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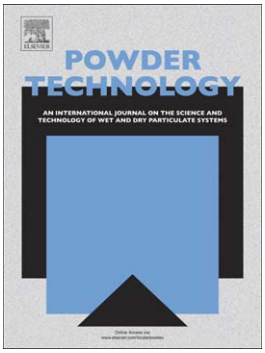

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Oxidative stability of walnut (*Juglans regia* L.) and chia (*Salvia hispanica* L.) oils microencapsulated by spray drying

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Abstract

This work was primarily aimed to evaluate the use of maltodextrin in combination with hydroxypropyl methylcellulose as wall materials for microencapsulation of both walnut and chia oils by spray drying. The effect of rosemary extract on the oxidative stability of the microencapsulated oils was examined under prolonged storage conditions. After a 45-day storage, the microencapsulated chia oil with rosemary extract showed lower formation of primary oxidation compounds than the control without additives, highlighting a protective effect of the antioxidant. Nevertheless, during the entire storage period the unencapsulated chia oil reported lower oxidative damage than their encapsulated counterparts, showing that the spray drying process affected its stability negatively. Walnut oil microencapsulation protected the oil from oxidation in comparison with its unencapsulated counterpart which was further apparent from the 30th day of storage. The addition of 1600 ppm of rosemary extract effectively protected microencapsulated walnut oil, showing -throughout the storage period- lower oxidation values in comparison with its analogous unencapsulated oil. The microcapsule surface oil and the images obtained by scanning electron microscopy at the beginning and at the end of the assay showed that the wall material has not been altered during storage.

Key words: walnut oil, chia oil, microencapsulation, spray drying, oxidative stability

Highlights

Microencapsulation significantly protected walnut oil against oxidation.

Rosemary extract had a strong effect on walnut oil oxidative stability.

The microencapsulation technique negatively affected the oxidative stability of chia oil.

Wall matrix was not altered during the storage period.

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Abbreviations

CO: chia oil

EE: encapsulation efficiency

FA: fatty acids

HPLC: high pressure liquid chromatography

HPMC: hydroxypropyl methylcellulose

HPV: hydroperoxide value

K_{232}: conjugated dienes

K_{270}: conjugated trienes

LSD: least significant difference

M: microcapsules

MD: maltodextrin

MUFA: monounsaturated fatty acid

ND: not detected

OSI: oxidative stability indexes

PUFA: polyunsaturated fatty acid

RE: rosemary extract

SEM: scanning electron microscopy

SO: surface oil

SY: solid yield

TO: total oil content

UV: ultra violet

WO: walnut oil

1. INTRODUCTION

Currently, walnut and chia seeds are crops of high economic interest for the food, pharmaceutical and cosmetic industries. Both, walnut and chia seeds, contain high levels of oil, 52-70 % and 25-38 %, respectively [1,2,3,4,5,6,7,8]. The major components of these oils are triacylglycerols, in which monounsaturated (mainly oleic acid) and polyunsaturated (PUFA, linoleic and α-linolenic acids) fatty acids (FA) are present in high amounts [9,1,7,10]. Omega-3 fatty acids play an essential role in physiology, especially during fetal and infant growth [11], and in the prevention of cardiovascular diseases, and are

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antithrombotic, antiinflammatory, antiarrythmic agents favoring plaque stabilization [12]. While these oils can be consumed directly as salad dressings, an interesting application would be to have them built into food products of mass consumption. Unsaturated fatty acids (ω-3, ω-6 and ω-9) are chemically unstable in the presence of oxygen, light, moisture and heat. Frankel [13] informed that in neat systems without an added initiator, linoleate was 40 times more reactive than oleate, and linolenate was 2.4 times more reactive than linoleate.

Microencapsulation of oils by spray drying in a polymeric matrix is an alternative that has been used by several researchers in order to protect unsaturated fatty acids [14,15,16,17]. This technology is defined as the process of enveloping a solid, liquid or gaseous substance within another substance in very small sealed capsules from which the core material is gradually diffused through the capsule walls at controlled rates under specific conditions [17]. Among the different techniques developed to encapsulate food ingredients, such as physical methods (e.g. pan coating, air-suspension coating, centrifugal extrusion, vibration nozzle and spray drying) and chemical methods (e.g. interfacial polymerization, in-situ polymerization, and matrix polymerization), the most common technique applied in this field is spray-drying because it is rather inexpensive and straightforward [18,17]. Spray drying involves the atomization of emulsions into a drying medium with high temperature, resulting in a very fast water evaporation, which results in a quick crust formation and in a quasi-instantaneous entrapment of the core material [14,18,19,20,16]. The main objective is to build a barrier between the component in the particle and the environment. This barrier may protect against oxygen, water, light, and also avoids contact with other ingredients. The efficiency of protection and controlled release depends mainly on the composition and structure of the established wall, and also on the operating conditions during the production and use of these particles (temperature, pH, pressure, humidity). An area of research of increasing interest is the use of biopolymer blends as wall materials that may allow the increase of the encapsulation efficiency and shelf life of microcapsules [21,22,23]. The selection of wall material influences the emulsion stability during its formation and after the drying process affecting the characteristics of the resulting microcapsules [24,25,26,27]. The usual encapsulating agents are proteins (e.g. milk and gelatine), gums (e.g. acacia and alginate), carbohydrates (e.g. sucrose, maltodextrins, modified starch, cyclodextrins and cellulose), lipids, fats, waxes, lecithins (emulsifiers) and fibers. Maltodextrin is a hydrolyzed starch commonly used as wall material in microencapsulation of food ingredients [18]. It offers advantages such as relatively low cost, neutral aroma and taste, low viscosity at high solid concentration and good protection against oxidation. However, the biggest problem of this wall

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material is its low emulsifying capacity. Also, it can be used in combination with other surface active biopolymers, such as gum Arabic [28,29], modified starches [30,29] and proteins [31,32], in order to obtain an effective microencapsulation by spray drying. Hydroxypropyl methylcellulose (HPMC), water soluble, non ionic cellulose derivative has been widely used in foods products. The presence of hydroxypropyl and methyl groups renders the cellulose molecules hidrophobicc and makes them surface active and adequate for oil microencapsulation [14,18,30]. Most of the works found in the literature on microencapsulation of PUFA-rich oils have been carried out by using fish oils [31,33,34,35]. Fish oils are similar to walnut and chia oils because they are very rich in PUFAs, although their compositions differ largely. Some works have reported the microencapsulation of fish oils containing approximately 27% [34], 29% [17], or 33% [36] of ω-3 fatty acids. Walnut and chia oils contain about 15 and 60 %, respectively, of α-linolenic (ω-3) FA [5,8,37]. Therefore, even though they could show a similar behavior when microencapsulated, their behavior may not be the same during spray drying and storage. Although encapsulation itself prevents lipid oxidation, additional stabilization with antioxidants is required to ensure maximum protection during processing and subsequent storage of microencapsulated bioactive ingredients [34]. Rosemary extract (*Rosmarinus officinalis*) has antioxidant properties and is widely used in food industry. The antioxidant activity of rosemary extract is associated with the presence of phenolic compounds, such as carnosic acid, rosmarinic acid, carnosol, rosmanol, rosmariquinone and rosmaridiphenol, which react with free radicals formed in the oxidation process [38].

Very little information is available on microencapsulation of walnut and chia oils [39,40,41] and none of the published works reported the influence of maltodextrin combined with hydroxypropyl methylcellulose as wall materials, on the oxidative stability of these oils.

This work was primarily aimed to evaluate the use of maltodextrin combined with hydroxypropyl methylcellulose as wall materials for microencapsulation of both walnut and chia oils by spray drying. In addition, the effect of rosemary extract on the oxidative stability of the microencapsulated oils was examined under prolonged storage conditions.

2. MATERIALS AND METHODS

2.1 Materials

Hydroxypropyl methylcellulose (HPMC, Methocel K99, Ciclo Química, Argentina), maltodextrin (MD, DE15, Distribuidora Nicco, Argentina) and soy lecithin (Distribuidora Nicco, Argentina) were used as an emulsion stabilizer and a protection for the oil drops in the emulsion and in the final powder.

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Walnut oil (WO) was obtained from healthy and mature kernels from Franquette variety (Catamarca, Argentina); while, chia oil (CO) was obtained from seeds coming from the province of Salta, Argentina (Nutracéutica Sturla SRL). WO and CO extraction were performed as described elsewhere [6,8]. Briefly, walnut kernels were ground and particles between 2.4 and 4.8 mm were selected using an automated screen and conditioned to obtain 7.5% (w/w) moisture content. Chia seeds were only hydrated to obtain 10% (w/w) moisture content [8].

Oil expression was carried out with a Komet screw press (Model CA 59 G; IBG Monforts, Monchengladbach, Germany), with a 5 or 6 mm restriction die, as appropriate, and a screw speed of 20 rpm.

Guardian Rosemary Extract 08 (RE, oil soluble) was from Danisco (Copenhagen, Denmark).

2.2 Emulsion preparation

For spray drying, blends of both HPMC/MD/WO and HPMC/MD/CO in water emulsions were prepared. MD (6 %) was dissolved in demineralized water at room temperature and then HPMC (3 %) was slowly added and stored for 24 h at 4 °C [42]. RE was incorporated to the oil by magnetic stirring for 5 min at 90 rpm and under these conditions a homogenous oil appearance was achieved. Blends of oil: soy lecithin (0.3 g lecithin /9 g oil) were prepared and then incorporated drop by drop into the suspension in a ratio 2:1 (wall material: oil) for 10 min, using a homogenizer Ultraturrax T18. The resulting emulsions (200 mL lots) were stored at 4 °C before the spray drying process. The droplet size of the emulsions ranged from 0.06 to 29 and 0.13 to 24 µm, for CO and WO, respectively.

2.3 Spray-drying microcapsules preparation

The spray-drying process was performed in a laboratory-scale Mini Spray Dryer Büchi B-290 (Büchi Labortechnik AG). A two fluid nozzle with a cap orifice diameter of 0.5 mm was used; the air atomizing pressure was kept constant at 6 bars for all the experiments. The microcapsules were obtained in triplicate under the following operating conditions: air inlet temperature, 163 °C; atomization air flow rate, 279 L/h; pump setting, 10 % and aspirator setting, 100 % [42].

2.4 Experimental design for storage stability test

Microencapsulated and bulk WO and CO were separated in 250 mL dark glass bottles – samples. The RE was added to oil samples according to quantities stated in Table 1, based on a previous work [43]. After mixing (magnetic stirrer, 10 min, 90 rpm), a homogeneous oil appearance was achieved. The oil samples were utilized to prepare the blend and then incorporated to the emulsion. The microcapsules obtained were set in 250 mL amber glass bottles and placed inside a thermostated chamber at 25 ± 1 ºC and 40% of

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relative humidity. Each treatment (consisting of a combination from oil plus additive) was prepared in triplicate. Bulk oil samples with and without RE were used as controls. Samples were stored for three months. Every fifteen days each individual sample was withdrawn from the chamber for scheduled analyses.

2.5 Solid yield (SY)

The solid recovered was calculated as the ratio of the powder weight collected after every spray-drying experiment to the initial amount of solids in the sprayed dispersion volume.

2.6 Powder analysis

2.6.1 Surface oil (SO) and encapsulation efficiency (EE)

Surface oil was measured by adding twice 30 mL of petroleum ether to 500 mg of powder contained in a filter paper, for 2 min and 30 s, respectively, at room temperature. The solution containing the extracted oil was transferred to a clean flask, which was allowed to evaporate and then dried at 60 °C until constant weight. The surface oil was calculated based on the difference between the initial clean flask and that containing the extracted oil residue. Total oil was assumed to be equal to the initial oil, since preliminary tests [41] revealed that all the initial oil was retained. The encapsulation efficiency (EE) was calculated as follows:

$$EE = (TO - SO)/TO * 100$$

Where TO is the total oil content and SO is the surface oil content.

2.6.2 Moisture content (MC)

Powder moisture content was determined by a moisture analyzer with halogen heating (model M45, OHAUS). Sample moisture content analysis was performed immediately after the spray-drying step.

2.6.3 Particle Analysis

Particle morphology was evaluated by scanning electron microscopy (SEM). Powders were attached to a double-sided adhesive tape mounted on SEM stubs, coated with 3–5 mA gold/palladium under vacuum and examined with a FEG SEM scanning electron microscope (Carl Zeiss - Sigma, Germany). Particle diameter was measured by image processing software ImageJ (National Institute of Health, USA). Fracture microcapsules were obtained by mechanical force.

2.7 Oil chemical analyses

The bulk and encapsulated oils were characterized using the following methods. The values of acid and conjugated trienes (K_{270}) were evaluated using standard AOCS [44] methods. Tocopherol content was measured by HPLC according to Pocklington and Dieffenbacher [45]. Chlorophyll and carotenoid compounds were determined using the method of Mínguez-Mosquera et al. [46]. To determine the carnosic

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acid content in rosemary extract (RE), HPLC and UV spectrofotometric analyses were carried out according to the procedures employed by Visentín et al. [47]. The total phenol content was measured by UV spectrophotometric analyses according to Siddhuraju et al. [48]. Oxidative stability indexes (OSI) were determined using the Rancimat analysis, and corresponded to the break points in the plotted curves [13]. Air flow rate was set at 20 L/h and temperature of the heating block was maintained at 110 ºC.

The hydroperoxide value (HPV) was determined by iodometric titration, which measures the iodine produced from potassium iodide by the peroxides present in the fat sample, using Kolanowski et al. [49] procedure with minimum modifications. A powder sample of 2.0 g (in the case of microencapsulated oil) or 2.0 g sample of bulk oils were dissolved in 7 mL chloroform. After that, 10 mL glacial acetic acid was added, and the mixture was stirred with a magnetic stirrer for a few seconds to ensure mixing. Then 0.5 mL saturated potassium iodide solution was added. After 1 min under darkness, 30 mL H_2O purified was immediately added and the titration was started. The liberated I_2 was titrated with 0.01 N $Na_2S_2O_3$ using a starch solution (1%) as an indicator, until the solution became colorless. The mixture was stirred magnetically for the whole time of titration procedure. The advantage of this modification in the analytical method was that the coating material dissolved in the chloroform liberated oil from the inside.

Conjugated dienes (K_{232}) value was determined using the spectroscopic method. A powder sample of 0.07 g was weighted in a 10 mL volumetric flask; the volume was complete with hexane HPLC grade. The flask was shaken every 15 min for 2 h. Then the samples were centrifuged at 13000 rpm for 15 min at 5 °C. Finally, the sample absorbance at 232 nm was determined.

Fatty acid composition was analyzed by gas chromatography [5].

2.8 Statistical analysis

Analytical determinations were the average of duplicate measurements from three independent samples. Statistical differences were estimated by ANOVA test at the 95% level ($p < 0.05$) of significance. Whenever ANOVA indicated a significant difference, a pair-wise comparison of means by least significant difference (LSD) was carried out.

3. RESULTS AND DISCUSSION

The microcapsules were characterized in terms of solid yield, moisture and fat contents (% encapsulation yield). Solid yield was 37.2 ± 1.34 % and 39.7 ± 0.78 %, for WO and CO microcapsules, respectively. The moisture content varied between 0.95 and 2.13 % for microcapsules with and without RE, respectively. The average encapsulation efficiency for WO and CO was 0.73 ± 0.03 g of oil encapsulated/g of total oil.

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Table 2 shows quality and compositional parameters of fresh WO and CO used for the microencapsulation storage stability test. Acid, hydroperoxide and the specific extinction coefficients (K_{232} and K_{270}) values were similar to those informed by Martínez et al. [6,8] for WO and CO, indicating that the oil extraction method did not adversely affect oxidative or hydrolytic oil indicators. Regarding fatty acid profile, WO contained 52.4%, 22.9% and 15.2% of linoleic, oleic and linolenic acid, respectively, while CO presented linolenic acid as the major fatty acid (61.8%), followed by linoleic acid (20.1%) and oleic acid (7.18). The ω-3/ω-6 fatty acid ratios of CO (3.07) and WO (0.29) are markedly higher than those from other vegetable oils, e.g. soybean oil (0.15) and olive oil (0.13) [10]. Although it seems clear that such fatty acid composition is favourable from a nutritional point of view, higher contents of linoleic and linolenic acids result in poorer oxidative stability and shorter shelf life of the oils. In this sense, Roman et al. [50] who studied the oxidative reactivity of unsaturated fatty acids from sunflower, high oleic sunflower and rapeseed oils informed that the unsaturation degree of the oils tested was the predominant parameter influencing the progress of oxidation kinetics.

The measurement of tocopherol isomers in these oils is well documented (Table 2). In both WO and CO *γ* - tocopherol was the main component, followed by *δ*- and *α*-tocopherol. *Beta*-tocopherol was not detected. Although the same order of abundance was informed by Martinez et al. [7] and Ixtaína et al. [10], the total tocopherol content reported in this study was higher than the content observed elsewhere (718 and 716 against 211 - 297 and 238 – 427 µg/g oil, respectively). Both WO and CO presented low levels of phenol as well as pigment (carotenoids and chlorophylls) components. Taking into account the chemical composition (mainly PUFAs) and oxidative stability indexes of WO and CO (Table 2), the lower thermo oxidative stability of CO is evident. Therefore, both oils may present a quite different behavior under selected process conditions, especially considering the drying temperature (163 ° C).

Figures 1 and 2 show the oxidation patterns during the storage of bulk and encapsulated WO and CO for 90 days. In contrast with data reported by Serfert et al. [34] immediately after the spray-drying process, no increase in lipid oxidation products was found in none of the oil samples, indicating an effective chemical stabilization during the homogenization experiments. As expected, during the storage stability test, WO and CO showed a quite different behavior. Regarding bulk and microencapsulated WO, there was a significantly faster increase in the hydroperoxide value (HPV) in bulk WO than in microencapsulated WO. Even though microencapsulation significantly protected WO against oxidation, rosemary extract had a strong effect on WO oxidative stability (Figure 1). At 90-day storage, the HPV amounted to 64.6 and 26.4 meq O_2/kg oil in WO and M – WO samples, respectively, whereas WO samples with rosemary extract, especially M - WO +

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RE 1600 sample, presented the lowest HPV (2.98 meq O_2/kg oil). Serfert et al. [34] and Ahn et al [51] also showed a significant reduction in the HPV of microencapsulated fish oil and high oleic sunflower oil added with rosemary extract. Microcapsule wall components seem to be an effective barrier against WO oxidative degradation. These results are in agreement with those reported by Calvo et al. [40] who found that a blend of maltodextrin (32.8%), carboxymethylcellulose (65.6%) and lecithin (1.6%) was effective in preventing walnut oil oxidative degradation. In the present work, a protective effect associated with the microencapsulation process was observed, which was enhanced by the addition of rosemary extract as a natural antioxidant.

On the other hand, the microencapsulation technique negatively affected the oxidative stability of CO with and without RE. A significant increase in HPV values was observed after 60-day storage in M - CO and M – CO + RE 1600 treatments (32.8 and 14.3 meq O_2/kg oil, respectively). Throughout the storage period, bulk CO showed significantly lesser oxidative damage than their microencapsulated counterparts, suggesting that encapsulated oil undergoes oxidation associated with some of the steps involved in the microencapsulation spray drying process. After 90-day storage, CO and CO + RE 1600 achieved HPV of 14.2 and 8.06 meq O_2/kg oil, respectively, whereas M – CO and M - CO + RE 1600 showed values of 214 and 110 meq O_2/kg oil, respectively. Similar trends were observed for K_{232}.

In order to estimate the oxidation rate of bulk and microencapsulated WO and CO with and without rosemary extract, the data were regressed linearly (data not shown). As discussed above, the rate of oxidation follows this order: M – CO > M – CO – RE 1600 > WO > M – WO > CO > WO – RE 1600 > CO + RE 1600 > M – WO + RE 1600. These results could be justified by the highest PUFA/MUFA ratio of CO (Table 1) which leads to the lowest oxidative stability previously reported [52,53,54,50].

The microcapsule images obtained by SEM at the beginning and at the end of the oxidative stability test showed a continuous surface structure, without pores or apparent fractures, indicating that wall matrix was not altered during the storage time (Figure 3). At the beginning of the assay, dispersed microcapsules were observed, separated from one another, which allows identifying individual particles (Figure 3, A1 and A2, WO and CO, respectively). After 90-days storage, the formation of agglomerates was evident; they may have formed due to the hygroscopic characteristics of the microcapsules wall materials (Figure 3, B1 and B2, WO and CO, respectively). The morphology of WO and CO MD and HPMC microcapsules were quite different to those obtained by Ahn et al. [55] and Anwar and Kunz [35] who used milk protein isolate - maltodextrin and Soyafibe-S-EN100 – maltodextrin - hydroxypropyl betacyclodextrin - octenyl succinic anhydride as coating

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materials. The particles exhibited large size range, varying from 10 to 100 μm. The wall thickness of both microcapsules was between 0.277 and 0.474 μm (Figure 4). Anwar and Kunz [35] showed that the particle microstructure is an important cause of oxidation aside from coating material and heat treatment. Although freeze drying process involves no heat or very low drying temperature, the final particle morphology was a limiting factor, particularly in relation to oxygen diffusivity. As the same way, another aspect to take in account is oil migration from the core to the surface. The porous, irregular and flake-like structure of the freeze drying powder accelerated oxidation due to an easy oxygen access into matrices; the oxygen thus reached the non-encapsulated oil. Moreover, Moreau and Rosemberg [56] and Kagami et al. [57] reported that oxygen permeability of the wall matrix is affected negatively by the porosity of the wall and determines the oxidative stability of the core substance. Since in this work all the microcapsules were obtained with the same MD and HPMC combination and considering the similar morphology of the microcapsules, with no apparent pores, the differences observed between microencapsulated WO and CO in terms of stability may be due principally to the effect of the spray drying temperature on the unsaturation degree of the oils.

4. CONCLUSIONS

In this work it was possible to evaluate the complexity of lipid oxidation phenomena affected by the microencapsulation process and by the different fatty acid composition of the oils. Although the drying process slightly affected the stability of walnut oil, this effect was reduced by the presence of rosemary extract. Moreover, during the 90-day storage, M–WO–1600 ppm RE treatment had the lowest contents of primary oxidation compounds. This suggests a combined protective effect of the wall matrix (MD/HPMC) and the natural antioxidant (RE) used on walnut oil chemical quality. Since there were no differences in the composition of the wall matrix and considering the similar morphology of the microcapsules, with no apparent pores, the differences observed between microencapsulated WO and CO in terms of stability may be due principally to the effect of the spray drying temperature on the unsaturation degree of the oils. In this sense, the chemical damage caused by the temperature (163 °C) in chia oil, unlike that observed in walnut oil, could not be countered by the wall matrix or rosemary extract, showing that the system (wall matrix and antioxidant) was not effective in protecting the chia oil during the storage stability test.

Considering that the microencapsulation of edible oils by spray drying technologies shows a remarkable interest for agri-food industries due to low cost and available equipment, further assays using others spray drying conditions and microcapsule formulations need to be tested in order to maximize the oils chemical quality.

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List of comments

Reviewer #1: The article deals with an interesting subject and is well written. However, it is a simple work and shows few results. A large discussion on the oils composition is presented, which is not the focus of the paper. Particles size values are strange. Other particular comments are given below:-

Graphical Abstract: The graph of HPV x Days should exlpain the meaning of the abbreviations, such as HPV, CO, RE, M and WO. The x axis should be "Time (days)"
R: It was done.

Introduction: The second paragraph ("Microencapsulation of oil by spray drying…") is too long and should be more concise. In addition, the authors talk about the advantages of maltodextrin, but do not justify the use of hydroxypropyl methylcellulose. Why was this material choosen?
R: It was changed according to the reviewer suggestion.

- Line 124: Oil expression??? Do you mean "oil pressing"?
R: Oil extraction by mechanical pressing (screw press) is industrially called “oil expression” (See J Singh and PC Bargale (2000) Development of a small capacity double stage compression screw press for oil expression, Journal of Food Engineering 43, 75-82.

- Line 127: Please change "Spray-drying emulsion preparation" for "Emulsion preparation".
R: It was done.

- Lines 130-131: The authors mention that the concentration of MD and HPMC was chosen from previous studies (3 % and 3 % or 6 % and 3 %, respectively). What was the criterion to select the concentrations of 6% and 3%? It should be mentioned.
R: The paragraph was reformulated to clarify the methodology. The selection of wall material proportion was done in order to increase oil content in the emulsion based on a previous work. (Roccia et al 2014, see [42]).

- Line 131: How was the mixing performed? Which type of mixer? It should be detailed.
R: It was done. The mixing was performed with a laboratory magnetic stirrer during 10 minutes at 90 rpm.

- Lines 135-136: How was the droplet size measured? It should be mentioned.
R: The droplet size was measured by image analyses from pictures obtained by optical microscopy. The pictures were analyzed using the software Image J. More details about methodology were added on Particle Analysis section.

- Lines 141-142: It is not necessary to separate in items A, B, C and D.
R: It was done.

- How was the amount of rosemary extract chosen? Why 1600 ug/g oil?
R: It was included in the text. The selection was based on a previous work from the authors (Martínez et al, 2013 [43]). In this work, different concentrations of RE was evaluated by oxidative stability test performed by Rancimat Method. In addition, this concentration is in according with the manufacture suggestions (Danisco).

- Line 145: What do the authors mean by "careful mixing"?
R: The mixing process was achieved using a laboratory magnetic stirrer during 10 minutes at 90 rpm. This data were included in the text.

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- In Table 1, the treatments should be better described. For example: WO = Bulk WO without RE, M-WO = Microencapsulated WO without RE, etc. The title should be: Table 1. Treatments for the storage stability study of bulk and microencapsulated walnut oil (WO) and chia oil (CO), with or without rosemary extract (RE).- In the Results, the authors present the quality and compositional parameters of bulk oils, but not of the encapsulated oils, which would be very interesting.
R: It was done according reviewer suggestion. The main objective of this contribution was to study the oxidative stability of microcapsules. The quality and compositional parameters of microencapsulated oil are under study and will be discussed in another publication.

- The lower oxidative stability of M-CO and M-CO + RE can also be attributed to the higher surface area, which makes powders more exposed and more susceptible to oxidation. It should be discussed in the text.
R: It was done. The effect of the surface particle characteristics were discussed on line 277 and next.

- Lines 269-270: I have doubts on these particles sizes. 1470 and 1850 um are not the size of MICROcapsules. It is probably the size of the agglomerates, not of the capsules. I do not think MEV is a good method to measure particles size.
R: The reviewer comment is correct. We have made a type mistake. There particle size data was corrected. The particle size range was measured by image analyses from SEM images. The micrographs were analyzed using the software Image J.

- The figures resolution is poor.
R: The figures were modified.

Reviewer #2: Oxidative stability of walnut (Juglans regia L.) and chia (Salvia hispanica L.) oils microencapsulated by spray drying

The manuscript under review deals with a very interesting subject, the oxidative stability of two different oils (rich in monounsatured and polyunsaturated fatty acids) microencapsulated by spray drying. The effect of rosemary extract, which has antioxidant properties, on the oxidative stability is particularly investigated. For both oils under study, the experimental collected data proved the rosemary extract protective effect. While spray drying and the selected wall materials were found to be appropriate to encapsulate the walnut oil, a negative effect of the microencapsulation process on the oxidative stability of the chia oil was observed. Taking into account the scarce information about the encapsulation of these specific oils, the present paper can be considered valuable. However, and as concluded by the authors, it may not find wide application based on just one set of operating and formulation conditions. As the microcapsules attributes are a function of the spray
drying conditions, more operating points should be tested to understand the relationship between the quality of the obtained particles and the physicochemical phenomena occurring in the production unit in order to determine the process feasibility and optimum operation.

The paper is well organized and documented by experimental work. The literature review is adequate. The highlights well summarized the findings of the article. The graphical abstract well depicts the manuscript content. After providing some clarifications on the foregoing minor comments, the paper should be accepted for publication in the Powder Technology Journal.
Minor Comments
1. Line 132
The proportion of lecithin in the oils:lecithin blends should be given.
R: it was done; the amount of lecithin incorporated was 0,3g lecithin / 9g oil.

2. Line 133

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Even though the droplet size of the emulsions is reported, no information about the method used to determine it is given.
R: The droplet size was measured by image analyses from pictures obtained by optical and scanning electron microscopy. The pictures were analyzed using the software Image J.

3. Line 165
According to the authors, preliminary tests revealed that all the initial oil was retained by the spray drying product. Considering the relatively low yields obtained, the results of those tests should be included to probe that no oil is loss during the spray drying process.
R to line 156: A reference with the used methodology was included in the text.

4. Figures
the figures quality should be improved.
R: The figures were modified.

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Figure Captions

Figure 1: Hydro peroxide value (HPV) evolution during the storage time of WO and CO with and without microencapsulation.

Figure 2: Conjugated dienes (k232) evolution during the storage time of WO and CO with and without microencapsulation.

Figure 3: Scanning electron micrographs of microencapsulated WO and CO at the beginning of the stability test (A1 y A2, respectively) and after 90 days (B1 and B2, respectively).

Figure 4: WO and CO microcapsules submitted to fracture.

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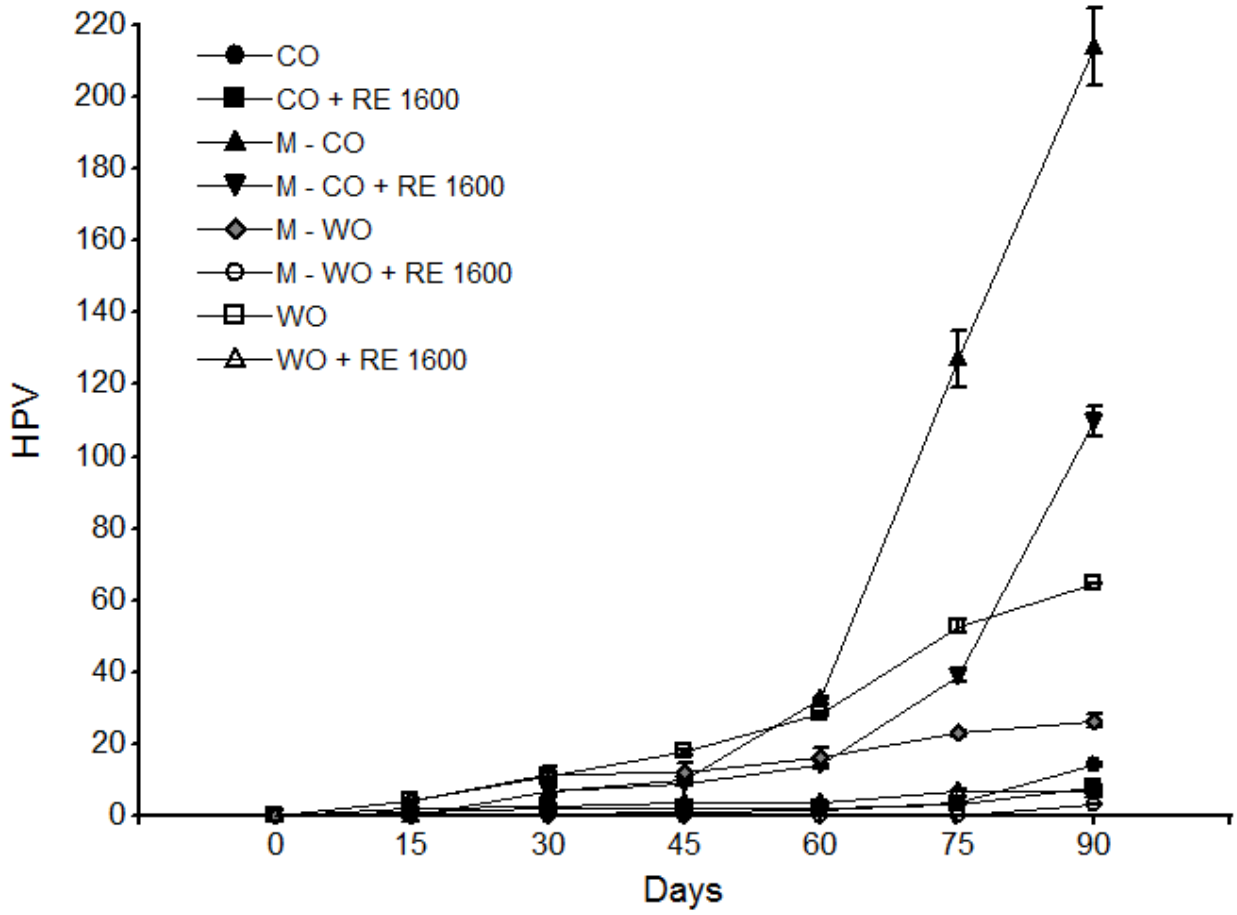

Figure 1

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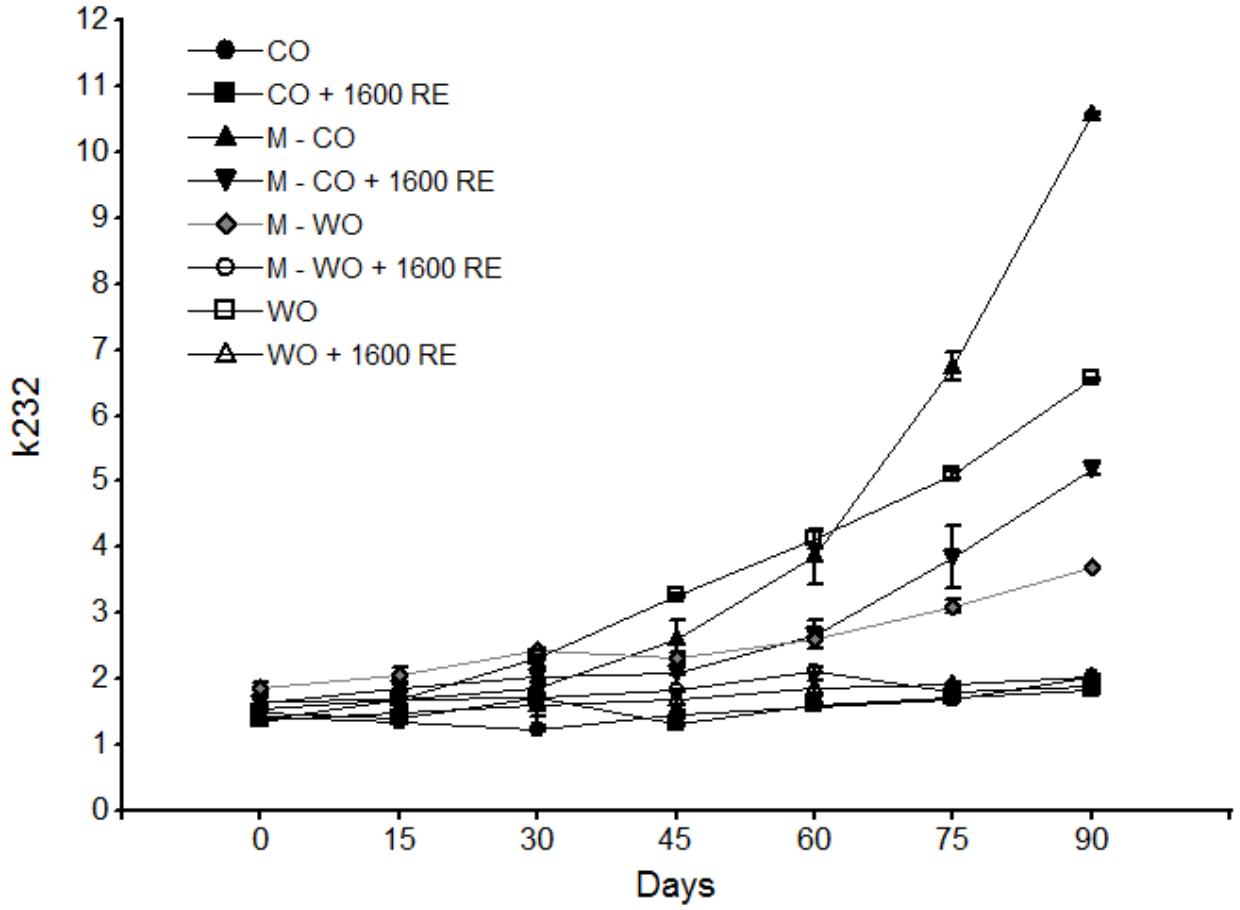

Figure 2

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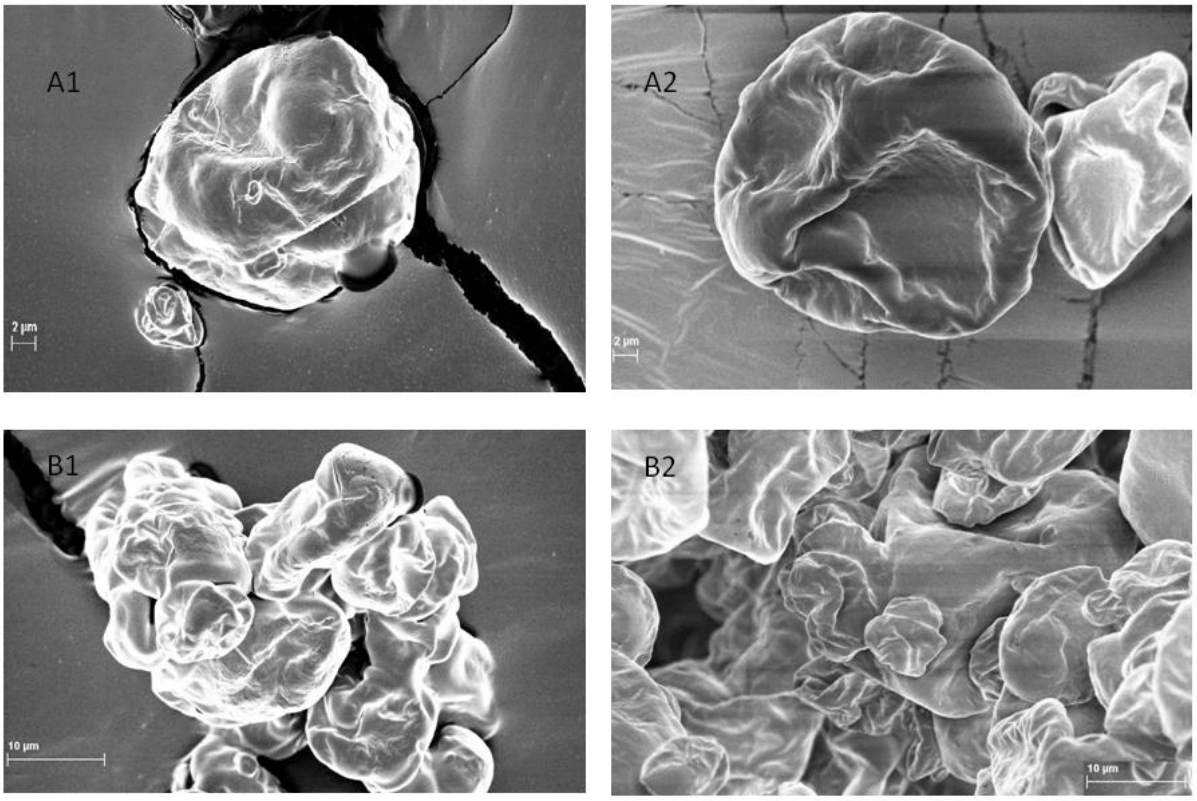

Figure 3

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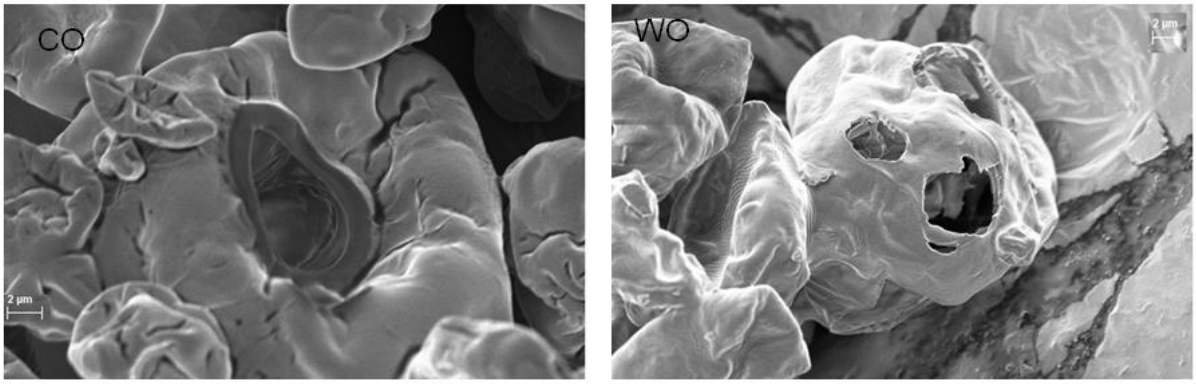

Figure 4

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Table 1. Treatments for the storage stability study of bulk and microencapsulated walnut oil (WO) and chia oil (CO), with and without rosemary extract (RE).

Code	Treatment
WO (bulk oil)	WO without RE
M - WO (microcapsules)	WO without RE
CO (bulk oil)	CO without RE
M - CO (microcapsules)	CO without RE
WO + RE 1600 (bulk oil)	RE 1600 µg/g oil
M - WO + RE 1600 (microcapsules)	RE 1600 µg/g oil
CO + RE 1600 (bulk oil)	RE 1600 µg/g oil
M - CO + RE 1600 (microcapsules)	RE 1600 µg/g oil

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Table 2. Quality and compositional parameters of walnut (WO) and chia oils (CO) used for microencapsulation storage stability test.

Parameter[a]	WO	CO
Acid value (% Oleic acid)	0.04 ± 0.006	0.13 ± 0.003
Hydroperoxide value (meq O_2 kg-1 oil)	ND	ND
Conjugate dienes (K_{232})	1.18 ± 0.01	1.42 ± 0.05
Conjugate trienes (K_{270})	0.60 ± 0.001	0.15 ± 0.02
Oxidative stability index (h)	2.88 ± 0.22	1.14 ± 0.10
Tocopherol content (µg/g oil)		
α tocopherol	69,0 ± 4.50	ND
β tocopherol	ND	ND
γ tocopherol	539 ± 39.7	651 ± 41.9
δ tocopherol	110 ± 9.25	64.8 ± 2.68
Carotenoids (µg/g oil)	0.93 ± 0.05	5.41 ± 0.09
Chlorophylls (µg/g oil)	0.52 ± 0.02	4.66 ± 0.06
Total Phenols (µg/g oil)	ND	49.1 ± 3.96
Fatty acid composition (relative abundance)		
Palmitic acid (16:0)	7.20 ± 0.04	7.46 ± 0.12
Palmitoleic acid (16:1)	0.08 ± 0.01	0.05 ± 0.01
Heptadecanoic acid (17:0)	ND	0.13 ± 0.01
Heptadecenoic acid (17:1)	ND	0.03 ± 0.01
Stearic acid (18:0)	2.14 ± 0.01	2.98 ± 0.07
Oleic acid (18:1)	22.9 ± 0.02	7.18 ± 0.12
Linoleic acid (18:2)	52.4 ± 0.02	20.1 ± 0.13
Linolenic acid (18:3)	15.2 ± 0.03	61.8 ± 0.38
Arachidic acid (20:0)	ND	0.21 ± 0.02
Eicosenoic acid (20:1)	ND	0.06 ± 0.01
PUFA/MUFA	2.94	11.2
Iodine value	157 ± 0.09	213 ± 0.88

[a] Mean value ± standard deviation (n = 3). ND: not detected.

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Marcela L. Martínez obtained her PhD degree on Chemical Engineering at School of Exact, Physics and Natural Science, Córdoba National University (UNC), Argentina. She is Professor of Production, Security and Quality Practices, and Professor of Food Technology and Process in the post-grade master course in Food Science and Technology (UNC). Dr. Martínez is a researcher of the National Council of Scientific and Technological Research (CONICET, Argentina), conducting her research activities at Science and Technology Food Institute – ICTA (CONICET-UNC). She has published several research works, and many communications in national and international conferences.

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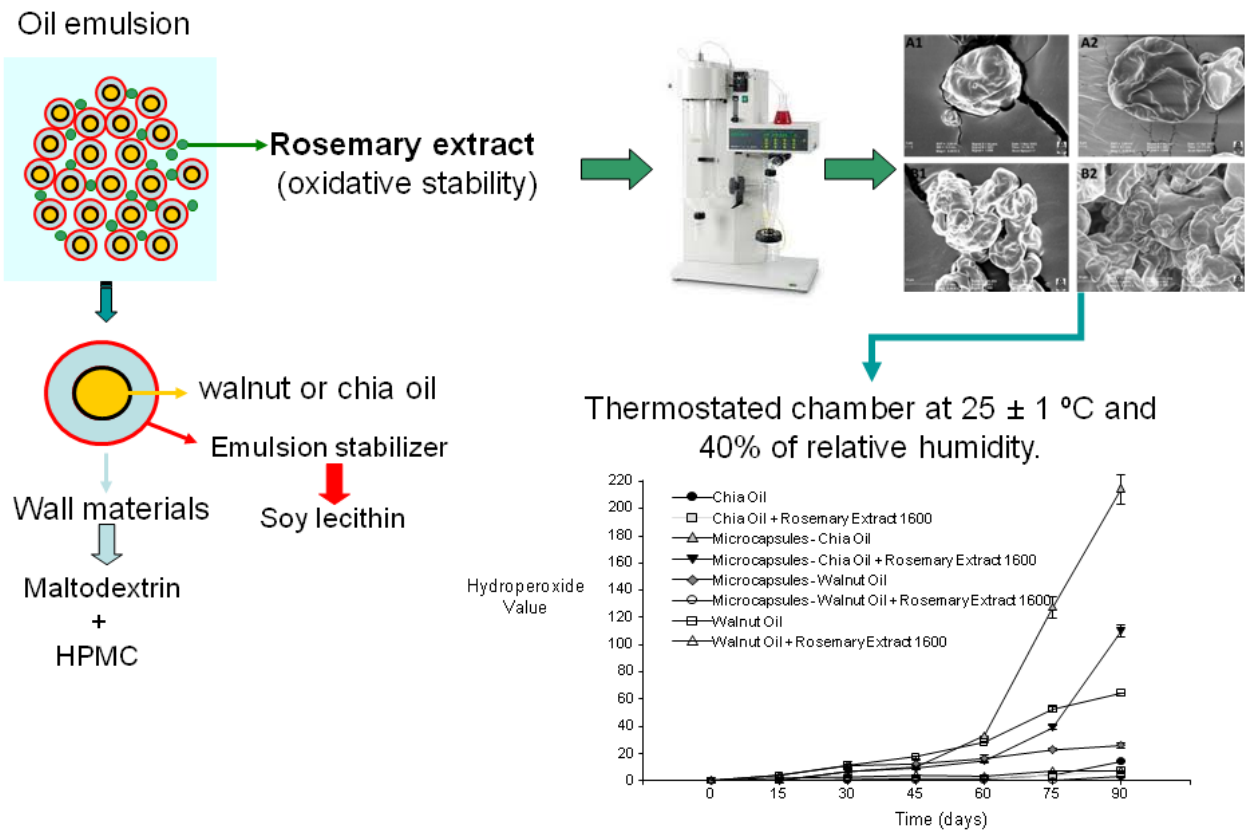
Oxidative stability of walnut (*Juglans regia* L.) and chia (*Salvia hispanica*) oils microencapsulated by spray drying



The main aim of the study was to evaluate the use of maltodextrin combined with hydroxypropyl methylcellulose as wall materials for microencapsulation of both walnut and chia oils by spray drying. Also, the effect of rosemary extract on the oxidative stability of the microencapsulated oils was examined.

Graphical abstract

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Highlights

Microencapsulation significantly protected walnut oil against oxidation.

Rosemary extract had a strong effect on walnut oil oxidative stability.

The microencapsulation technique negatively affected the oxidative stability of chia oil.

Wall matrix was not altered during the storage period.

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