similarly polydispersed distribution in all the formulations. The particle size distribution showed a wide range from 2.56 to 18.9 μm for different formulations. These variations were due to the aggregation of smaller particles around the larger ones. Gharsallaoui *et al.* (2007) reported that spray-drying yielded a very fine powder (10 - 50 μm) or large-sized particles (2 - 3 mm), depending upon the nature of feed emulsions. The results of the present work are in close comparison with the verdict of Carneiro et al. (2013) who microencapsulated flaxseed oil using different combinations of MD, GA, whey protein concentrate, and two different modified starches. Their findings delineated that the mean particle size varied from 17.98 to 23.03 μm. They further elaborated that greater size was observed by the particles obtained from mixture of MD and GA due to higher emulsion viscosity. Senatore *et al.* (2010) reported that the average diameter of spray-dried microencapsulated epoxidised linseed oil was 16 μm.

Morphology of EFOPs by Scanning Electron Microscopy (SEM)

The structure of spray-dried particles can be affected by different factors such as processing conditions which can influence shape and size. In the present work, SEM was used to analyse the microstructure of various EFOPs. The micrographs indicated that the encapsulation and spray-drying produced mostly spherical-shaped particles with several sizes. There were apparently no considerable fractured particles; however, slight agglomeration was observed along with apparent dents on the surface of particles. Intact particles guarantee better protection and retention of the core material since there are less chances of permeability of air/gasses. Also due to intact surface morphology, the encapsulation efficiency is high as the core material did not move out of the particle. However, the surface smoothness of microparticles cannot ensure better encapsulation efficiency. Hence, although the microparticles produced using IN in the present work had smoother surfaces, they had lower encapsulation efficiencies. A broad particle size distribution is a characteristic of spray-dried particles. At the same time, it was observed that wall material and the combinations affected the morphology of the particles (Figure 2). The particles produced from emulsion that contained IN were smoother as compared to others and had less dents (teeth), whereas the GA-containing particles showed more dents and irregular surface morphology. A similar trend of morphological characteristics was observed by Tonon *et al.* (2012). They also encapsulated flaxseed oil in GA and demarcated that particles were intact but showed concave and shrivelled surfaces, and this resulted from the formation of a vacuole and the immediate drying of particles. Senatore *et al.* (2010) also reported the agglomeration of spray-dried particles. Another reason for the agglomeration might be due to the presence of surface oil and the absorption and/or presence of moisture (Shivakumar *et al.,* 2012).

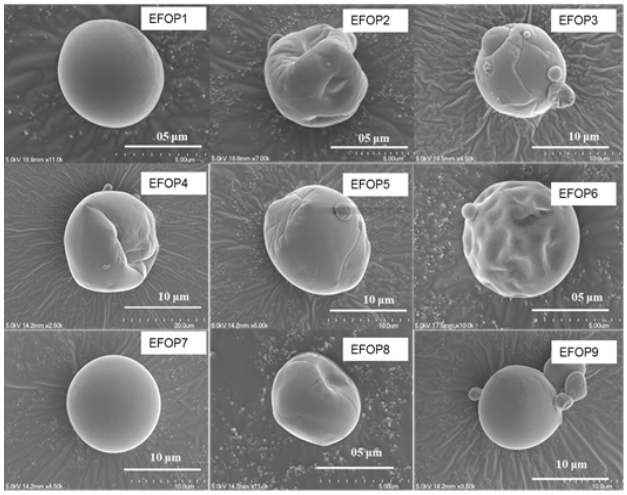

Figure 2. Morphology of particle surface of EFOPs as observed by scanning electron microscope.

Dissolution behaviour

One of the most vital properties of microcapsules is the efficiency and the speed of powder to dissolve/disperse in water (Klinkesorn *et al.,* 2005). The presence of surface fat or oil may negatively affect the dissolution behaviour (Kwapińska and Zbiciński, 2005). The dissolution profile of different EFOPs at 30 ± 1°C is shown in Figure 3. It was evident that the various EFOPs revealed a different dissolution behavior. However, all the formulated powder treatments were dissolved completely in water within less than 20 min. The treatment EFOP2 showed the lowest dissolution rate while EFOP1 showed the highest dissolution rate followed by EFOP6. EFOP2 contained GA as the wall material while IN and IN:MD (1:1) was the wall material in EFOP1 and EFOP6. The observed difference might be due to the different particle size of various EFOPs. There was an inverse relationship between the particle size and dissolution rate. The d_{90} (90% of the particles) was 22.2, 15.2, and 9.37 µm for EFOP1, EFOP6, and EFOP2, respectively. The larger particle size resulted in less compact packing of the powder and more free spaces in between the larger particles. Water can then easily penetrate the free space and increase the dissolution rate of the powder (Goyal *et al.,* 2014). Ferrari *et al.* (2012) found an inverse relationship between particle size and wettability of encapsulated blackberry oil powders. The dissolution was 134, 82, and 116 s for GA (11 µm), MD (49 µm), and their mixture (28 µm), respectively. Similarly, Goyal *et al.* (2014) explicated that the higher particle size and higher surface oil in NaCas flaxseed oil particles was one of the reasons for their slow dissolution rate.

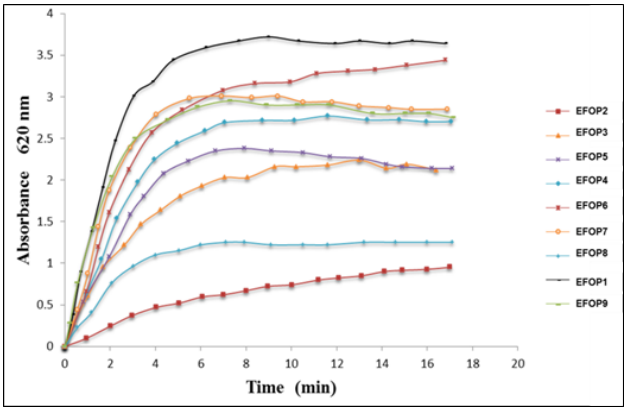

Figure 3. Dissolution behaviour of various EFOPs.

X-ray diffraction analysis of EFOPs

In general, if the XRD pattern shows several sharp peaks, it is recognised as a crystalline material whereas if the pattern is a broad background, it is considered as an amorphous material (Caparino *et al.,* 2012). The XRD patterns of raw materials are shown in Figure 4. It can be inferred from the Figure that all the polymers were amorphous in nature.

Figure 4 also represents the XRD patterns of EFOPs which were produced by spray-drying. The diffraction patterns obtained for the EFOPs are predominantly related to an amorphous material. It was clear that the wall materials before spray-drying and the EFOPs obtained after spray-drying had almost similar patterns. Bhandari and Hartel (2005) reported that spray-drying has less influence on the structure of biopolymers, since there is a quick drying of emulsions in this process, so molecules did not find enough time to crystallise. Similarly, in our XRD patterns, there were small changes between the raw materials before spray-drying and microparticles generated after spray-drying. The production of amorphous powders is desirable since they are more soluble (Drusch *et al.,* 2006). It can be concluded that EFOPs produced from GA, IN, and MD had favourable dissolution, since all the particles were completely dissolved in less than 20 min. Fernandes *et al.* (2016) encapsulated ginger essential oil in blends of MD, IN, and whey protein isolate and reported that all wall materials were amorphous before and after spray-drying. Campelo *et al.* (2017) developed lime essential oil microcapsules using prebiotic matrices (IN, oligofructose, and whey protein isolate). Their findings regarding XRD-difractograms showed that microcapsules were of amorphous nature.

DSC analysis

Thermal behavior of microcapsules was studied using DSC analysis in order to determine

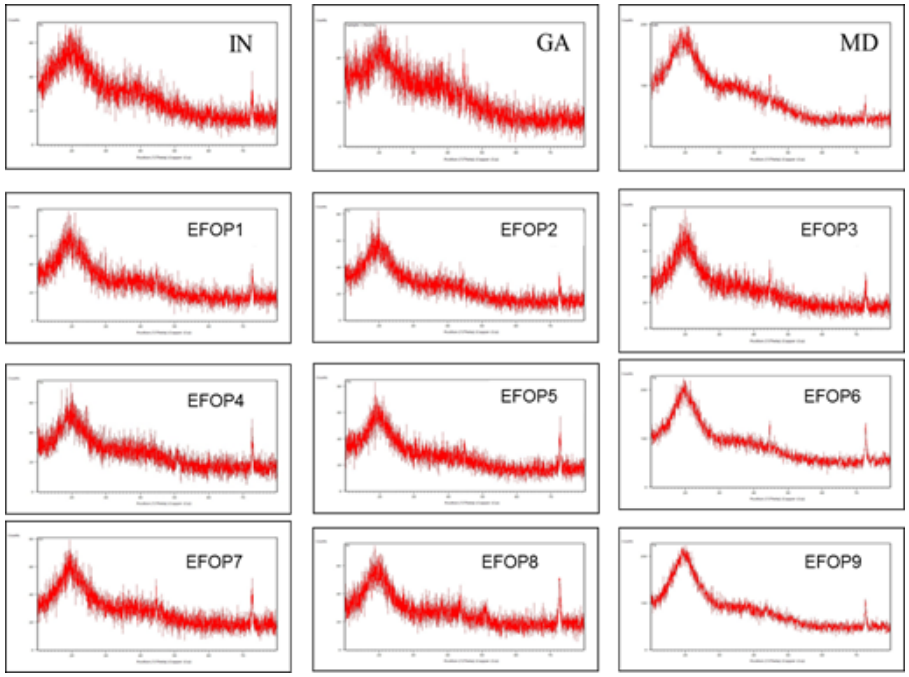

Figure 4. XRD difractograms of various EFOPs.

their storage stability. This method gives information about the physical and chemical alterations that can occur as a result of heat. DSC thermograms (Figure 5) showed that all the samples had exothermic peaks ranging from 60.05 to 66.21°C, at which the wall materials underwent thermal degradation. The presence of only one major peak in all the treatments indicated that encapsulating materials were homogeneously mixed. The peaks less than room temperature also showed that all the microcapsules were stable at room temperature.

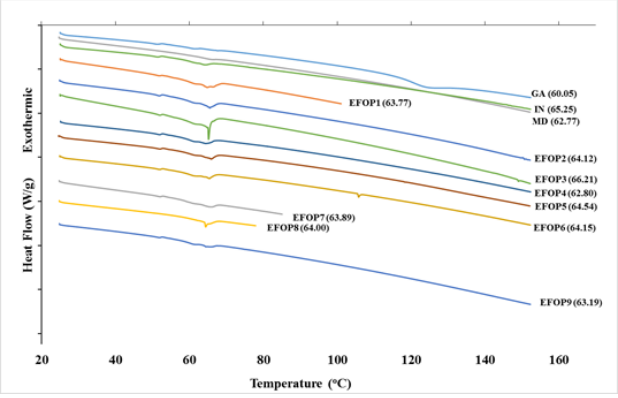

Figure 5. Thermal behaviour of various EFOPs as studied by DSC analysis.

Conclusion

In the present work, the performance of three polymers (GA, MD, and IN) for flaxseed oil microencapsulation by spray-drying was evaluated. The obtained microparticles were characterised and found to be different according to the nature and proportion of different wall materials. EFOP5 (containing GA and MD at 1:1) showed the best encapsulation efficiency (92%) result; but, the EFOP8 (containing GA, MD, and IN at 2:1:1) had a comparable EE (85%) with that of EFOP5. The results for encapsulation efficiency also showed that GA increased the emulsifying property of MD and IN. GA, being expensive, can be replaced with MD and IN for the encapsulation purposes as the objective of the present work was to develop a better combination of three polymers for flaxseed oil microencapsulation. Furthermore, IN will be beneficial owing to its prebiotic properties.

Acknowledgement

The authors are thankful for the funding provided by the Higher Education Commission of Pakistan through the International Research Support Initiative Program (IRSIP 33 Agri 24), thus enabling the present work to be carried out at the Department of Food Science and Technology, University of Nebraska-Lincoln, USA. The authors would like to thank Dr. Y. Joe Zhou at Microscopy Core Research Facility of the University of Nebraska-Lincoln, USA, for their help in scanning electron microscope imaging.

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