The Potential Application of Plant Essential Oils/Extracts as Natural Preservatives in Oils during Processing: A Review

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Abstract: The present work summarizes recent investigations carried out about the usage of natural antioxidants in lipid-rich food during processing. Synthetic antioxidants have been used as food additives to retard lipid oxidation and development of off-flavor for over 50 years. However, the literature has expressed safety concerns and health risks (toxic and carcinogenic effects) associated with the use of synthetic antioxidants recently. Natural antioxidative substances from the polyphenols of edible plants are believed to be safer and may provide with additional health benefits and more effective compared to synthetic antioxidants. Due to the fact that natural antioxidants are additives that people mixed with food and consumed for centuries, they are known to be safe by the consumer. Therefore, it is an area worth to investigate due to consumer concerns about health. In the literature, there are many studies showing that the natural antioxidants have important antioxidant effect. Plants (oil seeds, cereals, vegetables, fruits, herb and spices), compounds from animal source (peptides and amino acids), enzymes and some microorganisms are important natural antioxidants. Plant extracts have been widely used to retard lipid oxidation in foods during frying and accelerated storage processes. They were found as strong antioxidant sources due to their high contents of phenolic compounds. There are countless studies about natural antioxidants. However, they have not been investigated completely by means of toxicology.

Key words: Frying, oil, storage, stability, natural antioxidants, synthetic antioxidants.

1. Introduction

Oils have an important role in human life for healthy diet. Edible oils are known as good solvent/carrier for vitamins A, D, E and K. Besides of that they are energy source for the body. They are indispensable components of our diet due to their content of essential fatty acids.

A great demand on food prepared in a cheap and quick way due to fast increase in population, excess urbanization and economical drawbacks increases the production and consumption of the ready-foods especially fried foods. Therefore, most of the fats and oils are used in frying of the foods.

The frying is a fast, convenient and energy efficient way, therefore, it is widely used in fast-food restaurants and at home cooking. It has a special place among the other cooking methods since it increases palatability of foods due to fat absorption, crust formation and desired good flavors and odors [1]. Basically, deep-fat frying process can be described as the process of cooking foods by immersing them in an edible oil maintained at a temperature of about 150-200 °C [2]. Frying process involves both mass transfer, caused by water loss and oil uptake, and heat transfer. During the frying process, fats and oils are responsible for providing with an efficient heat transfer medium which is adapted to transmitting heat rapidly and uniformly to the surface of the food [3]. Due to high temperature, the presence of oxygen and steam released from the food, the oil
undergoes a series of chemical reactions such as hydrolysis, oxidation, thermal decomposition and polymerization [4, 5]. On the one hand, these reactions are responsible for the desired flavor, color and texture of the fried food, but on the other hand, they cause formation of undesirable products that adversely affect the consumer health. It has been proved that edible oils deteriorated excessively during frying have harmful effects on living organisms due to their high content of polar components [1, 4]. These components cause formation of toxic and cancerogenic compounds through the further reactions [6-8].

The type of oils and their oxidative stability are very important indicators in order to get idea about how long oils can be used. The choice of oil used for deep-frying should be considered by means of not only cost but also nutritional, physiological and culinary aspects. High oxidative stability, high smoke point, low foaming, low melting point and bland flavor are the important characteristics of good frying oils [9]. Low levels of trans fatty acids and saturated fatty acids that are basis of nutritional and diet physiological aspects also play important roles in selecting a frying oil [10]. Since the fatty acid composition alone is not enough to explain the stability of oils, a variety of minor components, such as tocopherols, polyphenols, phospholipids, carotenoids and certain sterols are also beneficial to oil stability during frying [2].

Edible oils are not stable products; therefore, they deteriorate through a variety chemical reactions in a short time. The most important of these reactions is lipid oxidation reaction [11]. Oxidative stability is not only a quality parameter, but also a great indicator of shelf life of oil [12]. Autoxidation of lipids starts with a free radical chain reaction that is generally initiated by exposure of lipids to heat, light, metal ions or ionizing radiation [13]. This free radical chain process which takes place at the double bond sites in lipid molecules consists of three steps: (1) initiation: formation of free radicals; (2) propagation: free radical chain reaction; (3) termination: formation of non-radical products.

When oil oxidizes, it produces a series of breakdown products in three stages. Primary oxidation products are peroxides or hydroperoxides that are formed due to acceleration of oxidation by the high temperature and the level of oxygen. Peroxides are further broken down into secondary oxidation products: alcohols, carbonyls, free fatty acids, aldehydes and ketones. The tertiary oxidation products; dimers and polymers are formed as a result of polymerization of secondary oxidation products. These products cause darkening of the oil color, formation of foam on the oil surface and an increase in viscosity of the oil [14].

Autoxidation can be inhibited or retarded by some methods such as vacuum packing, modified atmosphere packing and refrigeration/freezing [15]. Less stable liquid oils are chemically hydrogenated to enhance their oxidative stability. However, this process causes formation of considerable amounts of trans and positional isomer of fatty acids, which are nutritionally undesirable. In addition to this, the stability of polyunsaturated oils can be enhanced by physical methods such as careful blending of polyunsaturated oils with more saturated oils or antioxidants [9]. The use of antioxidants is the most preferable ways due to the cost and difficulties in the application of the methods mentioned above [15]. Antioxidants are used for the inhibition of free radical autoxidation in order to preserve polyunsaturated lipids from deterioration. Due to their lower cost, high stability and effectiveness, synthetic antioxidants such as Butylated hydroxyanisole (BHA), Butylated hydroxytoluene (BHT), Propyl gallate (PG), tert-Butylhydroquinone (TBHQ) are widely used for that purpose. On the other hand, the toxic and cancerogenic effects of them on the human health increase the use of natural antioxidants instead of synthetic ones [13].

Addition of natural antioxidants and precursors present in the plant kingdom into the frying oils is the best way of enhancing oxidative and flavor stability [9]. Natural antioxidants are considered as more potent,
more efficient and safer than the synthetic ones. Spices, herbs, teas, oils, seeds, cereals, cocoa shell, grains, fruits and vegetables are considered as sources of natural antioxidants. The herbs and spices have the highest antioxidant capacity. However, the amount that would be required per serving is extremely small but nevertheless giving a useful antioxidant boost to the meal. Studies are generally focused on plant extracts including different plant organs such as seeds, fruits, leaves and others [16]. In addition to plant extracts, essential oils, some plant oils having high stability and active components of the plants have been used as natural antioxidants for stabilizing the polyunsaturated oils.

Due to the growing interest in the use of these natural antioxidant sources in the pharmaceutical and agricultural industries besides of food industry, studies based on the natural antioxidants have come into prominence [17]. This review summarizes how the plants, essential oils of plants and plant extracts improve the stability of fats and oils during storage and/or processing at various conditions. A comparison of these potent natural antioxidants with the synthetic ones is also made.

2. Recent Studies

Recently growing interest in natural food additives especially, plant-derived ones, has decreased the considering of using synthetic antioxidants like BHA and BHT to retard the lipid oxidation in the foods. Many studies report the results of using plant-derived compounds as natural antioxidants in food products. These results led the researchers to conclude that some plant compounds could be considered as proper alternatives to synthetic antioxidants.

Plant extracts can be considered as potent sources of natural antioxidants in stabilizing the edible oils. For example, Jaswir et al. [18] investigated the effect of oleoresin rosemary extract, sage extract and citric acid on the oxidation stability of palm olein oil during frying. Results showed that all of the three extracts were effective on the lipid oxidation. Besides of reducing the oxidation rate of oil during frying, they enhanced the sensorial properties of the fried food. A combination of 0.059% oleoresin rosemary extract, 0.063% sage extract and 0.028% citric acid was found as optimum composition.

Kalantzakis and Blekas [19] investigated the antioxidant effect of Greek sage and summer savory on the thermal stability of vegetable oils (virgin olive oil, refined olive oil, sunflower oil and commercial oil blend) heated at frying temperature. According to the amount of total polar materials and $p$-anisidine values observed, the acetone extract of summer savory showed a better inhibitory effect against thermal oxidation than not only its ethanol extract but also the acetone extract of Greek sage. They have also reported that the latter extract was stronger against oil thermal deterioration than that of its corresponding ethanol extract. In other study, Bandak and Oreopoulou [20] studied the effect of the Majorana extract on the inhibition of lipid oxidation during the frying of potato in the refined corn oil at 185 °C. By monitoring changes in polar content, conjugated dienes, and peroxide value, majorana extract was observed to be an effective natural antioxidant by reducing oil oxidation during deep-frying.

Although, peels are generally thought to be underestimated as an agricultural waste, they have been found as potent sources of natural antioxidants in vegetable oils throughout the several studies. For instance, in a study [21], antioxidant efficacy of pomegranate peel extracts has been estimated in stabilization of sunflower oil. Thermal stability of methanolic extracts was evaluated by heating the extract at 185 °C up to 80 min and evaluating the antioxidant activity of extract for different intervals during storage period and exhibited 66.23% inhibition of peroxidation after 80 min heating time. Results reveal pomegranate peel to be a potent antioxidant for the stabilization of sunflower oil. In another study, Rehman et al. [22] tested the application of potato peels
extract as natural antioxidant on the soybean oil during storage at 25 and 45 °C for 60 days. In order to realize the antioxidant activity of the extract, free fatty acid, peroxide and iodine values were assessed during storage. According to the analysis results, potato peel extract exhibited very strong antioxidant activity in soybean oil during storage which was almost equal to the antioxidant activity of synthetic antioxidants (BHA and BHT). Similarly, Mohdaly et al. [23] tested the protective effects of potato peels and sugar beet pulp extracts in stabilizing sunflower and soybean oils, compared to the synthetic antioxidants, by measuring peroxide values, conjugated dienes, conjugated trienes and p-anisidine values of the oils during accelerated storage at 70 °C. At the end of the study, it was reported that potato peels and sugar beet pulp could be considered as potent sources of natural antioxidants to preserve vegetable oils against oxidation. The order of oxidative stability was found as follow: TBHQ > potato peel extract > BHT = sugar beet pulp extract > BHA.

Leafy plant extracts can be considered as effective on the oxidation of oils. For example, Shyamala et al. [24] investigated the antioxidant effect of the leafy vegetables on the fried and stored sunflower oil and groundnut oil. Four leafy vegetables; cabbage, coriander leaves, hongone and spinach were analyzed for antioxidant activity by standard methods and all of these vegetables were found as good hydroxy radical scavenger and they had excellent stability at high temperatures. In the study, Nor et al. [25] investigated the effect of Pandanus amaryllifolius leaves extract on the deep frying process with palm olein. According to the tests such as peroxide, anisidine and iodine values, free fatty acids, polar compounds and oxidative stability, natural extract was found as more effective than BHT in retarding oxidation and deterioration of palm olein. Similarly, Chiou et al. [26] studied the effect of olive tree leaves extract which is rich in polyphenols on frying of potatoes in sunflower oil, olive oil and palm oil. The extracts were found effective on the oxidative stability by monitoring total phenol content, tocopherol content, oxidative stability index, and antioxidant capacity. In another study, Arabshahi-Delouee et al. [27] investigated the antioxidant activity of drumstick leaves extracts at different concentrations in soybean oil during heating of oil for 1 hour at 180 °C. After heating test, conjugated dienes, conjugated trienes, thiobarbituric acid (TBA) and peroxide values were found lower than the control oil (without antioxidant) and the oil with BHA. As a result the extract was shown to have an excellent antioxidant activity in soybean oil compared with synthetic antioxidant BHA. Likewise, Nor et al. [28] studied the antioxidative effect of Curcuma longa (turmeric) leaf extract on the refined, bleached and deodorized palm olein during deep frying process at 180 °C for up to 40 h. According to the peroxide values determined during frying, extract was found to be effective on retarding oil oxidation and deterioration. Results also showed that the extract was more efficient than the synthetic antioxidant BHT. The good heat-stable antioxidant property of Curcuma longa was considered due to its polyphenol content leading it to be used as a good alternative to synthetic antioxidants.

Other investigation was carried out by Samotyja and Malecka [29] to test the antioxidative properties of extract from blackcurrant seeds and commercially available rosemary extracts in soybean oil. The antioxidant activity of plant extracts was compared with those of α-tocopherol and BHT. Taking primary oxidative changes into consideration, all extracts studied perform better than used standard antioxidants BHT and α-tocopherol. In another study, Ramadan and Wahdan [30] tested protective effects of black cumin seed oil and coriander seed oil in stabilization of corn oil during storage at 60 °C for 15 days. Results of peroxide values, conjugated dienes and conjugated trienes, indicated that oxidative stability of oils blended with extracts were better than the control oil. Also, Iqbal and Bhanger [31] found that the garlic extract used at high concentration showed stronger antioxidant
effect than BHT and BHA in the stabilization of sunflower oil during accelerated storage at 185 °C for 80 min.

In addition to plant extracts, essential oils can be considered as effective natural antioxidants on the edible oils during heat treatment. Du and Li [32] studied the antioxidant effect of cassia essential oil on the deep frying process of beef. The effect of cassia essential oil on five different edible oils (sunflower oil, soybean oil, rapeseed oil, palm oil and peanut oil) was investigated by measuring peroxide and TBA values as dependent variables. According to their results, the cassia essential oil was found to be effective for all of the vegetable oils, being the most effective in palm oil. Similarly, Bensmira et al. [3] studied the antioxidant effects of thyme and lavender essential oil on sunflower oil at three different frying temperatures (150, 180, 200 °C). According to the statistical analysis, a significant difference was found between oils treated with antioxidant and untreated with antioxidant. Both antioxidants prevented formation of free fatty acids and peroxides and increase in viscosity. However, there was no significant difference between the antioxidant activities of the two essential oils statistically. In other study, Özcan and Arslan [33] investigated the antioxidant effects of essential oils from rosemary, clove and cinnamon on hazelnut and poppy oils and compared them with the synthetic antioxidant BHA and the control groups. During the storage of the samples at 50 °C in darkness for 14 days, the antioxidant effect of the essential oils was determined by measuring peroxide values at regular intervals. According to the results, the essential oils showed stronger antioxidant effect compared to control groups. The effectiveness of these essential oils was ordered as: cinnamon > clove > rosemary. However, BHA was found to be more effective than the essential oils despite that it exhibited no antioxidative effect on the first few days of storage.

In some studies, oils were directly flavored with plants instead of using them in the forms of extract or essential oil. For instance, Ayadi et al. [34] investigated the effects of some Tunisian aromatic plants such as rosemary, lemon, basil and thyme on the thermal stability of olive oil. Flavored olive oils were kept in glass bottles and stored at 60 °C and 130 °C for 55 days and 6 h, respectively. The resistance to oxidation of the oils was evaluated by measuring peroxides, K232 and K270 values (indicative of the conjugation of trienes and the presence of carbonyl compounds) and change in chlorophyll, carotenoids and polyphenol contents. Despite that these aromatic plants caused a slight increase in free acidity and viscosity of aromatized olive oils, they helped to improve thermal resistance and stability of the oils. The effectiveness of these plants was ordered as: rosemary > thyme > lemon > basil. Another group, Karoui et al. [35] investigated the effect of thyme flowers instead of leaves on the thermal stability of corn oil during heating of oil for 30 min at three different temperatures (150, 180 and 200 °C) and deep-frying (180 °C). By measuring the changes in peroxide value, free fatty acid content, specific absorptivity values, color and chlorophyll, carotenoid and total phenol contents, it was shown that thyme flowers also improved the thermal stability of corn oil during heating when comparing with the control oil. It was thought that it was due to the migration of particulate compounds such as organic acids, phenolic compounds, pigments, antioxidants from thyme flowers during the aromatization process. However, Houhoula et al. [7] studied the effect of oregano in the both forms of ground spice and extract on the oxidative stability of cottonseed oil during frying of potatoes. The results showed that both additives ground spice and ethanol-derived extract could decrease the formation of conjugated dienes, polar compounds, polymerized triacylglycerols, dimeric triacylglycerols and the $p$-anisidine value of the frying oil. They estimated the storage stability of the produced chips by monitoring peroxide value and conjugated dienes in the oil absorbed. At the end of the study, oregano extract was found as more effective than ground one on the
oxidative stability of oil.

In several studies, instead of extracts and essential oils, some oils having high oxidative stability were used to slow down the oxidation. With respect to this, Chung et al. [36] studied the antioxidant effect of the roasted sesame oil on the soybean oil during frying of flour dough at 160 °C. The effect of sesame oil on the oxidative stability was investigated by determining fatty acid composition, conjugated dienoic acid, \( p \)-anisidine, and free fatty acid values. According to the test results, the sesame oil was seen to improve the thermo oxidative stability of the frying oil. By determining the tocopherols and lignan compounds in the frying oil, it was seen that contents of tocopherols and lignan compounds in frying oil had been decreased during frying. It was also realized that tocopherols protected lignan compounds in sesame oil from degradation during frying process. Likewise, Mohdaly et al. [37] tested protective effects of sesame cake extract (SCE) in stabilizing sunflower oil (SFO) and soybean oil (SBO) compared to the synthetic antioxidants by measuring their peroxide values, conjugated dienes, conjugated trienes and \( p \)-anisidine value during accelerated storage at 70 °C. It can be concluded that SCE can stabilize SFO and SBO very effectively. The SCE was also found to be a good alternative for BHT and BHA. Therefore, it can be recommended as a potent source of antioxidants for stabilization of vegetable oils. Similarly, Farhoosh and Kenari [38] investigated the effect of sesame oil (SEO) and rice bran oil (RBO) on the rancidity of canola oil during frying process of potato pieces at 180 °C. Two levels of these natural antioxidants were studied; 3% and 6%. Total polar compound content, conjugated diene value, acid value and carbonyl value were measured as deterioration parameters. According to the results, both antioxidants were found to be positively effective on the stability of canola oil. However, the frying performance of canola oil was better in the presence of SEO than RBO. The best frying performance was obtained by using 3% of both SEO and RBO together. In other study, Ardabili et al. [39] studied the frying performance of the canola oil in the presence of 5%, 10%, and 15% levels of the virgin olive (VOO) and pumpkin seed (PSO) oils during the frying of potatoes at 180 °C. To evaluate the effects of these natural additives, acid value, carbonyl value, total polar compounds and total tocopherols content of the oil samples were assessed during the frying process. According to the results, addition of both VOO and PSO improved the frying stability of the canola oil but the PSO was found as more effective than VOO due to different phenolic composition of PSO.

In some studies, the active components of the plants were also used as antioxidants. Related to this, Zhang et al. [40] tested protective effects of carnosic acid in stabilizing sunflower oil, compared to the synthetic antioxidants, by measuring their peroxide values, TBA-reactive substances, free fatty acid contents and \( p \)-anisidine value during accelerated storage at 60 °C. Results indicated that carnosic acid exhibited stronger antioxidant activity in sunflower oil than BHT and BHA. However, its antioxidant activity was less than that of TBHQ. In addition to this, Zunin et al. [41] tested the effect of addition of two different amounts of carnosic acid (0.01 and 0.1 g/100 g oil) on the oxidative stability of virgin olive oil at two different temperatures (accelerated aging temperature, 60 °C and deep frying temperature, 180 °C). A dose dependent inhibition of lipid oxidation was observed at 60 °C, whereas there was no protective effect of carnosic acid against oxidation at 180 °C. In other study, investigation carried out by Yeo et al. [42] revealed that sesamol may replace synthetic free radical scavengers like TBHQ and BHA in processed foods treated at high temperatures. Also, Erkan et al. [43] investigated the antioxidative effects of carnosic acid (one of the most active antioxidative components of \textit{Rosmarinus officinalis} L) and sesamol (extracted from sesame seeds) on sunflower oil under temperature-controlled microwave heating as a function of time and temperature (40-80 °C). The results were interpreted in
terms of antioxidative effects of carnosic acid and sesamol. Carnosic acid was found to be a more effective antioxidant than sesamol for sunflower oil.

3. Conclusions

Since food habits worldwide are mostly based on deep fried foods, oxidative−resistant oils are needed and demanded. This demand can be only conveniently met through the addition of natural antioxidants into the frying oils. Natural antioxidants recently have gained popularity since these components may contribute to the health benefit of consumers besides of the stabilization of food products. The studies having been made recently show that plant-derived materials especially, herbal extracts and their active components have an important effect on increasing the oxidative stability of vegetable oils. Using natural antioxidants increases the shelf life of the oil and food containing oil and increases the longer usage of frying oils.

In the literature, most of the studies are focused on the plant extracts due to their higher content of active components than the essential oils. However, the essential oils have been observed to have effect on the oxidative stability of edible oils, but this effect is not stronger than that of synthetic antioxidants. Studies also revealed that using plant extracts is more effective than directly addition of the plants for stabilizing the oils.

It is also realized from the studies that blending of the edible oils with the oils having higher oxidative stability is a good way for stabilizing the edible oils. But it is a doubt that these high stable oils could be considered as stronger antioxidants than the synthetic ones.

The studies based on investigating the antioxidative effects of the plants should be carried out by considering that the oxidation efficiency of the additives depends strongly on the structure of the lipid system and composition of the lipid-containing foods, besides of the processing and the storage conditions [44]. Due to the fact that the natural antioxidants have not been investigated completely by means of toxicology, the investigations related to this could be increased. When there is no doubt that the natural antioxidants have beneficial health effects and no toxic effects, the use of synthetic antioxidants in oil and fat industry will be completely terminated.

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