AACL BIOFLUX
Aquaculture, Aquarium, Conservation & Legislation
International Journal of the Bioflux Society

Characterization of dried fish oil from Menhaden encapsulated by spray drying

[1]Bahareh Mehrad, [1]Bahareh Shabanpour, [2]Seyed M. Jafari,
[1]Parastoo Pourashouri

[1] Faculty of Seafood Technology, Gorgan University of Agriculture Science and Natural Resources, Gorgan, Iran; [2] Faculty of Food Science, Gorgan University of Agriculture Science and Natural Resourses, Gorgan, Iran. Corresponding author: B. Mehrad, Bahar.mehrad@gmail.com

Abstract. This study aimed to evaluating the phisycochemical characterization of encapsulated fish oil from menhaden and potential of maltodextrin in combination with different wall materials in its microencapsulation process by spray drying, in order to maximize encapsulation efficiency. Maltodextrin (MD) mixed with fish gelatin (FG), κ carrageenan (κc) and both of them. The feed emulsions used for particle production were characterized for stability. The best encapsulation efficiency was obtained for MD:FG followed by the MD:FG+κc combination, while the lowest encapsulation efficiency was obtained for MD:κc, which also showed poorer emulsion stability. Particles were hollow, with the active material embedded in the wall material matrix, and had no apparent cracks or fissures.
Key Words: fish oil, menhaden, spray drying, κ Carrageenan, fish gelatin.

Introduction. Functional food including n-3 lipids is one of the fastest growing food product groups in the US and Europe (Frost and Sullivan Research Service 2005). Mainly, marine lipids have received growing notice for the past decade because of their useful health properties of eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA) on diseases like cardiovascular diseases (Yaqoob 2004), rheumatoid arthritis (Kremer 2000) and Crohn's disease (Belluzzi et al 2000). The best way to secure an increased intake of these healthy fatty acids in the people is through increase dietary fish consumption or partakes of fish oil capsule. An alternative way of increasing the intake of EPA and DHA could be via incorporation of fish oil into food products by substituting vegetable or animal fat into products such as salad dressing, milk and yogurt (Let et al 2005, 2007).

Due to their unsaturated character, n-3 polyunsaturated fatty acids (PUFA) are highly prone to accelerated oxidative rancidity. Lipid oxidation causing formation of undesirable off-flavours (e.g. fishy and rancid off-flavour) and unhealthy compounds such as free radicals and reactive aldehydes (Let et al 2003). Therefore, for successful development of PUFA enriched food, preventing the happening of the lipid oxidation is important.

Microencapsulating the oil with different biopolymers could be a very good way to overcome these adverse features (Cho et al 2003; Díaz-Rojas et al 2004). This structure could serve as: (a) vehicle for carrying the useful component to the preferred place action; (b) protection of the functional component from chemical or biological degradation (e.g. oxidation); (c) masking of the undesired properties of active component (e.g. odour and taste); and (d) controlling the release of the functional ingredient (Weiss et al 2006). This technology has been extensively used in pharmaceutical as well as in food industry.

Choosing the best wall materials and encapsulation technique are important steps in food encapsulation. Previous researches have emphasized that the best way to emulsify fish oil is to use a kind of wall materials, which could function, as a carrier matrix and as an emulsifier (Sheu & Rosenberg 1998). Even the best mixture of

biopolymers for encapsulating fish oil used with different encapsulating methods can create both stable and unstable powders.

Hence, in this research, three biopolymers (maltodextrin (MD), Gelatin from cold-water fish skin (FG) and κ-carrageenan (κc)) were evaluated in combination as wall materials for fish oil (fish oil from menhaden - which was bought from Sigma-Aldrich).

Maltodextrin is a filler matrix (Rosenberg et al 1993), which is cheap, extremely soluble in water and able to produce stable emulsion (Anandaraman & Reineccius 1986). Gelatin from marine sources (warm-water and cold-water fish skins, bones, and fins) is a potential substitute for bovine gelatin (Kim & Mendis 2006; Rustad 2003). One of the most important benefits of gelatin from marine sources is that they do not have the risk of occurrence of Bovine Spongiform Encephalopathy. Fish gelatin is acceptable for Islam, and can be used with least limitations in Judaism and Hinduism. Carrageenan is linear water-soluble sulfated polysaccharides that extracted from red seaweeds. Because of their biocompatibility and ability to produce thermoreversible hydrogels, carrageenan has been widely used as gelling agent in food and pharmaceutical industries (Stephen et al 1995). Within the carrageenan family, κ carrageenan originates the strongest gels and, hence, in the last decade this biopolymer has been studied a lot as a carrier for controlled drug release (Daniel-da-Silva et al 2011; Keppeler et al 2009; Leong et al 2011; Santo et al 2009).

Spray drying (SD) is one of many standard methods to encapsulate food ingredients. SD is a common method used for encapsulation of food components (Desobry et al 1997). It is an easy and low-cost technique in which either proteins or polysaccharides or a mixture of both can be used to create the wall for microcapsule. However, SD has some disadvantages, first disadvantage is high temperature for drying (sometimes higher than 200°C) and second one is only appropriate for matrices that are extremely soluble in water (Gharsallaoui et al 2007). Spray drying an emulsion that includes sensitive components such as fish oil is risky. Due to the high drying temperature used in spray drying, deterioration of sensitive components because of oxidation has been reported (Hogan et al 2003; Kolanowski et al 2006).

The main objective of this study was to investigate the effect of wall materials types (fish gelatin, κ carrageenan, maltodextrin and the combination of fish gelatin and κ carrageenan) on microencapsulation of fish oil using spray drying. The outcomes are compared based on encapsulation efficiency and powders physicochemical properties by related examinations.

Material and Method. The experiments were conducted between November 2013 to September 2014 in Gorgan University of Agricultural Science and Natural Resources labs. The materials were obtained from the following sources: fish oil (from menhaden), κ-carrageenan, gelatin (from cold water fish skin), acetic acid and sodium hydroxide purchased from Sigma-Aldrich Inc., St Louis, MO, USA. *n*-hexane (95%), isooctane, methanol and sulfuric acid were purchased from Dr. Mojallali Inc., Tehran, Iran. All chemicals used in this study were of analytical grade. Purified water was used for the preparation of all solution. All experiments and analysis were carried out in triplicate.

Emulsion preparation. Wall materials ratio including FG and κc, maltodextrin as filler matrix and soy lecithin as emulsifier are summarized in Table 1. Coating materials were dissolved in distilled water, followed by gentle stirring with a magnetic stirrer (for 30 min) to achieve a homogenous shell solution. The solution was allowed to hydrate for 24 h before emulsion preparation to ensure a full dissolution of materials, followed by cooling down to room temperature. The wall material concentration was 20% (Fernandes et al 2013a, b) and the amount of fish oil used was 25% of the mass of the wall materials (Jafari et al 2008). Coarse emulsions were prepared using an Ultraturrax IKA T25 homogeniser (Germany) at 12,500 rpm for 5 min. There was no addition of antioxidant during preparation of emulsion.

Table 1

Composition of the wall materials for the each treatment used as a feed solution for the spray-drying process

#	*Wall material (g 100 g^{-1} of solution)*				*Coar material - fish oil (g 100 g^{-1} of solution)*
	FG	*Kc*	*Maltodextrin*	*Soy lecithin*	
1	7.5	-	32.5	-	10
2	-	2.5	37	0.5	10
3	7.5	2.5	29.5	0.5	10

Emulsion characterization

Creaming stability measurement. Ten grams of emulsion were transferred into a test tube (internal diameter 15 mm, height 125 mm) and then stored for 1 month at room temperature. After storage, some emulsions separated into an optically opaque "cream" layer at the top and a transparent (or turbid) "serum" layer at the bottom. We defined the serum layer as the sum of any turbid and transparent layers. The total height of the emulsions (HE) and the height of the serum layer (HS) were measured. The extent of creaming was characterized as % serum = 100 (HS/HE) (Surh et al 2007).

Emulsion color. Color of emulsions was measured using a tintometer (Lovibond CAM-System 500, UK). L* is the lightness, a* and b* represent the colors where –a* is greenness, +a* is redness, –b* is blueness, and +b* is yellowness.

Spray drying of emulsions. The emulsions were prepared in the same way as described before and reserved in glass beaker. Directly after the emulsification, the spray drying of emulsion took place. A spray-dryer (model MSD 1.0; Labmaq do Brasil, Ribeirão Preto, Brazil) equipped with a two-fluid nozzle atomiser was used to convert the liquid emulsions into solid powders. Emulsions were fed into the spray dryer chamber, drying time was very short. The inlet and outlet temperature of spray dryer were set ±180°C and ±80°C respectively. Microcapsules were collected in a glass container. The produced microcapsules were transferred instantly into a glass container and immersed in ice-water bath (Anwar & Kunz 2011).

Powders analysis

1. Moisture content. The moisture content of the microcapsules was determined gravimetrically by drying them in oven at 105°C for 24 h. The moisture content (%) was recorded for each sample after stable weight was obtained.

2. Color of microcapsules. Color of microcapsules was measured using a tintometer (Lovibond CAM-System 500, UK). L* is the lightness, a* and b* represent the colors where –a* is greenness, +a* is redness, –b* is blueness, and +b* is yellowness.

3. Free surface oil of powders. Fifteen mL *n*-hexane was added to 2.5 g microcapsule sample. The resulting solution was mixed with a vortex mixer (Chiltern International, Slough, UK, operating at the speed of 4) for 2 min and then centrifuged (Centrifuge 5702, Eppendorf, Hamburg, Germany) at 8,000 rpm for 20 min. The supernatant was filtered with the whatman filter paper and filter paper then washed twice with *n*-hexane (Baik et al 2004; Hardas et al 2000). After that, *n*-hexane was evaporated in a rotary evaporator (RE 111 Rotavapor, Type KRvr TD 65/45, BUCHI, Switzerland) at 70°C, and the solvent-free extract was dried in oven at 105°C. The amount of free surface fish oil was determined gravimetrically.

4. Total oil. Two mL of acetate buffer (pH 3.0) was added to 0.5 g microcapsule sample and vortexed for 2 min. The resulting solution was then extracted with 25 mL *n*-hexane/isopropanol (3:1 v/v). The tubes were then centrifuged for 15 min at 1600 rpm

(Centrifuge 5702, Eppendorf, Hamburg, Germany). The clear organic phase then collected and aqueous phase was re-extracted with the solvent mixture (Baik et al 2004; Hardas et al 2000). After filtration through anhydrous Na_2So_4 the solvent was evaporated in a rotary evaporator (RE 111 Rotavapor, Type KRvr TD 65/45, BUCHI, Switzerland) at 70°C, and the solvent-free extract was dried at 105°C. The amount of total oil was determined gravimetrically.

5. Calculation of encapsulation efficiency. From the quantitative determinations above detailed, the encapsulation efficiency (EE) was calculated as follows:

EE = encapsulated oil (g/100 g powder) × 100/total oil (g/100 g powder)
encapsulated oil = total oil-surface free oil

6. Water solubility of microcapsules. Powders samples (0.1 g) were placed in centrifuge tubes containing 5 mL distilled water and incubated at 37°C for 5 h. The supernatants were obtained from centrifugation (KUBOTA, Japan) at 2500 ×g for 15 min and top up to 10 mL. The concentration of the soluble protein in the aqueous phase was determined using Biuret method.

7. Controlled release and core retention. Controlled-release method was performed according to the modified method of Gan et al (2008). Encapsulates (0.1 g) were placed in a glass test tube containing 2 mL of pepsin solution (2 mg mL^{-1} citric acid, pH 2.0). The suspension was incubated at 37°C in a water bath for 5 h. Samples from each test tube were drawn at intervals of 1 h until 5 h of incubation and placed at a Whatman filter paper to adsorb the released oil. The filter papers were then dried and the released oil content measured. The controlled-release of the fish oil from the encapsulated powder was expressed as proportion (%) of fish oil released from the encapsulates to fish oil retained in the dry encapsulates.

8. Particle size analysis. The particle size of the particles were determined by mixing 0.01 g of each samples with 0.5 mL water at room temperature (25±2°C) at a constant stirring rate of 400 rpm using a magnetic stirrer. The droplet size distributions of the resulting dispersions were determined by the Nano Zetasizer (Malvern Instruments Ltd., Worcestershire, UK). Triplicate experiments were performed.

Statistical analysis. In all cases, samples were analyzed in triplicate (n = 3). Significance of results was tested by an analysis of variance (ANOVA) and Duncan's Multiple-Range Test. Significance of differences was defined at $p < 0.05$.

Results and Discussion. This research was conducted to evaluate the effects of different wall materials on phisycochemical characteristics of spray-dried microcapsules. Fish oil was emulsified with three combinations of matrices as listed in Table 1, and dried by spray dryer to produce fish oil powders.

Emulsion characterization. The percentage of separation observed in the emulsions produced with different types of wall materials are shown in Table 2.

The stability assessment showed that MD:FG emulsion was stable, but κc and MD:κc+FG emulsions were unstable to sum extent, which demonstrated the formation of a small separation layer and a foam phase, 24 h after their homogenization. This was unexpected, since lecithin and FG are well known by their good emulsifying ability. In line with Dickinson & Matsumura (1991) the unfolding of the protein molecules at the droplets surface, which would increase protein–protein interaction leading to flocculation during emulsification and as a result reducing the emulsion stability, may have caused this effect. The unfolding of protein molecules of the oil–water interface may lead to changes in secondary and tertiary structure, and consequently exposure of their residues which would be linked (–S–S– linkages or disulphide linkages) within the native globular structure leading to the formation of intermolecular interaction at the oil–water interface

and flocculating. Another hypothesis that can be considered to explain this unusual behaviour is that the stability of protein-stabilized emulsions is a function of pH and other parameters.

Table 2

Characterization of emulsions prepared with different types of wall materials

Formulation	*% Separation*	*L**	*a**	*b**
MD:FG	-	90.32^{a}	2.54^{bc}	0.27^{b}
MD:κc	13.2±0.09	88.17^{a}	2.27^{c}	0.63^{a}
MD:FG + κc	1.4±0.01	90.03^{a}	3.64^{ab}	0.59^{a}

L* is the lightness, a* and b* represent the colors where –a* is greenness, +a* is redness, –b* is blueness, and +b* is yellowness. Comparison within the rows was shown in the table with the data written as mean (n = 3). Means within the same column not followed by the same letter are significantly different at $p < 0.05$ level of significance, according to Duncan's Multiple-Range Test.

Therefore, depending on the emulsions' pH, the emulsifying capacity of FG may have been lower than usual (Huynh et al 2008), affecting the emulsion stability. Color data of the microcapsules has shown that MD:κc emulsion had the lowest values in lightness (L*) and higher values in blueness (b*). This is mainly due to using lecithin as emulsifier in this treatment.

Moisture content. Moisture content is an important attribute of products since is directly related to stability. Dried products are more stable than liquid formulations, on the subject of the physicochemical aspects and microbiological spoilage. The moisture is mainly related to the drying conditions, however the composition of the formulations also plays an important role because drying could promote changes in water binding and dissociation that will affect the properties the dried product. High moisture content affects the shelf life of encapsulated fish oil (Baik et al 2004; Drusch et al 2006). This is probably because of a decrease in the glass transition temperature to values below the storage temperature, leading to relatively higher mobilities of molecules and reaction rates.

The generated microcapsules were analyzed according to the method explained previously and results are reported in Figure 1. Moisture content of microcapsules varied from a minimum value of 3.05% (MD:FG) to a maximum value of 3.46% (MD:FG+κc). There was no significant difference in the moisture content of microcapsules as affected by wall materials. It could be related to the same amount of water applied for aqueous phase preparation or smaller difference in particle size of finished microcapsules, which is an index of crust formation time. The crust keeps water within the particle, so that the interior moisture cannot be simply evaporate. Results showed that moisture content of microcapsule coated with MD:FG was lower than two other matrices. FG has lower molecular weight, and therefore, water diffusivities are greater for solutions of MD:FG than for that of MD:κc (El-Sayed et al 1990). Proteins could therefore, migrate quickly to the surface of droplet and form a continuous glass phase at droplet surface earlier than does κc. The formed case on droplet surface is consequently transformed to a tough leather-like skin and avoids the moisture evaporation. Crust acts as a barrier against moisture evaporation and thus powders with higher moisture content were produced. Rosenberg & Sheu (1996) reported a similar observation for the effect of lactose on crust formation during microencapsulation of volatiles in whey protein-based wall systems. In addition, the lactose glass phase reduces the diffusion of solvent through the wall by enhancing the hydrophilic character of the wall matrix (Moreau & Rosenberg 1996).

Rahman & Labuza (1999) suggest that the diffusion of oxygen may vary with different water content and water activity. The rate of diffusivity depends on the porosity of matrices and adsorbed water may form a protective layer against oxidation. Thus, the physical, chemical, and microbial stability of food depends highly on the water content (Rahman & Labuza 1999). According to the mentioned theory, MD:κc powders which had the highest moisture contents showed better stabilities than other samples. Therefore,

this study suggests that higher water content in a sample could correlate with better stability against oxidation. In addition, the moisture content is critical for formed microcapsules. High moisture will induce high viscosity and stickiness of microcapsules, resulting in the formation of inter-particle bridges that lead to caking and particle collapse and the release/oxidation of the core material (Beristain et al 2002; Drusch et al 2006, 2007; Le Meste et al 2002; Partanen et al 2005).

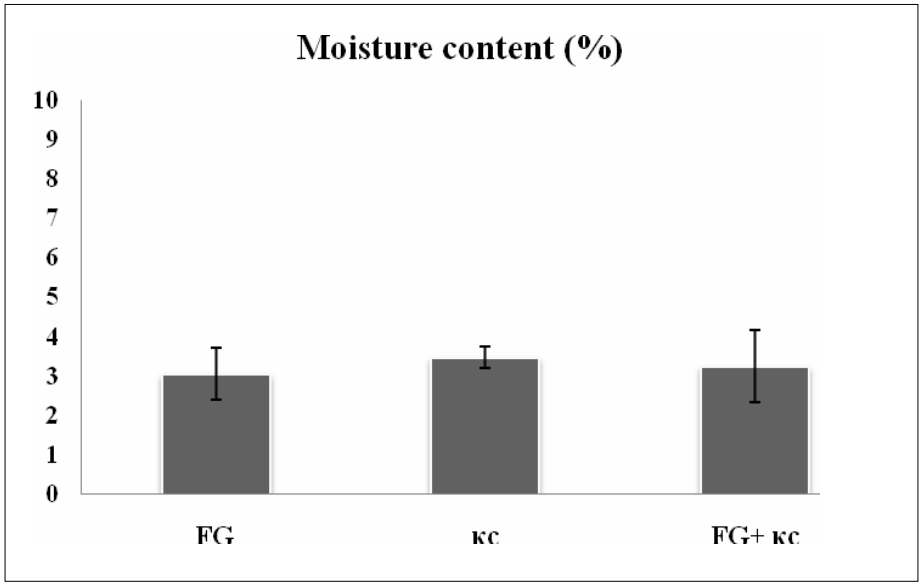

Figure 1. Moisture content of microcapsules (%). FG = microcapsules with fish oil as core and fish gelatin+Maltodextrin as wall material, κc = microcapsules with fish oil as core and κ carrageenan+Maltodextrin as wall material, FG + κc = microcapsules with fish oil as core and fish gelatin and κ carrageenan+Maltodextrin as wall materials.

Color of microcapsules. L* value is a measure of the lightness and a* and b* values are the measurement of redness and yellowness color of microcapsules respectively. Color data of the microcapsules is shown in Table 3. MD:κc had the lowest values in lightness (L*) and higher values in redness (a*).

Comparing the effects of wall materials on color of microcapsules show that b* is higher in MD:κc, which is related to using lecithin as emulsifier in this treatment, or maybe because of the high amount of free surface oil in this treatment (Drusch et al 2006).

Table 3

Color measurements of microcapsules with different wall materials

Formulation	*L**	*a**	*b**
MD:FG	92.54^{a}	2.82^{b}	0.92^{b}
MD:κc	91.35^{b}	3.43^{a}	1.87^{a}
MD:FG + κc	92.39^{a}	3.11^{ab}	0.67^{b}

L* is the lightness, a* and b* represent the colors where –a* is greenness, +a* is redness, –b* is blueness, and +b* is yellowness. Comparison within the rows was shown in the table with the data written as mean (n = 3). Means within the same column not followed by the same letter are significantly different at $p < 0.05$ level of significance, according to Duncan's Multiple-Range Test.

Surface oil, total oil and encapsulation efficiency. The surface oil expresses the amount of oil that is nonencapsulated and it is an important parameter determining the product quality because the non-encapsulated oil is prone to oxidize thus may lead to the development of off-flavors and affect the acceptability of the product (Drusch & Berg 2008). The presence of large amounts of oil on the surface of powders is undesirable, since the surface oil not only deteriorates quickly causing off-flavor but also affects the wettability and dispersability of powders (Drusch & Mannino 2009). Previous researches

showed that the amount of surface oil increased with the increasing emulsion droplet size. The possible explanation for the higher remaining oil on the surface of particles in spray drying method was the breakdown of the large emulsion droplets during atomization. This problem led to lower stability of the particles during storage, since there was no protection against oxidation, and the hydro peroxides were easily decomposed and formed off-flavor products (Soottitantawat et al 2003). The result of surface oil is shown in Figure 2. MD:κc powders shown the higher amount of free surface oil than other treatments.

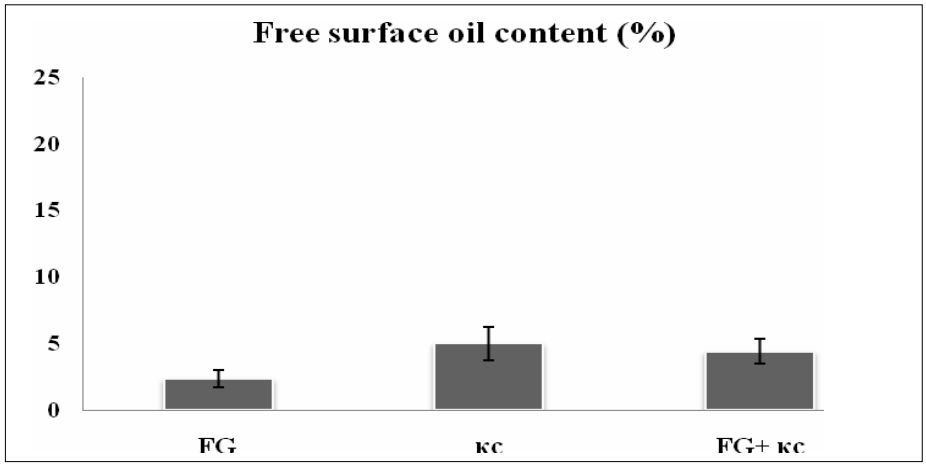

Figure 2. Free surface oil content of microcapsules (%). FG = microcapsules with fish oil as core and fish gelatin+Maltodextrin as wall materials, κc = microcapsules with fish oil as core and κ carrageenan+Maltodextrin as wall materials, FG + κc = microcapsules with fish oil as core and fish gelatin and κ carrageenan+Maltodextrin as wall materials.

The total oil content indicates the total extractable oil of microcapsules which includes both surface oil and encapsulated oil. Total oil of microcapsules is shown in Figure 3 where the MD:FG microcapsules had the highest amount of total oil.

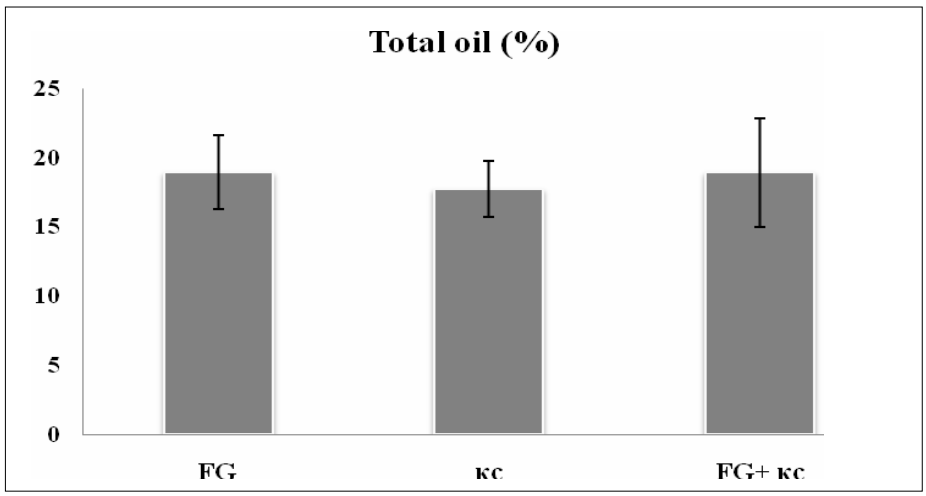

Figure 3. Total oil content of microcapsules (%). FG = microcapsules with fish oil as core and fish gelatin+Maltodextrin as wall materials, κc = microcapsules with fish oil as core and κ carrageenan+Maltodextrin as wall materials, FG + κc = microcapsules with fish oil as core and fish gelatin and κ carrageenan+Maltodextrin as wall materials.

Encapsulation efficiency (EE) reflects the real amount of fish oil that is encapsulated inside the matrix (Figure 4). Among the three formulas, MD:κc exhibited the lowest EE value. Encapsulation efficiency (EE) obtained with any wall material is statistically ($p < 0.05$) different from one to another. According to Figure 4, encapsulation efficiency varied from 71.77 to 87.65% and was significantly influenced by wall material.

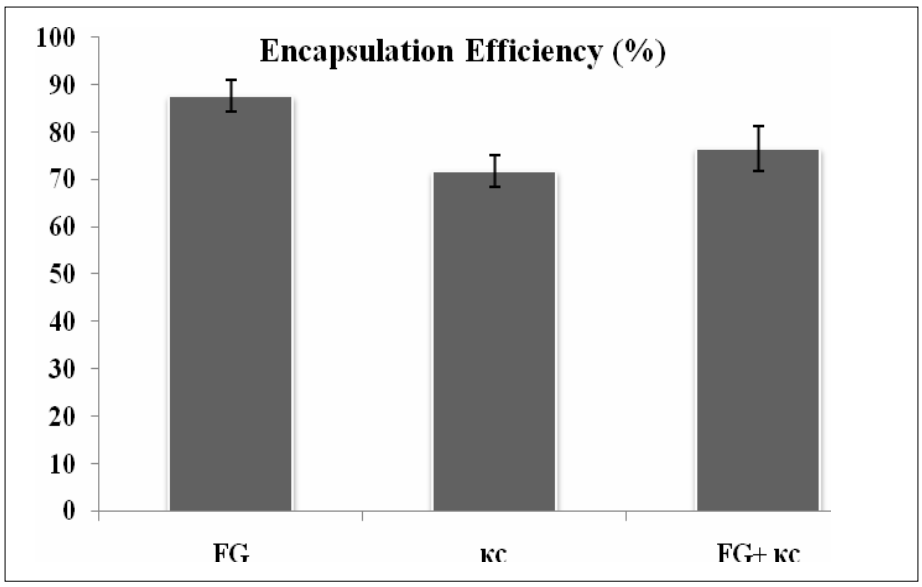

Figure 4. Oil recovery and encapsulation efficiency of microcapsules with fish oil and three matrices. FG = microcapsules with fish oil as core and fish gelatin+Maltodextrin as wall materials, κc = microcapsules with fish oil as core and κ carrageenan+Maltodextrin as wall materials, FG + κc = microcapsules with fish oil as core and fish gelatin and κ carrageenan+Maltodextrin as wall materials.

In the last years, special attention has been given to the studies aiming at improving the encapsulation efficiency during drying of food flavors and oils, by minimizing the amount of unencapsulated oil present at the surface of powder particles and thus preventing lipid oxidation and volatile losses, and extending product's shelf life (Desai & Park 2005). According to Jafari et al (2008), the main factors that affect encapsulation efficiency of microencapsulated oils and flavors are the type of wall material, the properties of the core materials (concentration, volatility), the characteristics of the emulsion (total solids, viscosity, droplets size) and the conditions of the drying process. Thus, it is important to optimize the drying process, in order to obtain the minimal surface oil in the powder particles.

The EE for MD:FG treatment was higher than other treatments which indicated that most of the oil was encapsulated and less oil was on the surface of the microcapsules. Both wall material selection and emulsion properties (stability, viscosity and droplet size) can affect the process efficiency and the microencapsulated product stability. A successful microencapsulation must result in a powder with minimum surface oil and maximum retention of the active material.

Water solubility and controlled release. Water solubility of microcapsules for controlled core release in an aqueous environment is very importance for the functionality of microcapsules. Microcapsule design requires limited or delayed water solubility (Lee & Rosenberg 2000). The water solubility of encapsulated powders is presented in Figure 5. This principle may be used to control the release of the core material.

It has long been recognized that controlled release technologies are of an interest not only for pharmaceutical industries but also for food industries. Encapsulation is always a key for putting a controlled release function in a food product (Lakkis 2007). However, in most cases, it is the encapsulant (i.e. shell material for reservoir-type, or outer matrix for matrix-type microcapsule) that has a critical effect on controlled release kinetics. Different biopolymers has proven to be a useful strategy to form shell walls with maximal protection and tailored controlled release properties for core substances (Islan et al 2012; Piculell et al 1995).

In all cases in this study, core release was time dependent. The efficiency of protection or controlled release mainly depends on the composition and structure of wall material (Young et al 1993).

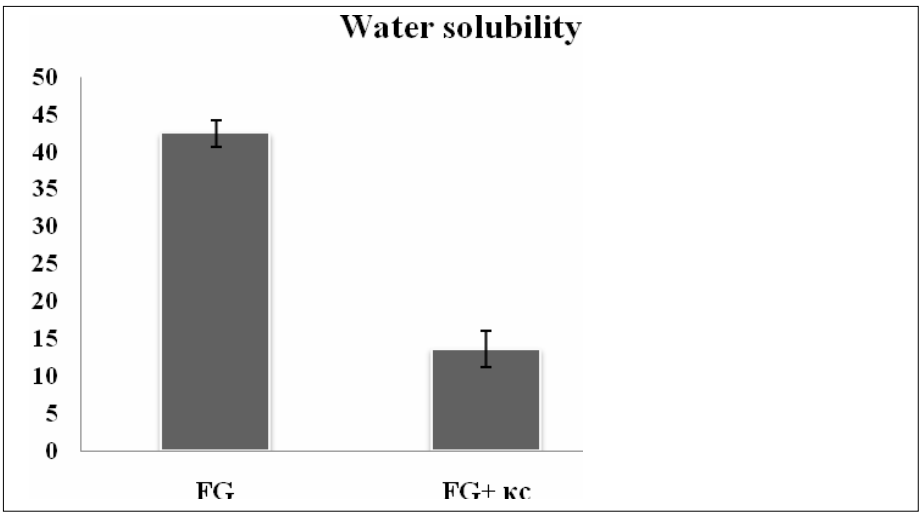

Figure 5. Water solubility of encapsulated powders. FG = microcapsules with fish oil as core and fish gelatin+Maltodextrin as wall materials and FG + κc = microcapsules with fish oil as core and fish gelatin and κ carrageenan+Maltodextrin as wall materials.

Controlled release of oil through different types of microcapsules is shown in Figure 6.

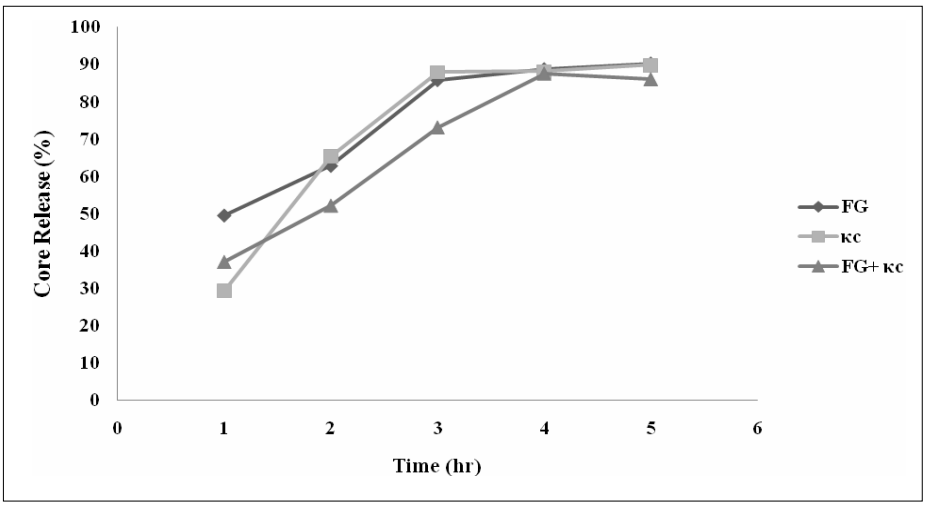

Figure 6. Cumulative release of fish oil from microcapsules during incubation at 37°C. FG = microcapsules with fish oil as core and fish gelatin+Maltodextrin as wall materials, κc = microcapsules with fish oil as core and κ carrageenan+Maltodextrin as wall materials, FG + κc = microcapsules with fish oil as core and fish gelatin and κ carrageenan+Maltodextrin as wall materials.

From Figure 6 it can be seen that fish oil release from powders is illustrated by two different phases. Initially (0-3 hr) a quick release of fish oil is observed ("burst effect") followed by a period during which the release becomes steady (after 3 hr) representing a constant release ("lag time"). The burst effect has been observed by other researchers in different polymeric matrices and may be because of the volume increase of polymer when immersed in liquid media. In aqueous liquid media, hydrophilic polymers start to hydrate causing relaxation of the polymer chain. In the present case, it is assumed that this effect is the main responsible for the initial release of fish oil from matrix; we believe that it is the fish oil that is not physically or chemically linked to matrix that will be released at this point of the process. In contrast, the subsequent almost zero order fish oil release phase results from the overlapping of several effects, such as the increase in the diffusion pathways with time, which is at least partially compensated by the increase in device porosity (polymer degradation), and the maintenance of approximately linear fish oil concentration gradients over prolonged periods of time within the microparticles

(Faisant et al 2003). The release characteristics of microcapsules are strongly dependent on their permeability (which was affected by the polarity, density, porosity, homogeneity and thickness of shell wall materials) (Donhowe & Fennema 1993).

None of the microcapsules used achieved complete core release after the incubation period showing that a certain amount of the fish oil was trapped within the network and was not released into the pepsin solution.

Conclusions. In this work it was possible to evaluate the performance of different wall materials combinations in the fish oil microencapsulation by spray drying. The MD:FG combination showed the best encapsulation efficiency result. This study clearly indicates the usefulness of fish gelatin as wall material in effective oil encapsulation in conjunction with Maltodextrin. Encapsulation of fish oil from menhaden in those matrices conducted also to differentiated releasing profiles that are explained by the dissimilar properties of the wall. Those different patterns may be very useful in finding the most suited formulation and processing condition for a specific end use of encapsulates.

Acknowledgements. The authors thank Gorgan University of Agricultural Science and Natural Resources for the financial support.

References

Anandaraman S., Reineccius G. A., 1986 Stability of encapsulated orange peel oil. Food Technology 40(11):88–93.

Anwar S. H., Kunz B., 2011 The influence of drying methods on the stabilization of fish oil microcapsules: comparison of spray granulation, spray drying, and freeze drying. Journal of Food Engineering 105:367–378.

Baik M. Y., Suhendro E. L., Nawar W. W., McClements D. J., Decker E. A., Chinachoti P., 2004 Effects of antioxidants and humidity on the oxidative stability of microencapsulated fish oil. Journal of the American Oil Chemists Society 81(4):355–360.

Belluzzi A., Boschi S., Brignola C., Munarini A., Cariani G., Miglio F., 2000 Polyunsaturated fatty acids and inflammatory bowel disease. American Journal of Clinical Nutrition 71:339S-342S.

Beristain C. I., Azuara E., Vernon-Carter E. J., 2002 Effect of water activity on the stability to oxidation of spray-dried encapsulated orange peel oil using mesquite gum (Prosopis juliflora) as wall material. Journal of Food Science 67:206–211.

Cho Y. H., Shim H. K., Park J., 2003 Encapsulation of fish oil by an enzymatic gelation process using transglutaminase cross-linked proteins. Journal of Food Science 68:2717-2723.

Daniel-da-Silva A. L., Ferreira L., Gil A. M., Trindade T., 2011 Synthesis and swelling behavior of temperature responsive k-carrageenan nanogels. Journal of Colloid and Interface Science 355:512–517.

Desai K. G. H., Park H. J., 2005 Recent developments in microencapsulation of food ingredients. Drying Technology 23(7):1361-1394.

Desobry S. A., Netto F. M., Labuza T. P., 1997 Comparison of spray-drying, drum-drying and freeze-drying for beta-carotene encapsulation and preservation. Journal of Food Science 62(6):1158-1162.

Díaz-Rojas E. I., Pacheco-Aguilar R., Lizardi J., Argüelles-Monal W., Valdez M. A., Rinaudo M., Goycoolea F. M., 2004 Linseed pectin: gelling properties and performance as an encapsulation matrix for shark liver oil. Food Hydrocolloids 18:293-304.

Dickinson E., Matsumura Y., 1991 Time-dependent polymerization of β-lactoglobulin through disulphide bonds at the oil–water interface in emulsions. International Journal of Biological Macromolecules 13(1):26–30.

Donhowe I. G., Fennema O., 1993 Water vapor and oxygen permeability of wax films. Journal of the American Oil Chemists Society 70:867–873.

Drusch S., Berg S., 2008 Extractable oil in microcapsules prepared by spray-drying: localisation, determination and impact on oxidative stability. Food Chemistry 109(1):17–24.
Drusch S., Mannino S., 2009 Patent-based review on industrial approaches for the microencapsulation of oils rich in polyunsaturated fatty acid. Trends in Food Science & Technology 20(6–7):237–244.
Drusch S., Serfert Y., van den Heuvel A., Schwarz K., 2006 Physicochemical characterization and oxidative stability of fish oil encapsulated in an amorphous matrix containing trehalose. Food Research International 39:807–815.
Drusch S., Serfert Y., Scampicchio M., Schmidt-Hansberg B., Schwarz K., 2007 Impact of physicochemical characteristics on the oxidative stability of fish oil microencapsulated by spray-drying. Journal of Agricultural and Food Chemistry 55(26):11044–11051.
El-Sayed T. M., Wallack D. A., King C. J., 1990 Changes in particle morphology during drying of drops of carbohydrate solutions and food liquids. 1. Effects of composition and drying conditions. Industrial and Engineering Chemistry Research 29(12):2346–2354.
Faisant N., Siepmann J., Richard J., Benoit J. P., 2003 Mathematical modeling of drug release from bioerodible microparticles: effect of gamma-irradiation. European Journal of Pharmaceutics and Biopharmaceutics 56:271–279.
Fernandes R. V. B., Borges S. V., Botrel D. A., 2013a Influence of spray drying operating conditions on microencapsulated rosemary essential oil properties. Ciência e Tecnologia de Alimentos 33:171–178.
Fernandes R. V. B., Borges S. V., Botrel D. A., Silva E. K., da Costa J. M. G., Queiroz F., 2013b Microencapsulation of rosemary essential oil: characterization of particles. Drying Technology 31:1245–1254.
Frost and Sullivan Research Service. End-user analysis of the global omega-3 PUFA markets. London: Frost and Sullivan Research Service. 2005.
Gan C. Y., Cheng L. H., Easa A. M., 2008 Evaluation of microbial transglutaminase and ribose cross-linked soy protein isolate-based microcapsules containing fish oil. Innovative Food Science and Emerging Technologies 9:563–569.
Gharsallaoui A., Roudaut G., Chambin O., Voilley A., Saurel R., 2007 Applications of spray-drying in microencapsulation of food ingredients: an overview. Food Research International 40(9):1107–1121.
Hardas N., Danviriyakul S., Foley J. L., Nawar W. W., Chinachoti P., 2000 Accelerated stability studies of microencapsulated anhydrous milk fat. Lebensmittel-Wissenschaft & Technologie 33:506-513.
Hogan S. A., O'riordan E. D., O'Sullivan M., 2003 Microencapsulation and oxidative stability of spray-dried fish oil emulsions. Journal of Microencapsulation 20(5):675–688.
Huynh T. V., Caffin N., Dykes G. A., Bhandari B., 2008 Optimization of the microencapsulation of lemon myrtle oil using response surface methodology. Drying Technology 26(3):357–368.
Islan G. A., de Verti I. P., Marchetti S. G., Castro G. R., 2012 Studies of ciprofloxacin encapsulation on alginate/pectin matrixes and its relationship with biodisponibility. Applied Biochemistry and Biotechnology 167:1408–1420.
Jafari S. M., Assadpoor E., He Y., Bhandari B., 2008 Encapsulation efficiency of food flavours and oils during spray drying. Drying Technology 26(7):816-835.
Keppeler S., Ellis A., Jacquier J. C., 2009 Cross-linked carrageenan beads for controlled release delivery systems. Carbohydrate Polymers 78:973–977.
Kim S., Mendis E., 2006 Bioactive compounds from marine processing byproducts – a review. Food Research International 39:383–393.
Kolanowski W., Ziolkowski M., Weissbrodt J., Kunz B., Laufenberg G., 2006 Microencapsulation of fish oil by spray drying-impact on oxidative stability. Part 1. European Food Research and Technology 222(3–4):336–342.
Kremer J. M., 2000 n-3 Fatty acids supplement in rheumatoid arthritis. American Journal of Clinical Nutrition 71:349S-351S.

Lakkis J. M., 2007 Encapsulation and controlled release technologies in food systems. Blackwell Publishing Ltd., 256 pp.
Le Meste M., Champion D., Roudaut G., Blond G., Simatos D., 2002 Glass transition and food technology: a critical appraisal. Journal of Food Science 67:2444–2458.
Lee S. J., Rosenberg M., 2000 Preparation and some properties of water-insoluble, whey protein-based microcapsules. Journal of Microencapsulation 17:29–44.
Leong K. H., Chung L. Y., Noordin M. I., Mohamad K., Nishikawa M., Onuki Y., Morishitab M., Takayamabet K., 2011 Carboxymethylation of kappa-carrageenan for intestinal-targeted delivery of bioactive macromolecules. Carbohydrate Polymers 83:1507–1515.
Let M. B., Jacobsen C., Meyer A. S., 2005 Sensory stability and oxidation of fish oil enriched milk is affect by milk storage temperature and oil quality. International Dairy Journal 15:173-182.
Let M. B., Jacobsen C., Meyer A. S., 2007 Ascorbyl palmitate, gamma-tocopherol, and EDTA affect lipid oxidation in fish oil enriched salad dressing differently. Journal of Agricultural and Food Chemistry 55:2369-2375.
Let M. B., Jacobsen C., Frankel E. N., Meyer A. S., 2003 Oxidative flavour deterioration of fish oil enriched milk: effects of oil type and ethylenediaminetetraacetate (EDTA). European Journal of Lipid Science and Technology 105:518-528.
Moreau D. L., Rosenberg M., 1996 Oxidative stability of anhydrous milkfat microencapsulated in whey proteins. Journal of Food Science 61:39–43.
Partanen R., Hakala M., Sjövall O., Kallio H., Forssell P., 2005 Effect of relative humidity on the oxidative stability of microencapsulated sea buckthorn seed oil. Journal of Food Science 70:37–43.
Piculell L., Bergfeldt K., Nilsson S., 1995 Factors determining phase behaviour of multi-component polymer systems. In: Biopolymer mixtures. Harding S. E., Hill S. E., Mitchell J. R. (eds), Nottingham, UK: NottinghUniversity Press, pp. 13–35.
Rahman M. S., Labuza T. P., 1999 Water activity and food preservation. In: Handbook of food preservation. Rahman M. S. (ed), Marcel Dekker, New York, pp. 339–382.
Rosenberg M., Sheu T. Y., 1996 Microencapsulation of volatiles by spray-drying in whey protein-based wall systems. International Dairy Journal 6(3):273–284.
Rosenberg M., Young S. L., Brooker B. E., Colombo V. E., 1993 Whey proteins as microencapsulating agents – microencapsulation of anhydrous milkfat – structure evaluation. Food Structure 12(1):31–41.
Rustad T., 2003 Utilisation of marine by-products. Electronic Journal of Environmental, Agricultural and Food Chemistry 2:458–463.
Santo V. E., Frias A. M., Carida M., Cancedda R., Gomes M. E., Mano J. F., Reis R. L., 2009 Carrageenan-based hydrogels for the controlled delivery of PDGF-BB in bone tissue engineering applications. Biomacromolecules 10:1392–1401.
Sheu T. Y., Rosenberg M., 1998 Microstructure of microcapsules consisting of whey proteins and carbohydrates. Journal of Food Science 63(3):491–494.
Soottitantawat A., Yoshii H., Furuta T., Ohkawara M., Linko P., 2003 Microencapsulation by spray drying: influence of emulsion size on the retention of volatile compounds. Journal of Food Science 68(7):2256–2262.
Stephen A. M., Philips G. O., Williams P. A., 1995 Food polysaccharides and their applications. New York: Marcel Dekker, pp. 205–217.
Surh J., Vladisavljevi Cacute G. T., Mun S., McClements D. J., 2007 Preparation and characterization of water/oil and water/oil/water emulsions containing biopolymer-gelled water droplets. Journal of Agricultural and Food Chemistry 55:175-184.
Weiss J., Takhistov P., McClements J. D., 2006 Functional materials in food nanotechnology. Journal of Food Science 71:R107-R116.
Yaqoob P., 2004 Fatty acids and the immune system: from basic science to clinical applications. Proceedings of Nutrition Society 63:89-104.
Young S. L., Salda X., Rosenberg M., 1993 Microencapsulating properties of whey proteins. 1. Microencapsulation of anhydrous milk fat. Journal of Dairy Science 76(10):2868–2877.

Received: 15 January 2015. Accepted: 09 February 2015. Published online: 11 February 2015.
Authors:
Bahareh Mehrad, Faculty of Seafood Technology, Gorgan University of Agriculture Science and Natural Resources, Basij Square, Gorgan, Iran, e-mail: Bahar.mehrad@gmail.com
Bahareh Shabanpour, Faculty of Seafood Technology, Gorgan University of Agriculture Science and Natural Resources, Basij Square, Gorgan, Iran, e-mail: b_shabanpour@yahoo.com
Seyed Mahdi Jafari, Faculty of Food Science, Gorgan University of Agriculture Science and Natural Resourses, Basij Square, Gorgan, Iran, e-mail: jafarism@hotmail.com
Parastoo Pourashouri, Faculty of Seafood Technology, Gorgan University of Agriculture Science and Natural Resources, Basij Square, Gorgan, Iran, e-mail: pourashouri.p@gmail.com

How to cite this article:
Mehrad B., Shabanpour B., Jafari S. M., Pourashouri P., 2015 Characterization of dried fish oil from Menhaden encapsulated by spray drying. AACL Bioflux 8(1):57-69.