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Original article

Changes in fatty acid profiles of microencapsulated fish oils prepared with citrus essential oils using a spray dryer

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Summary To decrease global fruit processing wastes, particularly citrus peel waste accounting for 50% weight of fresh fruits, have appeared as a promising and sustainable source for the industrial potential of biological valorisation into fuels or chemicals. For this purpose, this study aimed to evaluate the effects of the addition of citrus peel essential oils on the fatty acid profile of microencapsulated anchovy oil by spray drying at 0, 5, and 10 weeks of storage at ambient temperature (24 ± 1 °C). GC–MS analysis shows that the most common volatile components of lemon, grapefruit and orange essential oils were d-limonene (27.82%), limonene glycol (21.55%) and d-limonene (61.69%), respectively. Initial PUFA values were determined as 25.14%, 30.32%, 26.52%, and 30.87% for the control, grapefruit, lemon, and orange treatment groups, respectively. Although the ratio of ΣPUFA/ΣSFA was observed to vary between 0.48 and 0.94 in all groups, the ratio of Σn6/n3 was in the range of 0.10 for grapefruit and 0.58 for orange. The best citrus essential oils were grapefruit that preserved the content of PUFA in microencapsulated anchovy oil. Therefore, it is suggested that grapefruit EOs can be used effectively to preserve fish oil quality and extend the shelf life of microencapsulated fish oil.

Keywords anchovy oil, Citrus essential oil, fatty acids, microencapsulation, spray drying.

Introduction

The world food market is interested in foods with high nutritional value and health benefits. Thus, the interest in functional foods, which can be defined as those with health benefits beyond basic nutrition and contain whole, fortified, enriched or enhanced foods, has increased in the last decade due to awesome scientific evidence emphasising the significant link between diet and health. The importance of fish oil is well-known worldwide for human health and is often recommended by experts. The most important reason for this is the presence of long-chain polyunsaturated fatty acids in fish oil such as eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA). Fish oils are considered as functional supplements because they are rich in long-chain polyunsaturated omega-3 fatty acids, especially EPA and DHA. It has been stated that EPA and DHA deficiency contribute to the increased incidence of obesity, immune system disorders, hypertension, coronary heart disease (CHD), and possibly cancer (Ozyurt *et al.*, 2022). The human body cannot synthesise Omega-3 and omega-6 fatty acids. Therefore, they must be obtained from food. These fatty acids are essential for growth and development as well as maintaining health. Long-chain fatty acids form the building blocks of eicosanoids, which are stimulators of hormones such as prostaglandin. These hormone-like substances are effective in the formation of cell membranes. It is also involved in blood clotting, wound healing, and inflammation. In addition, they decrease the omega-3 triglyceride ratio, regulate blood pressure, improve heart failure, and support brain and retinal development (Maschietti & Pedacchia, 2014).The body can convert alpha-linoleic acid to long-chain EPA and DHA, but this conversion is very limited. For this reason, people need to take these long-chain omega-3 fatty acids directly from foods. Therefore, the consumption of fish oils is extremely important for human health. However, the use of fish oil in the food industry is still limited, because omega-3 fatty acids are very sensitive to oxidation. Lipid oxidation causes quality loss and deterioration of fish oil, especially in the storage and processing stages. As a result, the oxidation of oils causes an undesirable bitter taste in foods, thereby reducing the nutritional quality of

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foods. Therefore, more research is required to prevent oxidation in fish oil and, thus increasing its shelf life. In this regard, the quality of fish oils is maintained by using antioxidants and innovative processing technologies. One of the most important of these technologies is microencapsulation.

Microencapsulation is a technique in which solids, liquids, and even gases can be encapsulated in miniature closed capsules that can release their contents at controlled rates under certain conditions (Ribeiro & Veloso, 2021). The advantages of encapsulation are to isolate the core material from the degrading effects of oxygen, control the rate of separation from the microcapsule, delay the evaporation of a volatile core, improve the handling properties of sticky material. Additionally, this technique is applied for masking taste or odour, isolating a reactive core from chemical attack, and finally diluting the main ingredient material when used in very small amounts (Timilsena *et al.*, 2019). In recent years, special attention has been paid to increase encapsulation efficiency by minimising the amount of encapsulated oil on the surface of powder particles during spray drying of food flavours and oils, thus preventing volatile losses and lipid oxidation and extending the shelf life of the product (Desai & Jin Park, 2005). Although microencapsulation is an important technique for stabilising fish oils, additional stabilisation with natural antioxidants such as essential oils is necessary to provide maximum protection during processing and subsequent storage of microencapsulated bioactive components (Jeyakumari *et al.*, 2018). Therefore, researchers have been trying to characterise a number of biological properties of essential oils, including antimicrobial, antioxidant, antiviral, anticancer, anti-inflammatory, immunomodulatory, antimutagenic, and antiprotozoal activities (Durmus, 2020).

Turkey has very suitable ecological conditions and potential for citrus production, especially in the Mediterranean region. Thus, fruit processing wastes, especially citrus peel waste (accounting for 50% weight of fresh fruits) have emerged as a promising and sustainable source for biological valorisation into chemical compounds in order to reduce global fruit processing wastes. Essential oils (EO) of citrus groups are generated as a result of the citrus processing. They are considered as a natural source of flavonoids, which are one of the phenolic compounds. Recent research is generally related to the use of plant-derived antioxidants in foods and mostly in the form of adding to the raw material. However, studies on the use of fish oil microencapsulation are very limited. Thus, the current study focuses on the changes in fatty acid profiles of microencapsulated anchovy oils enriched with citrus essential oils by using a spray dryer.

Materials and methods

Fish oil

Anchovy (*Engraulis encrasicolus*) oil used as a raw material was obtained from a company located in Trabzon (Kobyalar Group Co., Trabzon, Türkiye). It was fresh and suitable for human consumption and the fatty acid profile of anchovy oil is demonstrated in Fig. S1. Sodium caseinate and maltodextrin (DE: 18–20, Alfasol, Turkey) as coating materials for encapsulation of anchovy oil (Hamasol, Turkey) were used. EOs of grapefruit, lemon and orange peels were purchased from a local company located in Adana (Biomesi Bioagrotechnology, R&D).

Preparation of emulsions

In many scientific studies, the ratio of fish oil emulsion/coating material as a coating material covers 10%–40% of the total solution on the microencapsulation of emulsified fish oils. In the current study, the treatment emulsion groups comprise of EO (2.5%), fish oil (10%), maltodextrin (10%), and sodium caseinate (10%). The rest of the solution was composed of water (67.5%). Since essential oil was not used in the control group, the water ratio was increased to 70%. Essential oil and coating materials were GRAS. The emulsion was prepared by dissolving 1:1 sodium caseinate and maltodextrin (DE:18–20) in 55 °C water for 1 h and then left them outside to cool down. Fish oil was added to this mixture and homogenised in ultra-turrax at 14 000 rpm for 10 min in a cold environment and thus the control group was formed. For the treatment groups, grapefruit, orange and lemon EOs were added to the mixture and homogenised with ultra-turrax.

Spray drying process

The emulsions were dried and pulverised in a 0.7 mm diameter spray nozzle laboratory spray dryer (Buchi Mini Spray Dryer, B-290, Switzerland). The aspiration rate was about $30 m^3/h$ and the feeding rate was 20 mL/min. The inlet temperatures was 160 °C and the outlet temperature was controlled at 90 ± 2 °C. Spray-dried microcapsules were stored in capped glass bottles at room temperature for further analysis.

Determination of volatile compounds of EOs

Analysis was determined by Agilent Brand 7890B GC, 7010B MS system. The prepared samples were injected into the DB-Wax (60 m × 0.25 mm i.dx 0.25 µm, J&W Scientific-Folsom, USA) capillary column with 1ul. Injection temperature was started at 250 °C,

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column temperature was started at 40 °C and adjusted to 240 °C by increasing 3 °C per minute. Helium (H_2) was used as the carrier gas. The electron energy was 70 eV and the mass range were 30–600 m/z. The split ratio was 1:20.

Determination of total phenolic content (TPC) in EOs

Total phenolic content of each EOs was determined according to the method of Folin–Ciocalteu (Gamez-Meza *et al*., 1999). Concentration for gallic acid standard ranges from 20 to 1000 µL and results were reported as GA equivalents (mg GAE/g). A total of 11 gallic acid standards were prepared. For the first standard, 1.5 mL Na_2CO_3, 0.5 mL Folin Ciocalteu's reagent (Merck 109 001), 20 µL gallic acid (Sigma G7384) and 7980 mL distilled water were added. Later, although Na 2 CO 3 and Folin Ciocalteu's reagent remained constant in standards, the amount of gallic acid increased and the amount of water decreased. 20 µL of each EOs was dissolved in DMSO (dimethyl sulfoxide, Merck, Germany) to yield maximum solubilisation and homogenised with the same reagents. Afterwards, the samples were kept in the dark for 2 h at room temperature. The samples were then measured at 760 nm in a spectrophotometer (Shimadzu, UV 1800).

Lipid extraction and fatty acid analysis

Lipid extraction analysis was determined according to the method of Bligh and Dyer (1959). Fatty acid methyl esters from the extracted lipid were made according to the Ichihara *et al*. (1996). On the 25 mg extracted oil sample, 4 mL of 2 M KOH and 2 mL of n-heptane were added. Then, it was mixed in vortex for 2 min at room temperature and centrifuged at 4000 rpm for 10 min and the heptane layer was analysed by gas chromatography (GC).

Statistical analysis

The data obtained at the end of the study was used with the SPSS 22.0 statistical package program and the changes due to storage between the groups were evaluated with the Duncan multiple comparison test (Duncan 1955). The level of significance was taken as $P < 0.05$.

Results and discussion

Determination of volatile compounds and total phenolic contents (TPC) of EOs

The major components of orange, lemon, and grapefruit essential oils analysed by GC/MS are demonstrated in Fig. S2. The main constituents identified in lemon essential oil are d-limonene (27.82%), carveol (11.78%), mentha (10.48%), caryophyllene oxide (6.43%), carvone (6.40%), p-menthadien-1,2-diol. (5.21%) and nootkatone (5.17%). Other compounds in low concentration are alpha terpineol (2.17%), limonene oxide (2.02%), linalool (1.92%), nerol acetate/valencen (1.39%), copaene (1.24%), limonene epoxide (1.10%), geraniol acetate/lavandulol acetate (1.05%), humulene-1,2-epoxide (1.03%), isoaromadendrene epoxide (1.09%) and less than 1% other ingredients (p-cymene, caryophyllene, δ-cadinene/cedrene, perillol, spathulenol) was detected (Fig. S2). Yazgan *et al*. (2019) reported that the main volatile compound identified in lemon oil was d-limonene with percentage of 52.85%. A study by another researcher showed that the main compounds identified in lemon oil were d-limonene (46.93%), γ-terpinene (16.89%), tri-cyclen (6.67%), 1-beta-pinene (4.69%), and 2- beta-pinene (3.86%) (Moosavy *et al*., 2017). The current study exhibited high levels of d-limonene (27.82%), carveol (11.78%) and mentha (10.48%) compounds in lemon essential oil. Franceschi *et al*. (2004) found similar results in their study and they determined that lemon essential oil contains mainly limonene (58.6%). Limonene (37.63%–69.71%), beta-pinene (0.63%–31.49%), gamma-terpinene (0.04%–9.96%) and p-cimene (0.23–9.84%) were also reported by Bourgou *et al*. (2012) as the main components in lemon oil. Özogul *et al*. (2015) demonstrated that main volatile compounds identified in lemon oil were limonene (71.77%) and β-pinene (6.11%). GC–MS analysis of the volatile components of grapefruit peel EO showed 18 compounds (Fig. S2). Limonene glycol (21.55%) was the main component. D-limonene was the second main identified compound accounting for 12.20% of the total compounds detected, which were similar to the values reported by Karioti *et al*. (2007) and Njoroge *et al*. (2005). However, Özogul *et al*. (2021) found that the main constituent of the grapefruit EO was d-limonene (82.86%). In the current study, other compounds present (>1%) include limonene epoxide (8.10%), carveol (7.55%), valencen (5.87%), beta bisabolene (5.80%), p-cymene (4.35%), carvone (4.08%), mentha (3.90%), α-bergamotene (3.20%), caryophyllene oxide (3.11%), and geraniol acetate/lavandulol acetate (3.01%). The major ingredient of orange peel oil was d-limonene (61.69%) (Fig. S2), followed by mentha (11.98%), carveol (3.44%), limonene oxide (2.99%), valencen (2.81%), linalool (2.19%), and linoleic acid ethyl ester (2.19%), respectively. It was reported that the antioxidant activity of limonene, the most important component of orange peel oil, was quite high (Roberto *et al*., 2010). Moreover, it was reported that d-limonene exhibited antimicrobial, anticarcinogenic,

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anti-inflammatory and antioxidant properties (Yu *et al*., 2017; Zahi *et al*., 2017). In general, the composition of essential oils varies remarkably with plant origin, plant parts, geographical position, climate conditions *etc*. (Si *et al*., 2012;).

Total phenolic content amounts of lemon, grapefruit and orange EOs were determined as 19.11, 25.75 and 19.58 mg GAE/g, respectively. Sir Elkhatim *et al*. (2018) found that grapefruit peels had the highest total phenolic content, followed by lemon and orange (77.3, 49.8 and 35.6 mg GAE/g peels, respectively). Similarly, in the current study, the highest phenolic content was obtained from grapefruit peels. Abeysinghe *et al*. (2007) determined that the total phenolic content of *Citrus unshiu* (Satsuma mandarin), *Citrus sinensis* (orange) and *Citrus reticulata* (mandarin) ranged from 18.5% to 38.5%.

Fatty acid composition

Fatty acids of anchovy oil used as raw material before microencapsulation are given in Fig. S1. The SFA content of anchovy oil was found to be 29.96%. As a result of microencapsulation, the SFA content of the groups increased, ranging from 32.43% to 36.19%. The MUFA content of raw anchovy oil was 20.70% which is less than those of encapsulated the samples. As for the PUFA, its content of raw anchovy oil was determined as 34.78%, which was higher than those of the treatment groups. The decrease in PUFA in microencapsulated samples may be due to oxidation of PUFAs during the spray drying process and preparation of emulsions. Özyurt *et al*., (2020) reported the initial SFA, MUFA and PUFA contents of the anchovy oil as 29.92%, 20.75% and 34.75%, respectively. However, they reported that after the microencapsulation process, these rates ranged from 34.54% to 38.31% for SFA, 21.65–23.75 for MUFA, and 26.24–30.97 for PUFA. Pourashouri *et al*. (2014) found that the PUFA content of the encapsulated oil was between 31.11% and 34.51% in fish oils in which different coating material combinations were applied, while this ratio was 35.00% in crude fish oil. EPA, DHA, and n6/n3 ratios for raw anchovy oil were determined as 8.46%, 22.43%, and 0.08, respectively. Polavarapu *et al*. (2011) reported that microencapsulated oil had slightly lower EPA and DHA percentages than crude fish oil after emulsion and spray drying. In this study, a decrease in EPA and DHA contents was determined compared to crude fish oil after the encapsulation process.

The saturated fatty acids (SFA) composition changes in all groups during storage are given in Table S1. The total SFA contents on the first day of storage were determined as 36.19%, 33.17%, 34.48%, and 32.43% for the control, orange, lemon, and grapefruit treatment groups, respectively. Statistical differences were observed among all groups and storage days ($P < 0.05$). It was reported that the SFA rates for raw anchovy were as 39.66%, 32.33% and 29.92%, respectively (Kaya & Turan, 2008; Özyurt *et al*., 2020). Studies on microencapsulation of fish oil showed that the content of saturated fat in microcapsules increases relatively after microencapsulation (Wan *et al*., 2011). In the previous study (Durmuş, 2021), an increase in SFA content during storage in microencapsulated anchovy oil with sage was observed. Similarly, Unnikrishnan *et al*. (2019) stated that encapsulate application in sardine oil caused an increase in the SFA rate. In addition, it was also reported that lipid oxidation accelerates and hydroperoxide content increases during emulsification and spray drying (Serfert *et al*., 2009). While the total SFA levels increased in all groups with the storage period, the highest increase in SFA content was determined in the orange group on the last day of storage, followed by the control, lemon and grapefruit groups. The fatty acids with the highest value among SFA were determined as myristic acid (C14:0), palmitic acid (C16:0) and stearic acid (C18:0) in all groups. On the first day of storage, the myristic acid values of the control, orange, lemon and grapefruit groups were determined as 6.33%, 5.39%, 6.34% and 5.94%, respectively. These values increased with the storage period and statistical differences were detected in all groups during the storage period. As for the palmitic acid content, the highest values were determined in the control (20.27%) and orange (19.67%) groups on the first day of storage. An increase in palmitic acid level was observed during the storage period, and the highest value was determined in the orange group of 23.61% on the last day of storage. Another prominent fatty acid was stearic acid. Its content was determined to be between 4.91% (orange and grapefruit groups) and 5.56% (the control) at the beginning of storage. Similar to other saturated fatty acids, the content of stearic acid increased with storage time.

The changes in the composition of MUFA during storage in microencapsulated anchovy oils produced using citrus EOs are given in Table S2. The highest MUFA values were determined in the control (24.46%) group on the first day of storage, followed by the lemon (23.75%), orange (22.90%), and grapefruit (22.33%) groups. In general, it was observed that the addition of citrus EOs increased the total MUFA amount and no statistical differences were observed among all groups except the grapefruit group in the last week of storage. Annamalai *et al*. (2015) stated that encapsulation application did not cause a significant change in MUFA values in encapsulation of cod liver oil. On the other hand, Durmuş (2021) reported an increase in the MUFA level during storage of

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microencapsulated anchovy oil with sage. It was determined that the palmitoleic acid rate on the first day of storage was between 4.64% and 5.39%. On the last day of storage, increases in palmitoleic acid level were observed in all groups except the control group. The main fatty acid among MUFA was oleic acid. The highest oleic acid level was determined in the control (15.11%) group on the first day of storage, followed by orange (14.84%), lemon (14.54%) and grapefruit (13.65%) groups, respectively. An increase in the oleic acid content was determined with the storage period. At the end of storage, the lowest oleic acid was determined as 15.64% in the grapefruit group. The level of vaccenic acid was found to be between 1.72% and 1.90% at the beginning of storage, and its value increased with the storage period. Considering the eicosenoic acid, the lowest value was 1.03% in the orange group at the beginning of storage. Similarly, it was observed that this value was the lowest in the orange group on the last day of storage.

The changes in PUFA composition during storage in microencapsulated anchovy oils with citrus EOs are given in Table S3. At the beginning of storage, total PUFA values were determined as 25.14%, 30.87%, 26.52%, and 30.32% for the control, orange, lemon, and grapefruit treatment groups, respectively. The highest fatty acids among PUFA were linoleic acid (C18:2n6), linolenic acid (C18:3n3), eicosapentaenoic acid (EPA, C20:5n3) and docosahexaenoic acid (DHA, C22:6n3) in all groups. The highest linoleic acid level was determined in the orange group (7.85%) at the beginning of storage, followed by the lemon (2.13%) group. Fluctuations in the level of linoleic acid were observed with the storage period. At the end of storage, the linoleic acid was 2.08% (the control), 7.34% (orange), 2.16% (lemon), and 2.15% (grapefruit), respectively. The level of linolenic acid (C18:3n3) was found to be between 1.04% and 1.18% at the beginning of storage. Statistical differences were not observed ($P > 0.05$) in linolenic acid level among the control, lemon, and grapefruit groups at the beginning of storage. Annamalai *et al.* (2015) used ginger essential oil in fish oil and also fish gelatin and maltodextrin as wall materials, showing a protective effect on PUFA content from oxidation in the fish oil encapsulates compared to the control.

Eicosapentaenoic (EPA) acid is one of the polyunsaturated fatty acids, a very important fatty acid in terms of human health and nutrition. At the beginning of storage, EPA rate was determined between 6.29% and 7.42% and these values decreased with storage period. The highest EPA value on the last day of storage was determined in the grapefruit (5.90%) group, followed by the control (5.40%), lemon (5.27%) and orange (4.05%) treatment groups. In general, towards the end of the storage, the EPA level decreased in all groups and the highest decrease was determined in the orange group.

Another important polyunsaturated fatty acid is docosahexaenoic acid (DHA). DHA level at the beginning of storage was determined as 14.81%, 15.13%, 15.87%, and 18.76% for the control, orange, lemon, and grapefruit groups, respectively. On the last day of storage, the DHA was determined as 11.58%, 8.62%, 11.47%, and 13.07%, respectively. DHA content decreased with storage and DHA level was determined as grapefruit > control > lemon > orange at the end of storage. During storage, the DHA value of the treatment groups was higher than those of the control group. It was determined that the addition of citrus essential oils preserved the DHA content in fish oil. On the other hand, Özyurt *et al.*, (2020) and Polavarapu *et al.* (2011) reported that the microencapsulation process decreased the EPA and DHA rates. Similarly, Bakry *et al.* (2016) investigated the effect of microencapsulation using peppermint oil on fatty acids of tuna oil, and reported that the level of EPA + DHA, which was 33.36% in crude oil, decreased to 16.33% after microencapsulation. In the current study, the amount of EPA and DHA, which was 30.89 in crude fish oil, decreased to 21.10%, 21.24%, 22.46% and 26.18% in the control, orange, lemon and grapefruit groups, respectively, after microencapsulation. This is could be due to temperature used in the microencapsulation process, causing oxidation.

Eicosapentaenoic acid and docosahexaenoic acid are abundant fatty acids in fish (Fan & Song, 2022). Therefore, fish are an important dietary source of EPA and DHA, which are essential for human metabolic activities (Castejón & Señoráns, 2020). In this study, the values of EPA + DHA were found between 21.10% and 26.18% for the 0th week. These two long-chain polyunsaturated fatty acids are oxidation-sensitive compounds, and their content in fish products can be altered by processing and preservation methods (Sampaio *et al.*, 2006). This explains the decrease in EPA + DHA values during 10 weeks of storage at 24 ± 1 °C. The higher values observed in the treatment groups may be due to the antioxidant activity of essential oils, which inhibited lipid oxidation in fish oil.

The changes in fatty acid indexes (ΣPUFA/ΣSFA, Σn3, Σn6, n6/n3, DHA/EPA) in microencapsulated anchovy oils with citrus EOs during storage are given in Table S4. The ratio of ΣPUFA/ΣSFA was observed to vary between 0.48 and 0.94. HMSO (1994) suggested that the PUFA/SFA ratio should be at least 0.45 for human health. According to the results obtained, it was determined that it was not below this limit value in all groups. The lowest ΣPUFA/ΣSFA value among all groups was observed in the control group. Although DHA/EPA ratios were determined as 2.35, 2.48, 2.41, and 2.22 for the control, orange,

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lemon, and grapefruit groups, respectively, at the beginning of storage, these ratios decreased at the end of storage and there were no statistical differences among the groups on the last day of storage. At the end of the storage, the highest DHA/EPA ratio was determined in the grapefruit group with 2.22, and the lowest ratio was observed in the lemon group with 2.07 at the 5th week of storage. Tatar & Kahyaoglu (2015) reported that the DHA/EPA ratio of crude and microencapsulated anchovy oil was 1.68 and 1.17, respectively. Similarly, Özyurt *et al*., (2020) determined the DHA/EPA and ∑PUFA/SFA ratios of anchovy oil microencapsulated with fish protein as wall material in the range of 2.46–250 and 0.70–0.89, respectively.

Σn3 constitutes a significant portion of the lipids in fish muscle. Although the highest Σn3 rate was observed in the grapefruit group at the beginning and end of the storage, the lowest value was observed in the control group at the beginning of the storage and in the orange group at the end of the storage. In terms of Σn6 content, the highest value at the beginning and end of storage was observed in the orange group, while the lowest values were determined in the grapefruit group for the beginning of storage and in the control group at the end of storage. While Σn3 level decreased towards the end of storage in all groups, Σn6 content tended to decrease similarly except the grapefruit group. This increase in the grapefruit group was found to be statistically insignificant between the control and lemon groups. The n6/n3 ratio of unsaturated fatty acids is associated with mortality caused by cardiovascular and cancer diseases (Monnard & Dulloo, 2021). It was suggested that this ratio could be maximum 4 (HMSO, 1994). Reduction in the ratio of Σn6/n3 fatty acids in the human diet is essential to help prevent coronary heart disease and reduce the risk of cancer (Drenjančević & Pitha, 2022). Özyurt *et al*., (2020) determined the ∑n6/n3 ratios in the samples of crude and microencapsulated anchovy oil in the range of 0.08 and 0.10–0.15, respectively. Durmuş (2021) determined that the ∑n6/n3 ratios of microencapsulated anchovy oils prepared with sage essential oil in different proportions ranged from 0.09 to 0.18. In this study, the Σn6/n3 ratio was between 0.10 (grapefruit) and 0.58 (orange), indicating within the recommended values for human health.

At the end of storage, an increase in SFA and MUFA and a decrease in PUFA were detected in all groups. Many researchers have attributed this decrease in polyunsaturated fatty acids to changes and transformations in free fatty acids. In particular, it has been attributed to the reduction of unsaturated fatty acids to peroxides and then to aldehydes, ketones, alcohols, and esters as a result of auto-oxidation reactions (Haq *et al*., 2018). In the present study, the lowest value in PUFA was determined in the control and lemon groups at the end of storage.

Due to strong flavour of essential oils, their direct use is restricted. Therefore, they can be added into another matrix, such as edible film/coatings or microencapsulation. In this study, encapsulation of fish oil with citrus essential oils masked the fishy odour. Annamalai *et al*. (2015) also microencapsulated fish oil with ginger essential oil by spray drying, providing a taste profile barrier eliminating fish oil taste and odour.

Conclusion

One of the major drawbacks of fish oil is the high content of PUFA, which is susceptible to oxidation as well as distasteful flavours. Spray-drying of fish oil is to protect it from oxidation thus it can prolong its shelf life. This study shows the suitability of microencapsulating fish oil together with citrus essential oils by spray drying with maltodextrin as wall material. Microencapsulated anchovy oil with citrus essential oils protected oil against lipid oxidation during storage. Among citrus essential oils, grapefruit essential oil gave the highest content of PUFA. Consequently, the results of the present study demonstrated that citrus essential oils, especially grapefruit can be effectively employed to maintain fish oil quality and extending the shelf life of microencapsulated fish oil.

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Author contributions

Mustafa Durmuş: Conceptualization (lead); investigation (lead); methodology (lead); writing – review and editing (lead).

Conflict of interest

The author declares that there are no conflicts of interest in this manuscript.

Peer review

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Data availability statement

All data generated and analysed during this study are presented in this article.

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Supporting Information

Additional Supporting Information may be found in the online version of this article:

Tables S1–S4 Supporting Information.

Figures S1–S2 Supporting Information.

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