



Terrence Madhujith and Subajiny Sivakanthan

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T. Madhujith (✉)

Faculty of Agriculture, Department of Food Science and Technology, University of Peradeniya, Peradeniya, Sri Lanka

e-mail: [madujith@yahoo.com](mailto:madujith@yahoo.com); [tmadhujith@gmail.com](mailto:tmadhujith@gmail.com)

S. Sivakanthan

Faculty of Agriculture, Department of Agricultural Chemistry, University of Jaffna, Jaffna, Sri Lanka

e-mail: [suba106@gmail.com](mailto:suba106@gmail.com)

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## Abstract

Edible plant oils play a vital role in daily diets of people worldwide. Stability against oxidation is the major factor limiting the application of most edible plant oils for cooking and processing. Most native plant oils vary greatly in their stability to oxidation depending on their composition. Oxidative stability of edible plant oils has been extensively studied to find out the ways of improving their stability against oxidation to widen their application. Synthetic antioxidants are effective to improve the oxidative stability of these oils, however, recently, following the evidences on possible toxicities of synthetic antioxidants, the use of natural plant sources as antioxidant is gaining interest. In addition, modification of composition of the oils through genetic modification is another successful means to improve the oxidative stability of these oils. This chapter focuses on the mechanism and factors of oxidation and ways of improving oxidative stability of oils.

## Keywords

Antioxidants · Autoxidation · Hydroperoxides · Photooxidation · Radicals · Refining

## 1 Introduction

Edible oils from various sources are important components of the daily diet of people from around the world. Plant oils account for more than 75% of edible oil consumption worldwide [1]. According to the US Department of Agriculture, 189.11 million metric tons of vegetable oils have been produced globally during the season 2016/2017. Further, the world vegetable oil production is increasing continuously, especially the production of palm, soybean and sunflower oil. Palm oil found to be the major plant oil ranked highest (more than 60%) in the world production followed by soybean oil [2]. In the recent past, the use of blended oils is becoming popular in several countries such as Europe, China, Thailand, and Japan. Two or more vegetable oils are mixed at different proportion to get blended oils for different nutritional and processing purpose [3]. Vegetable oils are the healthier choice relative to animal fats in view of their fatty acid contents and cholesterol-free nature [4].

Studies on edible oils remain one of the prime areas of research, and nowadays people are much health-conscious. Diet-related noncommunicable diseases,

especially coronary heart diseases, are the major cause of death in developed as well as developing countries. In this context, edible oils play a crucial role in maintaining human well-being. Edible oils differ in their characteristics based on their composition. Oils are used as cooking oils and ingredients in variety of foods. Thus, the oils undergo various processing, mainly, heat treatment. Therefore, the stability of these oils for processing as well as storage conditions is the major property of oils. Major deteriorative reaction occurring in most of the edible oils is the oxidation. Lipid oxidation primarily depends on the fatty acid composition and the presence of minor components. Lipid oxidation leads to the production of primary and secondary oxidative compounds which are harmful to health. This chapter emphasizes the mechanism and factors of oxidation of oils, effect of processing conditions and antioxidants in oxidation, oxidative stability of some major edible oils, and measurement methods.

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## 2 Mechanism of Lipid Oxidation

Lipid oxidation is the major cause of deterioration of the quality of edible oils. Oxidative stability of oils is defined as the resistance to oxidation during processing and storage [5]. Lipid oxidation breaks down the fatty acids, thus, causes the loss of nutritional quality, and produces undesirable color, flavor, and toxic components making the food unacceptable or less acceptable by consumers; thus it is an indicator to determine the quality and shelf life [6]. Different mechanisms have been proposed as responsible for the oxidation of edible oils during processing and storage such as autoxidation and photosensitized oxidation. The kind of mechanism depends on the kind of oxygen available. Triplet oxygen ( $^3\text{O}_2$ ) and singlet oxygen ( $^1\text{O}_2$ ) can react with oxygen to cause autoxidation and photosensitized oxidation, respectively [7].

### 2.1 Lipid Autoxidation

Unsaturated fatty acids are prone to autoxidation which proceeds via free-radical chain mechanism. Rate of oxidation depends on the degree of unsaturation and increases with increase in the number of double bond [8]. Free-radical mechanism of lipid autoxidation involves the attack of oxygen at the allylic position leading to the formation of hydroperoxides. These primary oxidation products further decompose into secondary oxidation products [9].

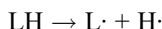
Lipid autoxidation involves three steps: initiation phase (induction period), propagation phase (exponential phase), and termination stage [10]. In the initiation step, when lipid (LH) is exposed to an initiator (heat, light or metal ions), hydrogen atom of double bond is abstracted, and alkyl radical ( $\text{L}\cdot$ ) is formed [11]. This free radical abstracts hydrogen from other lipid molecules and reacts with the hydrogen to form hydroperoxide (LOOH) (primary oxidation product) and another alkyl radical. Alkyl radical also reacts with molecular oxygen ( $^3\text{O}_2$ ) to form peroxy radical ( $\text{LOO}\cdot$ ), which abstracts hydrogen from other lipid molecules and reacts with hydrogen to

form hydroperoxide and another alkyl radical. These radicals catalyze the oxidation reaction, and autoxidation is called the free-radical chain reaction [7]. The rate of formation of peroxy radical and hydroperoxide depends only on oxygen availability and temperature [12]. Radicals react with each other to form non-radical species, and the reaction is terminated [7]. The most labile hydrogen atom in monounsaturated acids is on the carbon atoms adjacent to the double bond, whereas, in polyunsaturated acids, the most labile hydrogens are on methylene groups between two double bonds. The abstraction of hydrogen is followed by electron rearrangement to form conjugated dienes since the radicals formed are unstable [13].

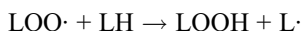
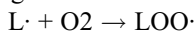
Hydroperoxides are nonvolatile, odorless, and very unstable and further break down via several steps into secondary oxidation products called carbonyl compounds such as aldehyde, ketones, acids, alcohols, acids, esters, and short-chain hydrocarbons via monomolecular and bimolecular reactions [7, 13–15]. Hydroperoxides begin to decompose as soon as they are formed. During the first stages of autoxidation, their rate of formation exceeds their rate of decomposition, and the reverse is true at later stages of autoxidation [16]. Most of the secondary oxidative products are responsible for the off-flavor in the oxidized edible oil. Aliphatic carbonyl compounds (alkanals, 2-alkenals, and *trans*, *trans*-2,4-alkadienals) have more influence on the oxidized oil flavor because of their low threshold values [7]. Breakdown of hydroperoxides also produces nonvolatile monomeric compounds, including di- and tri-oxygenated esters derived from the corresponding keto-, hydroxy-, hydroperoxy-, and epoxide esters. During induction period, only very little changes occur in lipids, followed by fast deterioration of lipids, and off-flavors become noticeable and the lipid is no longer edible [13].

Autoxidation of oleate (C18:1, n–9) produces four allylic hydroperoxides containing OOH groups on carbon 8,11 (*cis*-8-OOH, *cis*-11-OOH) and 9,10, (*trans*-9-OOH, *trans*10-OOH). Autoxidation of linoleate (C18:2, n–6) produces a mixture of *cis*, *trans* and *trans*, *trans* (9-OOH and 13-OOH) conjugated diene hydroperoxides. Autoxidation of linolenate (C18:3, n–3) produces a mixture of *cis*, *trans*, *trans*, *trans*, and *cis* (9-OOH, 12-OOH, 13-OOH and 16-OOH) conjugated diene hydroperoxides [13, 16].

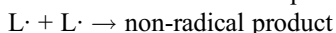
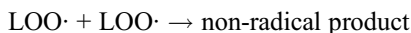
Initiation:



Propagation:



Termination:



Hydroperoxides initiate new chain reaction after reacting with free radicals [17].

## 2.2 Photooxidation

Oxidation of edible oils is accelerated by light, especially in the presence of sensitizers [7]. In the presence of light, photosensitizers such as chlorophyll and porphyrin convert triplet oxygen into singlet oxygen, which is a highly reactive non-radical molecule [15]. If wavelength of solar light is less than 220 nm, unsaturated fatty acids cannot absorb light; however photosensitizers absorb light energy and convert triplet state sensitizer to singlet-state sensitizer. Ground-state singlet sensitizers absorb light energy very rapidly and become excited singlet sensitizer which can return to their ground state via emission of light, internal conversion, or intersystem crossing. Fluorescence, heat, and excited triplet state of sensitizers are produced by emission of light, internal conversion, and intersystem crossing, respectively [7]. Photooxidation can proceed via two types of mechanism. An electron or a hydrogen atom transfers between an excited triplet sensitizer and a substrate, producing free radicals or radical ions (Type I). Excited triplet sensitizers react with triplet oxygen to produce superoxide anion by electron transfer. Superoxide anion produces hydrogen peroxide which reacts with superoxides and forms singlet oxygen in the presence of transition metals (iron or copper). Excitation energy of triplet sensitizers can be transferred to triplet oxygen by triplet–triplet annihilation to produce singlet oxygen, which reacts with the double bond of unsaturated fatty acids, producing an allylic hydroperoxide (Type II). The rate of type I and type II process may differ depending on the kinds of sensitizers, substrates, and concentrations of substrates and oxygen [7, 13, 15, 18–20]. Hydroperoxides formed by photooxidation are decomposed by the same mechanisms for the hydroperoxides formed by autoxidation. The mechanism of photooxidation is explained in detail by Choe and Min [7].

## 2.3 Lipoxygenase Catalyzed Oxidation

Lipoxygenase is an enzyme found in plants. It is another reason for the deterioration of edible plant oils, especially during oil extraction. Lipoxygenase reacts with oils containing 1,4-*cis*, *cis*-pentadiene system producing corresponding hydroperoxy derivatives and *cis-trans* conjugated hydroperoxide [21]. Reaction of this enzyme produces hydroperoxides, which decompose to form secondary oxidation products with strong undesirable flavors. Lipoxygenase produces similar volatile compounds to those produced by autoxidation [22]. Lipoxygenase contains a ferrous atom as inactive Fe (II) and is oxidized to active Fe (III) by fatty acid hydroperoxides or hydrogen peroxide. The active enzyme abstracts a hydrogen atom from the methylene group of a polyunsaturated fatty acid with the iron being reduced to Fe (II) producing conjugated dienes followed by reaction with oxygen generating peroxy radical and hydroperoxide [13, 20]. Rice bran and soybean are the two major plant sources containing lipoxygenase; thus the oils extracted from these sources can easily undergo lipoxygenase-catalyzed oxidation during processing.

### 3 Harmful Effects of Oxidation of Edible Oils

Lipid oxidation reduces the shelf life and nutritive value of food by limiting the content of essential polyunsaturated fatty acids [23]. Lipid oxidation products are considered to be harmful for health as they consist of compounds with mutagenic, carcinogenic, and cytotoxic properties [24]. These compounds cause health problems such as growth of tumor cells through lipid peroxidation as hydroxides of fatty acids are cytotoxic. Oxidation of long chain fatty acids causes neuromyopathic disease both in infant and adults [15, 25].

Consumption of lipids containing excessive free radicals may cause alterations in the redox state of human body, leading to lipid peroxidation [4]. Unstable free radicals tend to stabilize themselves by abstracting electrons from membrane lipids to start a self-propagating chain reaction causing structural rearrangement of the lipids. The rate of bond cleavage increases until the molecule gets stabilized. Oxidative damage to lipid structures eventually leads to disorganization and dysfunction of, as well as damage to membranes, enzymes and proteins [4, 26].

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### 4 Factors Affecting the Oxidative Stability of Edible Plant Oils

The oxidative stability of oil is influenced by the fatty acid composition of the oil; processing, heat or light; the concentration and type of oxygen; free fatty acids, mono-, and diacylglycerol concentration; transition metals; peroxides; thermally oxidized compounds; pigments; and antioxidants. These factors interactively affect the oxidation of oil, and it is not easy to differentiate the individual effect of the factors [7].

#### 4.1 Fatty Acid Composition

The natural fatty acid composition of oil and the positions of the fatty acids in the glycerol backbone determine the susceptibility and resistance to oxidation [27]. Oils containing more unsaturated fatty acids are oxidized more quickly than the oils containing less unsaturated fatty acids [28]. As the degree of unsaturation increases, the rate of oxidation increases [7] producing more complex mixtures of hydroperoxides. In addition, as the degree of unsaturation of fatty acids increases, the rate of formation and the amount of primary oxidation products are accumulated [29]. The relative rate of autoxidation of oleate/linoleate/linolenate was reported to be in the order of 1:40–50:100 on the basis of the oxygen uptake and 1:12:25 on the basis of peroxide development [13]. Soybean, sunflower, corn and rapeseed oils are often considered unsuitable for continuous frying due to their high content of polyunsaturated fatty acids [30]. Xu and others [31] studied the stability of camellia oil after frying potatoes by comparing with palm oil and peanut oil and reported that camellia oil possesses highest oxidative stabilities followed by palm oil, while peanut oil

showed the least stability. They have concluded that the fatty acid composition and tocopherol contents of oils are the important factors of oxidative stability of the oils. Camellia oil contains higher proportions of monounsaturated fatty acid compared to saturated and polyunsaturated fatty acid than other two oils, and the peanut oil contains higher proportion of polyunsaturated fatty acid. The monounsaturated fatty acid is relatively stable toward oxidation, while polyunsaturated fatty acids are highly susceptible to oxidation [32]. Even though the fatty acid composition of the oils is the major factor determining the oxidative stability of the oil, oxidative stability of oils is a function of several other factors [30].

## 4.2 Oil Processing

The oil processing affects the oxidative stability of an oil. All crude oils after extraction from their sources contain nontriglyceride components such as free fatty acids, partial acylglycerols, sterols, tocopherols, hydrocarbons, pigments (gossypol, chlorophyll), vitamins, phosphatides, protein fragments, trace metals, and resinous and mucilaginous materials. The objectionable nontriglycerides cause unwanted effects during processing such as darkening and formation of foam, smoke, and precipitate and develop off-flavors. Thus, crude oils are refined to remove the objectionable constituents [33]. Typical refining process consists of neutralization, bleaching, winterization, and deodorization steps. Even though the objective of the refining is to remove impurities with minimum damage to the oil, unfortunately, these processing steps may result in losses in naturally occurring bioactives such as tocopherols, tocotrienols, sterols, and phenols, and the extent of loss depends on the processing parameters and nature of the oil [33, 34]. Thus, the refined oils may have less oxidative stability than their unrefined counterparts [30].

## 4.3 Temperature and Light

Autoxidation of oils and the decomposition of hydroperoxides increase as the temperature increases. As temperature increases, the solubility of oxygen decreases drastically although all the oxidation reactions are accelerated [35]. At temperatures above 150 °C, hydroperoxides are reduced to very low level or become absent, and the formation of new compounds is very rapid, indicating that the rate of peroxide decomposition is higher than that of their formation [36].

The formation of autoxidation products during the induction period of autoxidation is slow at low temperature, and the content of polymerized compounds increases significantly at the end of the induction period. Light of shorter wavelengths has more detrimental effects on the oils than longer wavelengths, and the effect of light on oil oxidation becomes less as temperature increases [7, 12]. However, as temperature increases, the influence of light on oxidation becomes less important [35]. The effect of temperature on photooxidation is less prominent than on autoxidation. Light has more influence than temperature in photooxidation [7].

The packaging of oils is very important to minimize the photooxidation. Transparent plastic bottles can increase oil oxidation. The incorporation of UV absorbers such as Tinuvin 234 (2-(2-hydroxy-3,5-di (1,1-dimethylbenzyl)phenyl) benzotriazole) or Tinuvin 326 (2-(3'-*tert*-butyl-2'-hydroxy-5'-methylphenyl)-5-chlorobenzo-triazole) into the transparent plastic bottles can be effective to improve the oxidative and sensory stability of edible plant oils stored under light [7, 37, 38].

### 4.3.1 Frying

Frying is one of the commonly used culinary methods. In recent years, frying oils became an important component of daily diet due to rising demand for deep-fried products. However, much concern has been raised on the biological effects of consumption of fried foods containing oxidized lipids. During the frying process, oil is exposed to an extremely high temperature in the presence of air and moisture. It may result in a high rate of production and decomposition of peroxides. Deep-fat frying decreases the unsaturated fatty acids and increases polar material [39]. Thus, the quality of the frying oil is of paramount importance.

The chemistry of oxidation during frying is very complex since both thermal and oxidative reactions take place during frying at high temperatures. Reaction mechanism of thermal oxidation is principally the same as the autoxidation mechanism; however, the thermal oxidation takes place at faster rate than the rate of autoxidation [39–41]. These oxidative reactions are mainly influenced by the phenolic compounds, tocopherols, and fatty acid composition of the oil and frying temperature [4, 16, 42, 43]. Repeated frying accelerates the oxidation of oil leading to formation of primary and secondary oxidative products which are absorbed by the fried food and eventually get into the gastrointestinal tract and the circulation system after ingestion leading to health problems such as high blood pressure [4].

Juárez and others [44] investigated the discontinuous deep frying of churros in soybean oil, sunflower oil, and partially hydrogenated fats and found that, after 80.5 h of deep frying, all the oils exceeded 25% of total polar compounds except partially hydrogenated fat and losses of tocopherols during frying reached 76.0%. Marinova and others [39] investigated the oxidative stabilities of refined sunflower, grape seed, soybean, corn, and olive oils at frying temperature. Their results demonstrated that olive oil possesses better stability against thermal oxidation when compared to polyunsaturated oils. Further, it is shown that corn and soybean oils are most resistant to oxidation at frying temperature among unsaturated oils.

## 4.4 Oxygen Concentration

The oxidation of oil takes place when oxygen and catalysts are in contact with the oil. Both concentration and type of oxygen affect oxidation of oils. The oxygen concentration in the oil depends on the oxygen partial pressure in the headspace of the oil [45]. If the partial pressure of oxygen in the headspace is high, invariably high amount of oxygen becomes dissolved in the oil. The amount of dissolved oxygen is positively associated with enhanced oxidation of the oil. Prooxidants such as



transition metals and light accelerate the effect of oxygen concentration on the oxidation of oil [7, 45]. For example, addition of 70 ppm copper to rapeseed oil exposed to air for 35 days resulted in 70 times higher hexanal concentration compared to the sample devoid of copper [45]. If the oxygen concentration is sufficiently high, the rate of oxidation of oil is independent on oxygen concentrations and vice versa. Ratio of surface to volume of the oil also has influence on the oxidative stability. When the surface to volume ratio is increasing, oil can react more efficiently with oxygen; thus, the relative rate of oxidation is less oxygen-dependent with a low oxygen content [7, 45, 46].

#### 4.5 Prooxidants

Crude oils contain nontriglycerides which can act as prooxidants such as free fatty acids, mono- and diacylglycerols, metals, phospholipids, peroxides, and chlorophylls. Trace amounts of metals such as copper, iron, manganese, and nickel can be absorbed by plants during the growth and during fat and oil processing. These metals substantially reduce the oxidative stability of oils [47]. Transition metal ions are involved in redox reaction, which leads to hydroperoxide decomposition [13]. These metals' effects can be diminished by the use of chelating agents such as citric and phosphoric acids [47].

Free radicals produced during hydroperoxide decomposition act as initiators of autoxidation. Radicals generated from food contaminants may also participate in oxidation process acting as catalysts. Some molecules absorb UV light energy and are converted into an excited singlet state. Pigments such as riboflavin and porphyrins (chlorophyll, hemoglobin, myoglobin) and some synthetic dyes can act as initiators of lipid oxidation [13].

#### 4.6 Antioxidants

Antioxidants are the compounds which inhibit the oxidation of fats and oils. They extend the induction period or slow down the rate of oxidation [7]. The presence of antioxidants (naturally present or intentionally added) is one of the most important factors determining the stability of oils against oxidation.

Tocopherols, tocotrienols, carotenoids, phenolic compounds, and sterols are the naturally occurring antioxidants in plant oils. Among these, tocopherols are the most important antioxidants, especially in soybean, canola, sesame, sunflower, and corn oils. Palm oil contains a high amount of tocotrienols.  $\beta$ -Carotene is one of the important natural antioxidants present in unrefined plant oils.  $\beta$ -Carotene acts as antioxidant by light filtering, singlet oxygen quenching, sensitizer inactivation, and free-radical scavenging [7]. In addition to tocopherols, lignans are important antioxidants in sesame oils, which is well known to possess high stability against oxidation, even though it contains high level of unsaturation [46].

Non-refined oils have a better stability at elevated temperature than refined oils [30] as the refining can lead to reduction in the natural antioxidants. Szydłowska-Czerniak and Łaszewska [48] reported that the refining process of rapeseed oils decreased the antioxidant capacity by about 60% and total phenolic content by above 80%.

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## 5 Oxidative Stability of Important Edible Plant Oils

Edible plant oils can be categorized into three groups based on their fatty acid composition such as oils containing high levels of saturated fatty acids (lauric acid, myristic acid, palmitic acid, and stearic acid), oils with high levels of mono-unsaturated fatty acids (oleic acid), and oils containing high amount of polyunsaturated fatty acids (linoleic and linolenic acid). Examples for first category include coconut oil and palm kernel oil. Examples for oils with high levels of monounsaturated fatty acid include olive, canola, rapeseed, peanut, hazelnut, avocado, and sesame oils. Corn, soybean, sunflower, cottonseed, grape seed, and safflower are examples for the third category [49]. Table 1 shows the range of fatty acid found in important plant oils.

### 5.1 Coconut Oil

Coconut (*Cocos nucifera*) oil remains an important edible oil for the food industry for many years. It contains more than 90% of saturated fatty acids [51]. Coconut oil being a highly saturated oil is extremely stable against oxidation, therefore suitable for frying [52]. Lauric acid is the major saturate fatty acid present in coconut oil. The generation of *trans*-fatty acids is also very minimal during frying operations.

### 5.2 Palm Oil and Palm Kernel Oil

Palm oil and palm kernel oil are obtained from oil palm (*Elaeis guineensis*). In addition to palm oil and palm kernel oil, their fractions are also produced globally to be used for edible purposes. Palm oil is edible oil derived from the fleshy mesocarp of the oil palm fruit, and palm kernel oil is derived from the kernel of the fruit of the oil palm. Palm oil contains almost equal portions of saturated and unsaturated fatty acids, mainly as palmitic acid (42–47%) and oleic acid (37–41%). Palm oil is highly stable against oxidation [33, 53]. Palm kernel oil contains high amount of lauric acid (45–55%), thus known as lauric oil. Fractionation of palm oil into palm stearin and palm olein and palm kernel oil into palm kernel stearin and palm kernel olein further enhances their applications in foods with different stabilities [54].

**Table 1** Fatty acid composition of important edible plant oils

Fatty acid	C6:0	C8:0	C10:0	C12:0	C14:0	C16:0	C16:1	C17:0	C17:1	C18:0	C18:1	C18:2	C18:3
Coconut oil	ND-0.7	4.6–10.0	5.0–8.0	45.1–53.2	16.8–21.0	7.5–10.2	ND	ND	ND	2.0–4.0	5.0–10.0	1.0–2.5	ND-0.2
Palm oil	ND	ND	ND	ND-0.5	0.5–2.0	39.3–47.5	ND-0.6	ND-0.2	ND	3.5–6.0	36.0–44.0	9.0–12.0	ND-0.5
Palm kernel oil	ND-0.8	2.4–6.2	2.6–5.0	45.0–55.0	14.0–18.0	6.5–10.0	ND-0.2	ND	ND	1.0–3.0	12.0–19.0	1.0–3.5	ND-0.2
Palm olein	ND	ND	ND	0.1–0.5	0.5–1.5	38.0–43.5	ND-0.6	ND-0.2	ND-0.1	3.5–5.0	39.8–46.0	10.0–13.5	ND-0.6
Palm kernel olein	ND-0.7	2.9–6.3	2.7–4.5	39.7–47.0	11.5–15.5	6.2–10.6	ND-0.1	ND	ND	1.7–3.0	14.4–24.6	2.4–4.3	ND-0.3
Palm kernel stearin	ND-0.2	1.3–3.0	2.4–3.3	52.0–59.7	20.0–25.0	6.7–10.0	ND	ND	ND	1.0–3.0	4.1–8.0	0.5–1.5	ND-0.1
Palm stearin	ND	ND	ND	0.1–0.5	1.0–2.0	48.0–74.0	ND-0.2	ND-0.2	ND-0.1	3.9–6.0	15.5–36.0	3.0–10.0	ND-0.5
Palm superolein	ND	ND	ND	0.1–0.5	0.5–1.5	30.0–39.0	ND-0.5	ND-0.1	ND	2.8–4.5	43.0–49.5	10.5–15.0	0.2–1.0
Rapeseed oil	ND	ND	ND	ND	ND-0.2	1.5–6.0	ND-3.0	ND-0.1	ND-0.1	0.5–3.1	8.0–60.0	11.0–23.0	5.0–13.0
Rapeseed oil (low erucic acid)	ND	ND	ND	ND	ND-0.2	2.5–7.0	ND-0.6	ND-0.3	ND-0.3	0.8–3.0	51.0–70.0	15.0–30.0	5.0–14.0
Safflower seed oil	ND	ND	ND	ND	ND-0.2	5.3–8.0	ND-0.2	ND-0.1	ND-0.1	1.9–2.9	8.4–21.3	67.8–83.2	ND-0.1
Safflower seed oil (high-oleic acid)	ND	ND	ND	ND-0.2	ND-0.2	3.6–6.0	ND-0.2	ND-0.1	ND-0.1	1.5–2.4	70.0–83.7	9.0–19.9	ND-1.2
Sesame seed oil	ND	ND	ND	ND	ND-0.1	7.9–12.0	ND-0.2	ND-0.2	ND-0.1	4.5–6.7	34.4–45.5	36.9–47.9	0.2–1.0
Soybean oil	ND	ND	ND	ND-0.1	ND-0.2	8.0–13.5	ND-0.2	ND-0.1	ND-0.1	2.0–5.4	17–30	48.0–59.0	4.5–11.0
Sunflower seed oil	ND	ND	ND	ND-0.1	ND-0.2	5.0–7.6	ND-0.3	ND-0.2	ND-0.1	2.7–6.5	14.0–39.4	48.3–74.0	ND-0.3
Sunflower seed oil (high-oleic acid)	ND	ND	ND	ND	ND-0.1	2.6–5.0	ND-0.1	ND-0.1	ND-0.1	2.9–6.2	75–90.7	2.1–17	ND-0.3
Sunflower seed oil (mid-oleic acid)	ND	ND	ND	ND	ND-1	4.0–5.5	ND-0.05	ND-0.05	ND-0.06	2.1–5.0	43.1–71.8	18.7–45.3	ND-0.5

Source: Codex standard for named vegetable oils (CODEX-STAN 210 – 1999) [50]

ND Non-detectable

### 5.3 Olive Oil

Olive (*Olea europaea*) oil is best suited for food applications involving high temperatures such as frying. Fatty acid composition of the oils determines the suitability of oils for various applications. In this context, the fatty acid composition of the olive oil complies with the criteria of the stable healthy frying oils, that is, olive oil contains high ratio of monounsaturated-to-polyunsaturated fatty acid, low in saturated and polyunsaturated fatty acids, and most importantly very low in linolenic acid [35, 55].

Further, olive oil is considered to be a good-quality frying oil owing to its relatively low melting point; thus, the oil can drain easily from the fried food resulting in low content of trapped oil in the fried food [56].

Virgin olive oil has a higher resistance to oxidative deterioration compared to refined oils because of the presence of phenolic antioxidants such as polyphenols and tocopherols and low polyunsaturation. Polyphenols are eliminated or reduced drastically during the refining process [35].

### 5.4 Canola (Rapeseed) Oil

The edible canola/rapeseed oil is obtained from low erucic and low glucosinolate species such as *Brassica campestris*, *Brassica napus*, and *Brassica juncea*. Canola oil carries excellent nutritional value because of its unique fatty acid composition with a high amount of oleic acid (50–66%) and also contains linoleic acid (18–30%) and linolenic acid (8–12%) [33, 57]. However, canola oil has less oxidative stability because of the high amount of polyunsaturated fatty acids. Thus, canola oil is subjected to various modifications to improve their oxidative stability such as hydrogenation, fractionation, interesterification, and physical blending [57].

### 5.5 Soybean Oil

Soybean (*Glycine max* L.) oil is an important plant oil in terms of nutritional quality attributed to its high content of polyunsaturated fatty acids (n-6 and n-3) and tocopherols; however, it is highly susceptible to oxidation [58]. Soybean oil is mainly composed of polyunsaturated fatty acids such as linoleic acid (n-6) (56%) and  $\alpha$ -linolenic acid (n-3) (8%) which are considered essential fatty acids as they are necessary for growth and development of the human body. They act as precursors of prostaglandins and hormones which play an important role in the regulation of some physiological and biochemical functions of the human body [58]. However the high amount of linolenic acid reduces oxidative stability of the oil, leading to decreased shelf life [59].

Soybean oil contains relatively high amount of tocopherol compared to other major polyunsaturated oils such as rapeseed, corn, and sunflower [60]. Research

studies have shown that refined soybean oil is highly susceptible to oxidation because of its high content of unsaturated fatty acids and the absence or less amount of natural minor compounds with antioxidant effect [58].

## 5.6 Sesame Oil

Sesame (*Sesamum indicum*) oil contains mainly monounsaturated fatty acids (45–49%) and polyunsaturated fatty acid (37–41%). It contains thermally stable natural antioxidants as lignans; however, its high content of polyunsaturated fatty acids makes sesame oil highly prone to autoxidation [51].

## 5.7 Sunflower Oil

Sunflower (*Helianthus annuus* L.) oil contains linoleic (48–74%) and oleic acid (14–39%) as the major fatty acids [33]. High content of unsaturation makes the sunflower oil highly susceptible for autoxidation and less suitable for deep frying. As explained later in this chapter, new phenotypes with modified fatty acid composition, mainly with increased oleic acid content, have been developed through genetic and breeding techniques in order to make the sunflower oil suitable for thermal applications such as deep frying.

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# 6 Improving Oxidative Stability of Oils

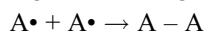
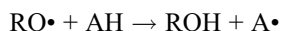
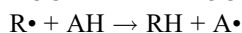
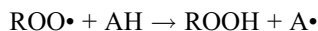
## 6.1 The Use of Antioxidants

Antioxidants are substances that when introduced into substrate at low concentration compared to that of an oxidizable substrate significantly inhibit oxidation of that substrate by inhibiting formation of free radicals or by interrupting propagation of the free radical [61, 62]. Addition of antioxidant during oil processing is one of the most effective means to retard fat oxidation. Antioxidants are of two types based on mechanism of action: primary antioxidants and secondary antioxidants. This can be further classified into natural and synthetic.

### 6.1.1 Primary Antioxidants

Primary antioxidants are chain-breaking antioxidants, that is, they are capable of neutralizing lipid free radicals by stopping their radical state by donating hydrogen. Butylated hydroxy anisole (BHA), butylated hydroxyl toluene (BHT), tertiary butylhydroquinone (TBHQ), tocopherols, and flavonoids are examples for primary antioxidants [62, 63].

The reaction mechanism can be explained schematically as follows (antioxidant molecule is denoted by AH).



Some primary antioxidants such as propyl gallate, proanthocyanidins, and ascorbic acid act as antioxidants via more than one mechanisms such as free-radical scavenging, oxygen sequestering, metal chelation, and light energy absorption [64].

### 6.1.2 Secondary Antioxidants

The secondary antioxidants (preventive antioxidant) retard the rate of oxidation through the removal of the substrate or singlet oxygen quenching mechanism [65–67]. Mechanisms of secondary antioxidants include metal chelating, singlet oxygen quenching and inactivation of photosensitizers and lipoxygenase [7]. Metals act as prooxidants by reducing the activation energy of the oxidation, especially in the initiation step, to accelerate oil oxidation. Metal chelators form insoluble metal complexes or provide steric hindrance between metals and food components. Examples include citric acid, EDTA, polyphenols, lignans, and ascorbic acid [7, 45, 64].

The mechanism of action of singlet oxygen quenchers involves deactivation of singlet oxygen to the ground-state triplet oxygen or getting oxidized themselves by singlet oxygen [7, 68]. Tocopherols, carotenoids, phenolics, and ascorbic acid retard oxidation of lipids through quenching singlet oxygen [69]. Photosensitized compounds such as chlorophyll and riboflavin transfer the energy to triplet oxygen to form singlet oxygen, or transfer an electron to the triplet oxygen to form a superoxide anion radical, which react with lipid to produce free radicals [7]. Energy of the photosensitizers is transferred to the singlet state of antioxidant to become a triplet state of antioxidant, which is changed to singlet state by transferring the energy to the surrounding or emitting phosphorescence. Carotenoids retard lipid oxidation through inactivating photosensitizers [70].

### Synergism

Addition of combinations of primary and secondary antioxidants is often reported to have synergistic effect, that is, combined action is more effective to retard lipid oxidation than the sum of their single effect and increases the length of induction period [71, 72].

### 6.1.3 Natural and Synthetic Antioxidants

Natural antioxidants are found in several plant sources such as grain, seeds, cereals, nuts, fruits, vegetables, and spices [73]. Plant extracts contain various antioxidants such as flavonoids (quercetin, kaempferol, myricetin), catechins or phenols (carnosol, rosmanol, rosamaridiphenol), and phenolic acids (carnosic acid, rosmarinic acid) [74–76]. Tocols are the natural antioxidants found in plant-based oils, which include four tocopherol and four tocotrienol isomers, each designated as

alpha, beta, gamma, and delta on the basis of the chromanol ring. The  $\alpha$ -tocopherol has been reported to be the most active isomer biologically, whereas the  $\gamma$ -tocopherol is perceived as the best antioxidant [47]. The vitamin E, most importantly  $\alpha$ -tocopherol, is the well-known antioxidant found in vegetable oil. They are known to give protection against peroxidation of polyunsaturated fatty acids [77]. Among all types of tocopherols,  $\alpha$ -tocopherol is the most unstable, thus easily destroyed at high temperatures [31]. Juárez and others [44] reported that there were significant losses of  $\alpha$ -tocopherol during frying because  $\alpha$ -tocopherol degrades more quickly at high temperatures than at room temperature [78].

Vegetable oils containing high amounts of unsaturated fatty acids such as soybean, sunflower, sesame, corn, and peanut oils are very unstable for continuous frying due to their content of polyunsaturated fatty acids. However, the presence of natural substances such as tocopherols, oryzanol, sterol fraction, squalene, among others enhances their stability at higher temperatures [30].

Several studies attempted to study the effect of addition of synthetic antioxidants to the edible plant oils. Azeez and others [67] investigated the effect of direct incorporation of TBHQ, BHT, and mixed (TBHQ and BHT) on the oxidative stability of palm olein, soybean oil, and linseed oil at room temperature and 70 °C for 168 h and reported that TBHQ had significant effect on the oxidative stability of palm olein at 70 °C, while TBHQ and BHT had synergetic effect on stability of soybean oil at room temperature and Linseed oil at 70 °C.

Gertz and others [30] investigated the efficacy of some antioxidants on stability of refined sunflower and rapeseed oils and reported that  $\alpha$ -tocopherol, tocopherol esters, and BHA exhibit low antioxidant effects at frying temperature, while ascorbic acid 6-palmitate and some phytosterol fractions were found to possess the greatest antioxidant activity.

Synthetic antioxidants, such as BHT, BHA, and TBHQ, are very effective in edible plant oils to protect against oxidation and, thus, widely used in many oils. However, recently, their use has been discouraged following findings related to possible toxicity and carcinogenicity of these synthetic antioxidants as evidenced by animal studies [79]. For example, BHA and BHT have been shown to possess carcinogenic activity in rodents [80–82]. This has drawn considerable attention to the use of natural antioxidants from plant sources as replacement for synthetic antioxidants for improving the oxidative stability of oils [58, 74].

During the past two decades, research studies have been focused extensively toward the use of natural plant extracts as sources of antioxidants to replace the synthetic antioxidants [79]. In recent years, tendency toward the use of agro-industrial by-products as sources of antioxidant is increasing [73, 74, 83, 84]. Compared to synthetic antioxidants, natural plant extracts have been shown to exhibit higher antioxidant activity and thermal stability which are the most important criteria for an antioxidant to be used for fats and oils [62, 79]. Some examples for such sources include  $\alpha$ -tocopherol, pomegranate peel [85, 86], green tea [87], olive waste [88], sesame cake [74], sesame seed [89], rosemary (*Rosmarinus officinalis* L.) [85, 90], *Eucalyptus citriodora* leaf [91], celery [92], oregano (*Origanum vulgare*) [85], cinnamon, and other spices and herbs [62]. In recent years, ascorbyl palmitate is

gaining popularity to be used in edible oils. Ascorbyl palmitate is “generally recognized as safe” (GRAS) according to the Food Drug Administration without limitation on levels to be used in food [70].

Antioxidative effect of  $\alpha$ -tocopherol (vitamin E), fat-soluble carotenoid, has been extensively studied [62]. Hraš et al. [93] studied the antioxidative activities of four natural antioxidants such as rosemary extract,  $\alpha$ -tocopherol, ascorbyl palmitate, and citric acid in sunflower oil stored at 60 °C. Among them, rosemary extract had the best antioxidative activity. Rosemary extract exhibited additive antioxidative effect when combined with citric acid and ascorbyl palmitate. Further they have reported that  $\alpha$ -tocopherol exhibited prooxidative effect. Choe and Min [7] explained that at high concentration, tocopherol radical may abstract hydrogen from lipids with very low concentration of peroxy radical and produces tocopherol and lipid radical, which may increase the lipid oxidation; thus, tocopherol act as prooxidant instead of antioxidant. Thus, the natural antioxidants may not be always preventive against oxidation [94].

Abdelazim and others [74] found that sesame cake extract possesses stronger antioxidant activity than BHT and BHA, however less than that of TBHQ. Hassani and Abdel-Razek [95] have found that addition of roasted sesame seed as a source of natural antioxidant can improve the stability of edible oils. Sesame seeds contain a myriad of natural antioxidant components including lignans such as sesamin, sesamol, and sesaminol. In addition to enhancing the stability of oils against oxidation, these compounds play a vital role in maintaining good health. Green tea extracts at higher concentrations than 200 ppm showed excellent antioxidant activity in both oils, and its efficacy was higher than that of BHA, BHT, and  $\alpha$ -tocopherol but less than that of TBHQ. Sayyad et al. [90] studied the effect of rosemary extract on thermoxidative stability of soybean oil and reported the higher stability of the soybean oil added with rosemary extract (3000 ppm) than the oil added with TBHQ (50 ppm).

Gertz et al. [30] reported that natural substances such as squalene, sterol fraction, quercetin, oryzanol, and ferulic acid are more efficient than synthetic antioxidants to enhance the stability of vegetable oils at higher temperatures. Further, ascorbyl palmitate is more efficient than BHA or  $\alpha$ -tocopherols to increase the stability of vegetable oils during frying at high temperature [30]. Alavi and Golmakani [66] reported that spirulina can improve the oxidative stability of olive oil.

Recently, a study has been reported on the antioxidant activity of encapsulated olive leaf extract in soybean oil. And their results indicated that nano-encapsulation of olive leaf could be a suitable novel technique to improve the antioxidant activity of natural sources [96].

## 6.2 Modification of the Fatty Acid Composition

Modification of fatty acid composition through natural plant breeding or genetic modification is another way of improving the oxidative stability of vegetable oils [49]. Refined edible oils such as soybean, rapeseed, sunflower, or peanut oils have



high contents of polyunsaturated fatty acids, linoleic, and linolenic acids; thus, they are not suitable for repeated deep fat frying. Even though palm kernel and coconut oils are more stable, their high content of saturated fatty acid limits their applications [97]. Genetic and breeding techniques are employed mainly to alter the composition of saturates, oleic acid, and linolenic acid. High and mid-oleic acid content can increase the oxidative stability at high cooking temperatures [98], especially during deep frying. In recent years, high and mid-oleic acid oil crops developed through breeding techniques by private companies are commercially available [49, 99]. Some examples are Nexera™ (Omega-9 canola and Omega-9 sunflower oils), Plenish™ high-oleic soybeans by E. I. du Pont de Nemours (Wilmington, DE, USA), and Vistive-Gold™ low-saturated high-oleic soybeans by Monsanto Co. (St. Louis, MO, USA) [49, 100, 101]. Some examples for oil crops developed by genetic means to contain higher oleic acid levels than normal include soybean (from 24% to 84%), palm (from 36% to 59%), canola (from 57% to 89%), sunflower (from 29% to 84%), peanut (from 55% to 76%), cottonseed (from 13% to 78%), and safflower (from 10% to 81%) [49]. Soybean phenotypes with linolenic acid less than 4% (low linolenic) and less than 2% (ultralow linolenic acid) have been developed through mutation [59].

High content of linolenic acid is the important factor responsible for the poor oxidative stability of some oils such as soybean oil. Partial hydrogenation of highly unsaturated oils can increase the oxidative stability significantly. However, the use of partially hydrogenated oils is discouraged because partial hydrogenation leads to formation of *trans* fats. Therefore, modification of fatty acid composition by breeding and genetic methods is effective [49]. New phenotypes of soybean, sunflower, corn, safflower, and rapeseed have been produced to have improved oxidative stability as well as nutritional value. High-oleic peanut oil developed through breeding has much greater autoxidation stability as compared to normal oleic peanut oil [102].

### 6.3 Blending

Blended oils are prepared by blending of different oils together. Blending is a way of modifying the fatty acid composition, physicochemical properties, and functional properties of edible oils without changing their chemical composition [89].

Okogeri [103] studied the frying stability of peanut oil blended with palm kernel oil at different ratios (90:10, 80:20, 70:30 and 60:40) and reported that all blends after used for frying contained less polar compounds than control.

### 6.4 Modifications in Oil Processing

The heat applied during the conventional oil processing technique accounts for the major loss of natural antioxidants present in edible oils. Cold-pressing oils may help retain higher levels of natural antioxidants to have acceptable shelf life

without added synthetic antioxidants [95]. Oils obtained by cold pressing are known as virgin oils, which are very popular due to their typical color, taste, and flavor. Since there is no heat treatment, most of the natural components are present in the oil [104]. It is reported that cold-pressed olive oil has a stronger antioxidant activity attributed to the presence of natural phenolic compounds [89]. Wroniak et al. [105] have reported that oil flushing with nitrogen was a very effective way to reduce the changes caused by oxidation in cold-pressed rapeseed and sunflower oil.

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## 7 Conclusion

Autoxidation and photosensitized oxidation are the main deteriorative processes that occur in edible plant oils during processing and storage. The oxidation of edible oil leads to the production of off-flavor and toxic compounds and diminishes the oil quality and shelf life. Oxidative stability of edible plant oils is thus the determining factor of the selection of suitable oil for different processing and storage methods. Oxidative stability of oils differs depending on fatty acid composition, the presence of minor components (antioxidants or prooxidants), and the processing or storage conditions, mainly, temperature, light, and oxygen. Oxidative stability of the oils can be improved by modifying the processing conditions such as the use of low temperature, exclusion of light and oxygen, removal of prooxidants, and the use of antioxidants.

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