

A Review: Use of Plants for Antioxidant Purposes in Fish Oil Microencapsulation

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Abstract

Fish oils, rich in essential n-3 fatty acids like EPA and DHA and constituting approximately 80% unsaturated fatty acids, play a pivotal role in contemporary nutrition. However, the susceptibility of fish oil to oxidation and its off-putting fishy smell present obstacles to its widespread consumption. Microencapsulation, a technique involving the application of protective films, emerges as a viable solution to not only enhance oxidative stability but also mitigate sensory drawbacks.

Despite the considerable global fish consumption, the daily intake of n-3 fatty acids in Western diets often falls short of recommended levels. In regions, where fish consumption is comparatively lower, the utilization of microencapsulated fish oils provides an alternative to guarantee sufficient intake. Commonly used coating materials involve proteins and carbohydrates, and the incorporation of antioxidants, be they synthetic or natural, further augments stability. Natural extracts, including rosemary, blueberry, oregano, and pomegranate peel, exhibit promise as potent antioxidants in the microencapsulation process.

This review explores various studies on microencapsulation of fish oils using different natural antioxidants. The findings highlight the potential of microencapsulation to address challenges in fish oil consumption, making it a promising avenue for improving dietary habits and promoting human health.

Introduction

The studies conducted to date indicate that many diseases in humans are caused by nutrients and dietary habits. Therefore, maintaining a healthy diet is crucial for human health. It is now well-known that diseases related to high cholesterol are largely attributed to red meat. Consequently, the consumption of foods rich in unsaturated fatty acids, which are more beneficial for health, is recommended (Kaya et al., 2004).

The importance of lipids in our diet lies in their richness in unsaturated fats. The n-3 series fatty acids play significant roles in biological and physiological processes in the body. Fatty acids are accumulated in the eyes, brain, testes, and placenta in the human body.

They contribute to the proper functioning of the eyes and brain, as well as regulating blood lipid concentrations (Kaya, 2004). High levels of n-3 fatty acids in the blood have been reported to lower triglyceride levels, increasing the risk of heart attacks. A 20-year study on urban populations revealed a decrease in heart disease-related deaths by 50% in men who consumed fish at least three times a week (Çaklı, 2007).

Due to their positive effects on human health, fish oils, containing about 80% unsaturated fatty acids, especially the essential n-3 fatty acids EPA and DHA, are considered a part of modern nutrition. n-3 fatty acids have been reported to have positive contributions to diseases such as cancer and diabetes, in addition to their beneficial effects on cardiovascular health. The British

Nutrition Foundation recommends a daily intake of at least 0.2 g of EPA+DHA for a balanced and healthy diet (Kris-Etherton, 2000).

Fish oil is highly sensitive to oxidation due to its content of polyunsaturated fatty acids, making it prone to rancidity. Oxidized fats consumed with food are reported to trigger blood clotting and arterial blockage (Turner et al., 2006). Another limiting factor for the consumption of fish oils is the fishy odor they contain, which can be unpleasant for consumers. Microencapsulation is one of the methods used to overcome these drawbacks and stabilize fish oil against oxidation.

Despite a global annual fish consumption rate of approximately 20.5 kg per capita, the daily intake of EPA and DHA in many Western diets is significantly below the recommended levels (Maki & Rains; 2012; Korkmaz et al, 2022;). Therefore, supplementing diets with microencapsulated fish oils has been considered an alternative method to ensure the intake of n-3 polyunsaturated fatty acids. The industrial demand for microencapsulated fish oils developed to meet this need is expected to be significant.

Microencapsulation refers to the process of covering a substance in very small particles with a film. Starch, cellulose, and proteins are commonly used as coating materials for this purpose. Microencapsulation has been reported to enhance the oxidation stability of fish oil, reduce the fishy odor, and make it suitable for adding to foods (Wan et al., 2011). Velasco et al. (2009) emphasized that microencapsulation is often made from proteins and/or carbohydrates, limiting oxygen permeability to protect the central material from lipid oxidation. However, they also noted the necessity of adding effective antioxidants to microcapsule emulsions to protect against quality losses in oils during processing and storage. Studies on the oxidative stability of microencapsulated fish oil have primarily focused on coating material selection, application temperature values, and drying methods, with less emphasis on the role of antioxidants (Agbashlo et al., 2012; Drusch et al., 2006; Anwar et al., 2012). A study evaluating the combined effects of phenolic components and microencapsulation on preventing lipid oxidation has also been conducted (Rubilar et al., 2012).

In practice, sodium caseinate, whey proteins, soy proteins, gelatin, modified starch, and arabic gum are commonly used as coating materials for microencapsulation. Hydrolyzed starches (glucose, lactose, corn syrup, and maltodextrin) are often added to emulsions as auxiliary coating materials (Calvo et al., 2010). The addition of antioxidants and antimicrobial substances to foods for preservation purposes can be either synthetic or natural. The reluctance towards synthetic additives due to potential harmful side effects has increased interest in studies on the use of non-chemical preservatives that exhibit antioxidant and antimicrobial effects (Gözü, 2009).

Due to their antioxidant properties, plants and spices such as hyssop (*Hyssopus officinalis* L.), laurel (*Laurus nobilis* L.), rosemary (*Rosmarinus officinalis* L.), sage (*Salvia officinalis* L.), thyme (*Thymus vulgaris* L.), clove (*Syzygium aromaticum* L.), and nettle (*Urtica dioica* L.) have mostly been used as food additives (Fernández-López et al., 2003; Gülçin et al., 2004).

Studies on Fish Oil Microencapsulation with Natural Extracts

Annamalai et al. (2015) encapsulated fish oil by coating it with fish gelatin, maltodextrin, and milk as coating materials, using ginger essential oil as an antioxidant. The researchers stated that using fish gelatin and maltodextrin as coating materials increased the encapsulation efficiency. Additionally, they reported that the addition of 0.25% ginger essential oil enhanced oxidative stability.

Li et al. (2015) conducted a study on the microencapsulation of coal fish liver oil, examining the antioxidative efficacy of blueberry extract. In their research, the addition of blueberry to the emulsion resulted in lower TBARS (Thiobarbituric Acid Reactive Substances) values (0.41 mmol/kg fat) compared to the control group (0.47 mmol/kg fat). Moreover, they found that the TBARS values of microcapsules with the extract (0.49 mmol/kg fat) were lower than those of the control group without the extract (0.57 mmol/kg fat). Additionally, after a 17-day storage period at both +4°C and room temperature, the samples with blueberry extract exhibited lower TBARS values. The researchers concluded that blueberry extract could be utilized as an antioxidant in the microencapsulation of fish oil.

Binsi et al. (2017), investigating the synergistic effect of rosemary polyphenols and gum arabic to protect fish oil encapsulates from heat-induced oxidative degradation and stabilize the capsule wall during spray drying, employed sodium caseinate and gum arabic as coating materials and rosemary extract as a coating stabilizer. The researchers noted that samples using rosemary in conjunction with gum arabic exhibited higher encapsulation efficiency and showed lower lipid oxidation during storage compared to those without rosemary. Examination of the microcapsule morphology revealed that samples with rosemary extract had a smooth surface and a spherical structure.

Jeyajumari et al. (2018) conducted a study to evaluate the potential of combining chitosan with bovine gelatin and maltodextrin as wall materials for microencapsulation of fish oil through spray drying. To enhance the oxidative stability of fish oil microcapsules, oregano (*Origanum vulgare* L) extract was added to the emulsion at 0.50 g/100 g. The spray-dried powder was reported to have a moisture content ranging from 2.8 to 3.2 g/100 g, containing spherical microparticles of different sizes, as indicated by scanning electron microscopy images. The encapsulation efficiency was reported to range between 68.94% and 81.88%. In the

study monitoring the oxidative stability of fish oil microcapsules at three different temperatures (60°C, 28 ± 2°C, and 4°C), the addition of oregano extract was noted to reduce the formation of secondary and tertiary oxidation products, as indicated by lower peroxide values and thiobarbituric acid reactive substance values compared to the control. In conclusion, the use of chitosan in combination with bovine gelatin and maltodextrin as wall materials was suggested to improve the surface morphology and encapsulation efficiency of microparticles. Additionally, the addition of oregano extract to fish oil before the commencement of spray drying was proposed to enhance oxidative stability during storage.

Another study found that the microencapsulation process of fish oil undergoes multiple changes in physical properties, such as bulkiness and dispersibility in aqueous phase and dry matrix, which can lead to autoxidation. To prevent this, an efficient stabilization was achieved using a ternary combination of antioxidants, synergistic compounds, and trace metal chelators, including rosemary extract, tocopherols, ascorbyl palmitate, lecithin, and citrem. The addition of rosemary extract significantly retarded the autoxidation of the microencapsulated oil, and the main active compounds in the extract, carnosic acid and carnosol, exhibited high antioxidative activity in the emulsified oil. Overall, the study provides valuable insights into the chemical stabilization of oils rich in long-chain polyunsaturated fatty acids during storage (Pop, 2011).

Yesilsu and Özyurt (2019) performed a study to evaluate the antioxidant activity of rosemary, thyme, and laurel extracts for the microencapsulation of fish oil and protection against heat-induced degradation. The peroxide formation kinetics of natural and commercial antioxidants (1000-1500 ppm extract and 250 ppm BHT) were investigated. The researchers reported that the peroxide values (PV) of microencapsulated fish oil with 1500 ppm rosemary were lower than the control group with commercial antioxidant (4.25 mEq O₂/kg oil vs. 3.08 mEq O₂/kg oil). Thiobarbituric acid (TBA) values for samples with 1500 ppm rosemary, 1000 ppm rosemary, and 1500 ppm laurel were determined to be 0.36 mmol MDA/kg oil, 0.56 mmol MDA/kg oil, and 0.59 mmol MDA/kg oil, respectively, compared to the control group with BHT (0.64 mmol MDA/kg oil). Additionally, the activation energies for samples with 1500 ppm rosemary and 1000 ppm rosemary were higher (31.62 kJ/mol and 30.82 kJ/mol, respectively) than the control group with 250 ppm BHT (30.46 kJ/mol). In conclusion, it was reported that rosemary and laurel extracts could be successfully applied to enhance the oxidative stability of anchovy oil through microencapsulation via spray drying.

In another study, the impact of olive leaf extract (OLE) at concentrations of 1500 and 2000 ppm under various temperatures (160, 170, and 180 °C) on the physicochemical attributes and lipid quality of fish oils throughout the microencapsulation procedure was

performed. The effectiveness of OLE as a natural antioxidant was compared with that of a synthetic antioxidant (BHT), alongside a control group devoid of any antioxidant. The group without antioxidants, subjected to spray-drying at 180 °C (C-180), exhibited the lowest microencapsulation efficiency at 70.01%. Following the microencapsulation processes, the peroxide values were comparable across all microencapsulated groups, except for the C-180 group. The saturated fatty acid (SFA) ratios closest to raw anchovy fish oil were identified in the groups with BHT and 1500 and 2000 ppm olive leaf extract dried at 160 °C. The most substantial reductions in EPA and DHA levels post microencapsulation were observed in groups lacking antioxidant supplementation, particularly those subjected to spray-drying at elevated inlet temperatures. (Özyurt et al., 2022).

In a study conducted by Trilaksani et al. (2020), tuna eye oil extracted using cold centrifugation was microencapsulated with mangrove fruit extract, maltodextrin, and arabic gum, and formulated as extrusion flake products. Mangrove fruit extract exhibited strong antioxidant activity with an IC₅₀ value. The optimal microcapsules were obtained from arabic gum-maltodextrin coating material, with the addition of 4000 ppm mangrove fruit extract, achieving an efficiency value of 93.71%, a round shape with a size of 6.26 µm and peroxide value of 97.68±0.39 (meq/Kg).

In another study, Rosemary (*Rosmarinus officianlis* L.) extracts were used as potential active compounds during the process of preparing omega-3 oil-based microcapsules to stabilize these microcapsules against oxidation and microorganisms. Various experiments were conducted to obtain effective extracts using solvents with different polarities and ultrasonication-assisted water extraction (UAE) technique. The results indicated that the water extract exhibited the highest phenolic content and radical scavenging activity (RSA %), even when compared with the synthetic antioxidant BHT. However, it was specifically noted that the radical scavenging activity of ultrasonication-assisted water extract significantly increased at a temperature of 45 °C and an extraction time of 30 minutes. Additionally, it was emphasized that microcapsules prepared with the addition of rosemary using the spray drying method were better protected against oxidation (Hamed et al., 2020).

In the study conducted by Al-Moghazy et al. (2022), fish oil, pomegranate peel extract, and probiotic bacteria (*Bifidobacterium lactis* BB12) were microencapsulated. The researchers reported that pomegranate peel extract served as an effective antioxidant, reducing the formation of both dienes and trienes. Additionally, they highlighted that the pomegranate peel extract assisted in preserving the unsaturated portion of fish oil when compared to other microcapsules without pomegranate peel extract.

Conclusion

In conclusion, the studies reviewed underscore the critical role of diet in human health, with particular emphasis on the impact of nutrients and dietary habits on various diseases. The focus on the detrimental effects of high cholesterol, primarily attributed to red meat consumption, has led to a growing recognition of the health benefits associated with the intake of unsaturated fatty acids, particularly those from the n-3 series.

The importance of n-3 fatty acids in biological and physiological processes, such as eye and brain function and blood lipid regulation, has been well-established. Despite the recognized positive contributions of n-3 fatty acids to cardiovascular health, the global and regional disparities in fish consumption, especially in Türkiye, have prompted the exploration of alternative methods to ensure adequate intake. Microencapsulation of fish oils emerges as a viable solution to address challenges such as oxidation sensitivity and fishy odor, thereby facilitating the incorporation of these health-promoting fatty acids into diets.

The literature survey reveals that microencapsulation, using various coating materials such as proteins and carbohydrates, enhances the oxidative stability of fish oil and mitigates undesirable sensory attributes. The addition of antioxidants, both synthetic and natural, further contributes to the protection of fish oil against quality losses during processing and storage. Notably, the studies demonstrate the efficacy of natural extracts, including rosemary, blueberry, oregano, and pomegranate peel, as antioxidants in the microencapsulation process, offering potential alternatives to synthetic additives.

Ethical Statement

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Author Contribution

This study is performed by single author.

Conflict of Interest

The author reports there is no competing interests to declare.

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