Review Article

Comparing Synthetic and Natural Antioxidants in Vegetable Oils: Effects on Oxidation and Oil Quality

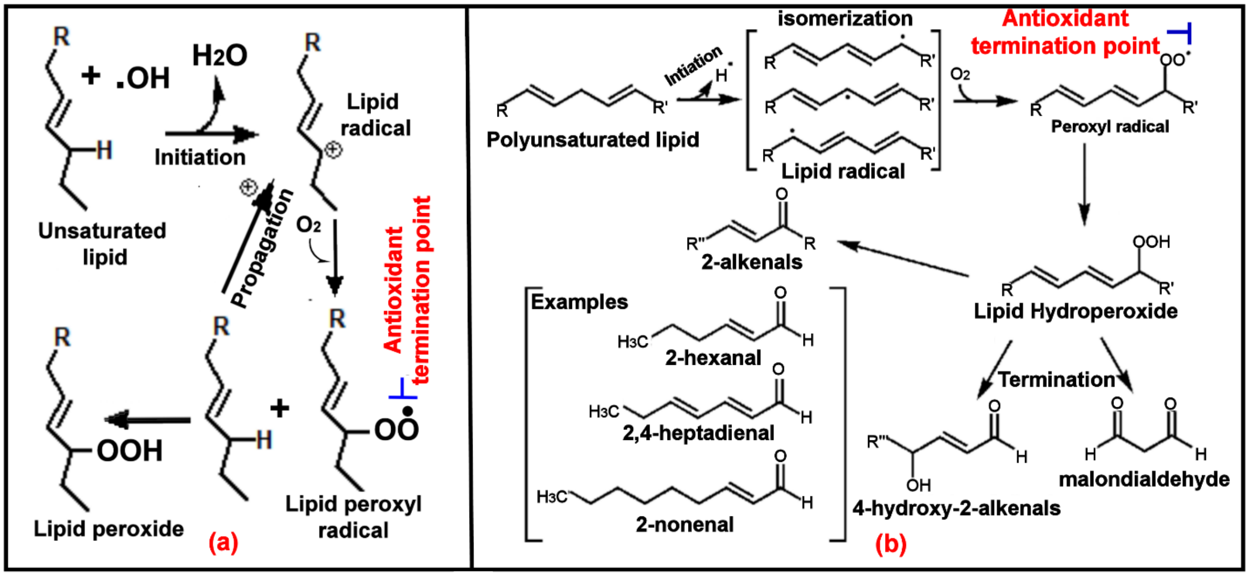
Abstract

Globally there is a huge attention towards natural products and in this case natural antioxidants. The attention has been driven by consumer demands and documented detrimental effects of synthetic antioxidants on human health. In view of this, this review has evaluated a range of scientific studies and experimental investigations providing a comprehensive analysis of oxidation related phenomena in a variety of edible oils used in the food industry. By systematically and deeply evaluating the mechanisms, effectiveness and potential limitations of synthetic and natural antioxidants, the review contributes valuable insights into the ongoing debate surrounding the application of natural and synthetic antioxidants in the food and culinary sectors. The findings of the review aids in unravelling the complex interactions between antioxidants and edible oil stability as well as highlighting the impacts of antioxidants on the nutritional quality and shelf life of edible oils. Evidence from the reviewed studies indicates that natural antioxidants may hold the solution to vegetable oils stability. This review serves as a reliable and up-to-date resource for food technologists, researchers and consumers interested in making informed decisions on selection and application of natural antioxidants in enhancing the quality of edible oils.

Key words: Antioxidants; Oxidation; Shelf life; Vegetable oils

1. Introduction

Globally there is an increase in demand of edible oils and fats for application in food, pharmaceutical, cosmetic and biofuel industry (Colombo et al., 2018). For instance between 1995 and 2011 there was a 48% increase in demand for edible oils (FAO, 2020). The edible oil market is huge with an estimation of 203 million tons by the US Department of Agriculture (USDA) in 2019 (Xiaotian, 2019). There is therefore a lot of effort to ensure the preservation of oil quality so as to stabilize the cost and sustainability of the market. Furthermore in order to meet consumer demands scientists are focusing on natural antioxidants as alternatives to synthetic antioxidants which are undesirable due to their toxic and carcinogenic effects (Bandonienė et al., 2002). Extensive studies have evaluated the potential of natural antioxidants extracted from rosemary,



flaxseed, sage, mint, sumac and thyme on enhancing oxidative stability of edible oils (Sharma et al., 2019). Evidence indicates that preventing oxidative damage of oils prevents economic problems associated with lack of usability of oxidized oils and disposal of oxidized oil (Mohammadi et al., 2016). This study aims at reviewing literature and further compare the potential of natural antioxidants with synthetic antioxidants in enhancing quality of edible oils.

Edible oils are very highly susceptible to deterioration through oxidation and microbial degradation resulting to nutrition losses and development of off flavors. Quality deterioration of oils results to formation of oxidation products that are reactive and toxic which ultimately possess as a health risk due to their potential of causing cancer development and inflammation (Negash et al., 2019). Among the deterioration changes in edible oils is the development of rancidity.

Rancidity is a chemical process that involves oxidation or hydrolysis of oils/fats on exposure to moisture, air, light or bacteria giving rise to products such as ketones, aldehydes or free fatty acids (Kuhnert, 2016) (**Fig. 1**). The products are unpleasant in taste and odor. There are three types of rancidity: oxidative, hydrolytic, and ketonic rancidity (Anand, 2021). Oxidative rancidity occurs when oil/food is exposed to oxygen, light and/heat. The oxidative products of this process are

peroxide derivatives that are toxic with a bad smell. The peroxide that arises out of oxidative rancidity degrades vitamins A and E in foods (Anand, 2021). Other products of oxidative rancidity include polymeric materials and oxidized sterols (Bilancia et al., 2007) **(Fig. 1).**

Figure 1:Free radical mechanisms of oxidative rancidity of (a) unsaturated and (b) polyunsaturated fats (Kuhnert, 2016).

Hydrolytic rancidity occurs when fats are hydrolyzed by lipase from bacteria at high temperature and pressure. High pressure and temperature will lead to the denaturation of bacterial lipase. It can also occur as a chemical reaction when foods are cooked at high temperatures (frying) whereby the ester bonds of the triglycerides are broken down, giving rise to free fatty acids and other products of undesirable flavors. This problem is common with fast foods restaurant that use frying oils for a long time (Zeece, 2020).

Ketonic rancidity occurs due to contamination with fungi such as *Asperigillus niger* and it is catalyzed by moisture (Hatton, 2017). This kind of rancidity only occurs in lauric acid oils and butter fats wherein the length of intermediate and short carbon chains (specifically carbon 14 and 6) is converted into methyl ketones. It arises out of fungi contamination which can be avoided unlike hydrolytic rancidity that occurs at high temperature (factors easily controlled on storage – *store in a cool dry place*). As such, the rancidity process that affects edible oils and makes them unfit for consumption during storage is oxidative rancidity (Esfarjani et al., 2019).

Monounsaturated and polyunsaturated fats (liquid at room temperature) are healthier than saturated fat (solid at room temperature) (Matthäus, 2010; Mattson & Grundy, 1985). As such, there is a preference by consumers for the mono and polyunsaturated fats (Matthäus, 2010). Whereas saturated fats are less susceptible to rancidity, mono and polyunsaturated fats are more prone (Choe & Min, 2006).The susceptibility arises due to weaker hydrogen-methylene bond strength in the fatty acid fragment (Choe & Min, 2006). Rancidity products can react with other components in the food like the amino acids or proteins leading to nutritional loss. Consequently, changes in color, viscosity, and solubility occur. Most importantly, loss of essential fatty acids could also take place (Omwamba, Artz, & Mahungu, 2011). The by-products of rancidity have also been linked with health concerns such as cancer, malformations during pregnancy, and a rise in blood pressure (Grootveld et al., 2006; Perumalla & Subramanyam, 2016; Vikeira et al., 2017). Therefore, oxidative rancidity is a significant factor with regard to nutritional value, quality of edible oils, shelf stability of the oils and generally affects the economics of the oils.

The oxidation process cannot be completely stopped. Once it has begun, it can only be retarded or minimized (Kamal Eldin, 2010). Recent trends, which aim to improve the storage stability of edible oils by shifting fatty acid alignment from unsaturated to increased amounts of monounsaturated fats, have been associated with significant nutritional loses (Matthäus, 2010). Preventing contact with factors such as temperature, oxygen, and metal traces has proven inefficient and uneconomical (Greyt, 2013). The most effective way to delay oxidation has been the use of antioxidants (Santos et al., 2004). As such, we review these antioxidants in detail and critically identify research gaps for attention by researchers. The work compliments other reviews in the subjects including: Machado et al., 2022; Lozano-Castellón et al., 2022; Fadda et al., 2022). However, the current work is unique in that it links the possibility of using natural antioxidants to solve the aforementioned challenges that arises when artificial antioxidants are used.

1.2. Use of antioxidants in oils

Most antioxidants function by reacting with radicals from the free radical chain mechanism to form more stable compounds (Taghvaei & Jafari, 2015). Others function by destroying the hydroperoxides formed, scavenging oxygen or by synergism. Artificial antioxidants like butylated hydroxytoluene (BHT), tert-butylhydroquinone (TBHQ), Citric acid and vitamin E (**Table 1**) have been used over time mainly because of their great stability, performance, and affordability. Nevertheless, their safety has always been controversial (Lourenço et al., 2019). Continued intake of artificial antioxidants has been associated with well-being issues among them skin reactions, gastrointestinal tract complications and in some cases increased risk of cancers (Botterweck et al., 2000; Choe & Min, 2006; Engin et al., 2011; Randhawa & Bahna, 2009).

Recently, researchers have tried to improve oil storage stability by altering the alignment of fatty acids: from polyunsaturated to advanced quantities of monounsaturated fatty acids. Such interventions result in nutritional losses and therefore, antioxidants remain the most effective technique to delay oxidation.

Table 1: Antioxidants in various edible oils (Santos et al., 2004).

Vegetable oils	**Fatty acids**			
	Monosaturated	Polyunsaturated	Saturated	Artificial antioxidants
Olive	71.3	12.7	16.0	NA
Rapeseed (A)	65.2	29.3	5.5	Citric acid/Vitamin E
Rapeseed	65.0	29.0	5.0	NA
Sunflower (A)	22.8	65.2	12.0	Citric acid/Vitamin E
Sunflower	23.0	65.0	12.0	NA
Corn (A)	33.5	51.0	15.5	Citric acid/TBHQ
Corn	34.0	50.0	16.0	NA
Soybean	24.3	60.0	15.7	Citric acid/TBHQ
Rice	40.8	40.1	19.1	NA

(NA) Absence of artificial antioxidants
TBHQ: tert-butylhydroquinone

2. Shelf life of vegetable oils

Vegetable oils are an important part of people's everyday diets all around the world. According to data from the United States Department of Agriculture, around 189.11 million tons of plant oils are produced globally (Madhujith & Sivakanthan, 2019). In the recent past, the world's vegetable oil manufacture has been surging incessantly especially for palm, soybean, and sunflower. Vegetable oils have a wide range of properties that are largely determined by their composition. As a result, these oils are utilized as ingredients in a wide range of dishes and food processing.

Oxidation is a major quality-degrading event in edible oils (Choe & Min, 2006). The oxidative strength of edible oils is determined by the raw material used, the processing procedures and the storage circumstances. Triacylglycerides, which are made up of different fatty acids make up around 96 percent of vegetable oils (Orsavova et al., 2015). Fatty acids, whether coupled to glyceride or free are prone to oxidation, which results in a variety of breakdown products (Omwamba et al., 2011). The most obvious characteristic change is the growth of unfriendly flavor and scent. Other variations include changes in viscosity, appearance, and solubility. Consequently, there is the destruction of vitamins and their precursors, loss of vital fatty acids, and development of odor-intensive combinations. These changes in the long run influence the sensory and nutritional value of vegetable oils (Matthäus, 2010).

2.1. Mechanism associated with oxidation of lipids in edible oils

High temperature, oxygen accessibility, and the availability of light and trace metals all influence the oxidation vulnerability of edible oils (Choe & Min, 2006). Therefore, the most imperative task in their production and distribution is to keep these features at a minimal to a point where no unfavorable variations are projected over a certain period (Greyt, 2013).

Hydroperoxides, which are odorless and tasteless, are the major results of lipid oxidation. The hydroperoxides undergo a sequence of reactions to create aldehydes, ketones, hydrocarbons, alcohol and lactones due to their high instability (Kerrihard et al., 2015). These products become very obvious in the oil once formed. The main pathway for the development of hydroperoxides is autoxidation yet still irradiation, enzymatic oxidation and photo-oxidation are also possible (Matthäus, 2010). The mechanisms of these pathways are technically similar although they differ in radical formation.

2.1.1. Autoxidation

Oxygen and unsaturated fatty acids either free or bound in triacylglycerol nanoparticles are the key participants in this reaction. It is a free fundamental sequence reaction that takes place in four phases; the starting phase, proliferation, chain branching and conclusion phase.

The initiation stage starts with the removal of a hydrogen atom from an integral lipid molecule to produce a radical, which is required to begin the chain reaction. A hydroperoxide radical is formed when the radical reacts with triplet oxygen (Kerrihard et al., 2015). This reaction occurs within a very short time since the fatty acid radical is unstable and does not require activation energy for the process to happen. The mechanism is also heavily reliant on the availability of oxygen as well as the oxygen concentrations in the oil. The concentration of oxygen in the oil decreases as the temperature rises. The generation of peroxy radicals is slowed or stopped as the solubility of oxygen in the fat decreases. Without the availability of oxygen, other reactions such as polymerization take over (Ahmed et al., 2016; Kamal Eldin, 2010).

Owing to the high reactivity of the peroxy radical, it takes up a hydrogen ion from another unsaturated fatty acid resulting in the formation of lipid peroxides, the main product of the edible oils breakdown process (Omwamba et al., 2011). The chain reaction continues after the very first stage, when the freshly formed radical reacts with triplet oxygen and takes up hydrogen from a separate unsaturated fatty acid resulting in the formation of more hydroperoxides (Ned, 2013). The autoxidation process then goes on exponentially.

Initially, the formation of hydroperoxides is slow then later accelerates to a noticeable level, a level known as the induction period (Šimon et al., 2000). The presence of various ions such as copper and iron speeds up the oxidation reactions (Omwamba et al., 2011). Due to the varied bond power of the hydrogen -methylene group in the fatty acid particles, the vulnerability of fatty acids to hydrogen removal is highly dependent on their level of unsaturation (Juan & Maria, 2003). The peroxy radical always removes the fatty acid molecule's weakly attached hydrogen. While hydrogen has a binding power of around 99 kcal/mol in saturated fatty acids, only about 80 kcal/mol is required to extract a hydrogen molecule from a methyl group in oleic acid and only 69 kcal/mol is required to isolate hydrogen from the double allylic methylene group in linoleic acid (Matthäus, 2010; Ying et al., 2018). By nature, it is 40 kcal/mol for linolenic acid which has two extra allylic methylene groups. As a result of different fatty acid particles having varied hydrogen bond strengths, they oxidize lipids at different speeds (Hongyan et al., 2013). Conclusively, this means that edible oils with increased concentrations of unsaturated fatty acids are subject to faster autoxidation during storage compared to fats with monosaturated and saturated fatty acids.

2.1.2. Photo-oxidation

Photo-oxidation can also activate the fatty acid particle and it occurs in two types (Porter, 2013): type I involves light that activates the catalyst, which then transmits energy to the fats, resulting in the formation of a reactive species that can react with triad oxygen; type II involves the excitation source combining with triad oxygen to generate receptive molecular oxygen, which subsequently combines with the light -activated fatty acid molecule. Fatty acid reactions are 1500 times faster with singlet oxygen than with triplet oxygen (Choe & Min, 2006). With no further stimulation, singlet oxygen reaction with the double bonded fatty acids can occur (Hongyan et al., 2013; Juan & Maria, 2003). This, therefore, means that oxidative deterioration occurs rapidly since it can progress without an induction period.

It is not possible to slow down oxidation in this case by use of antioxidants since in type II reactions there is no radical formation. However, the reaction can be inhibited by quenchers which take up the initiation energy of light minus formation of any reactive molecules (Choe & Min, 2006).

2.1.3. Irradiation

Irradiation can occur through the following highlighted stages (Ahmed et al., 2016; Madhujith & Sivakanthan, 2019; Matthäus, 2010). Direct production of electrons from fats by removal of hydrogen from a-linolenic acid's allylic methylene, since depolarization intensity is 105 times greater than the energy necessary for extraction. Production of additional radicals, such as peroxides by electrolysis of water which then extracts hydrogen from the allylic methylene species. The fatty acid's integral part then subsequently reacts via hydroperoxides free radical chain mechanism.

2.1.4. Enzymatic oxidation

Enzymatic oxidation by the enzyme hydroxylase, which belongs to the oxidoreductase category, also produces hydroperoxides. This enzyme is found in almost all living cells and catalyzes the reaction between oxygen and unsaturated fatty acids to produce peroxides. Unrestricted fatty acids are their primary source of energy. A few of them use triacylglycerides as a substrate as well, albeit with lower specificity.

2.2. Influence of oxidation on edible oils

2.2.1. Creation of secondary reaction products

Hydroperoxides are not so obvious to the consumer since they are tasteless and odorless. Quality variations only become apparent after the formation of aroma-active compounds (Matthäus, 2010; Porter, 2013). The amount and kind of chemically generated substances are determined by the fatty acid configuration. While hexanal is the main result of linoleic acid degradation, Trans, trans-2, 4-heptadienal are also produced from linolenic acid (Greyt, 2013). As a result, the rapid degradation of vegetable oils with high levels of linolenic acid is not only due to this fatty acid's high oxidation vulnerability but also the very low threshold values of the secondary aroma compounds generated from the decomposition of hydroperoxides (Madhujith & Sivakanthan, 2019; Matthäus, 2010). The decomposition of hydroperoxides can happen naturally or in the existence of metal ions.

2.2.2. Reduction in sensory quality during storage

This is the key and the most noticeable effect of oxidative deterioration. Generally, the negative characteristic of the oil is described as rancid which means ‘unfavorable, 'stinky' or 'nasty' (Khan et al., 2011). The rancid sensation perception is varied dependent on the oil's fatty acid structure and the degradation products that arise.

The green, beany and grassy smell of soybean oil during the primary packaging stage which quickly transforms to painty or fishy, is a good example of off-flavor development. This green-beany flavor is present in unfinished soybean oil but it is eliminated during purification, resulting in a pleasant and odorless oil (Greyt, 2013; Vieira et al., 2017). During storage however, some oils develop distinct off-flavors such as "animal flavor" in butter and tallow, "fishy" in canola, "grassy/painty" in oilseeds and soybean and "painty/rancid" in coconut oils (Villarino et al., 2007).

2.2.3. Effect on nutritional quality

Edible oils are a good source of essential fatty acids like linoleic and linolenic acid, as well as vitamin E active combinations like tocopherol and tocotrienols (Aladedunye & Przybylski, 2009). Oxidation leads to degradation of these compounds and if they were the single origin of nutritional lipids and vitamin E in the diet, then there will be a deficiency (Aladedunye & Przybylski, 2009; Vieira et al., 2017). Moreover, there is a reduction in amino acid availability in a protein-rich food prepared using oxidized oil as a result of reactions involving lipid degradation products (Madhujith & Sivakanthan, 2019; Omwamba et al., 2011).

A higher intake of oxidized oils causes a faster turnover of vitamin-E-active composites to maintain the body's immune system, which increases the need for vitamin-E-active fibers hence disrupting the antioxidative defense framework (Burton & Traber, 1990; John et al., 2001).

2.3. Protecting edible oils from oxidation

2.3.1. Modification of the fatty acid composites

This is done through genetic modification or natural plant breeding to improve the oxidative stability of the oil (Richard, 2012). Purified vegetable oils for instance peanut, soybean or sunflower oils possess increased concentrations of levels of unsaturated linolenic and linoleic acids; hence not recommended for recurrent high temperature cooking. Despite some oils like palm kernel oil being more stable with regard to oxidation, their usage is limited by high levels of saturated fatty acids (Sakurai et al., 2011) and therefore chromosomal and breeding methods are used to modify and change the saturates structure. High and mid-oleic acid content improves the oxidation endurance of oils at high cooking temperatures, such as pan-frying (Lee et al., 2018).

Mid and high-oleic acid oil yields have recently been established through breeding techniques; breeds involved among them are Monsanto Co.'s NexeraTM (Omega-9 canola and Omega-9 sunflower oils) and Vistive-GoldTM low-saturated high-oleic soybeans (Bellaloui et al., 2015; Kaushik & Grewal, 2017; Richard, 2012).

Oil crops resulting from genetic modification contain high quantities of oleic acids for instance; Soybean from 24 percent to 84 percent, palm from 36 percent to 59 percent, canola from 57 percent to 89 percent oleic acid, sunflower from 29 percent to 84 percent oleic, peanut from 55 percent to 76 percent oleic acid as well as cottonseed from 13 percent to 78 percent oleic acid (Richard, 2012). Scientists have also used mutation to create soybean phenotypes with linolenic acid levels of less than 4%, referred to as low linolenic and less than 2% referred to as ultra-low linolenic (Clemente & Cahoon, 2009). This is important because the high content of linolenic acid is a major contributor to the poor oxidative stability of some oils, such as soybean oil. The oxidative stability of highly unsaturated oils is greatly improved by partial hydrogenation. However, because of the creation of Trans fats, it is prohibited; thus, genetic and breeding strategies for modifying fatty acid composition remain useful (Richard, 2012). This not only increases the oils' oxidative stability but also their nutritional value.

2.3.2. Modifications involving processing of oil

The loss of antioxidants naturally occurs in edible oils largely due to the heat used in traditional oil processing procedures. With no additional antioxidants, the cold-pressing procedure allows oils to maintain high quantities of antioxidants and have a longer shelf life (Hassanein & Abdel-razek, 2012). Virgin oils are edible oils derived through cold-pressing and are well known for their particular taste, color and flavor. Most of the oil's natural ingredients are preserved due to the lack of heat treatment.

Cold-pressed olive oil according to (Abdel-razek et al., 2010), has a higher antioxidant activity due to the presence of natural phenolic components. Additionally, flushing sunflower and rapeseed oil with nitrogen during processing is important as it lowers the level of oxidative changes (Wroniak et al., 2016).

2.3.3. Blending

Blending is basically the combination of several oils. This method combines the excellent features of two separate oils to provide a compounded influence on the oil's quality. It produces oils with an altered fatty acid content as well as functional and physicochemical qualities while leaving the chemical makeup unchanged (Abdel-razek et al., 2010. Okogeri, (2015), investigated the frying durability of peanut oil combined with crude palm oil at varying percentages (90:10, 80:20, 70:30, and 60:40) and found that all mixtures had fewer polar molecules after frying than the reference. When weighing the benefits and drawbacks of using a single oil as a cooking medium, blended oils have been demonstrated to be more suited than solitary oils while also being more cost-effective.

2.3.4. Application of antioxidants

Antioxidants are compounds that, when added into a target substance in small quantities, limit oxidation by decreasing free radical generation or halting oxygen radicals' dissemination (Bansal et al., 2013; Bera et al., 2006). Antioxidant application during oil processing is one of the most effective and feasible methods for reducing oxidation in edible fats and oils. Primary antioxidants and secondary antioxidants are divided into two categories based on their method of action. They are further divided into natural and synthetic varieties.

2.3.4.1. Primary antioxidants

Primary antioxidants, also characterized as chain-breaking antioxidants can destroy lipid-reactive oxygen species by contributing hydrogen to prevent them from ever becoming reactive. Primary antioxidants include butylated hydroxyanisole (BHA), butylated hydroxyl toluene (BHT), tertiary butyl hydroquinone (TBHQ), tocopherol and flavonoids (Bansal et al., 2013; Dimitrios, 2006).

2.3.4.2. Secondary antioxidants

Secondary antioxidants decrease oxidation using the singlet oxygen quenching strategy (Ana et al., 2015; Bente et al., 2002). Metals ions function as pro-oxidants, inhibiting oxidation by reducing the amount of energy of the electrons, specifically in the initiation phase. Citric acid, EDTA, polyphenols, lignans and ascorbic acid are examples of metals that form insoluble metal complexes or offer steric hindrance between metals and dietary components (Hussain et al., 2015; Jiang & Youling, 2016).

Singlet oxygen quenchers work by deactivating singlet oxygen and converting it to ground-state (Rather et al., 2016). Tocopherol, carotenoids, phenolics and ascorbic acid quench singlet oxygen hence slowing lipid oxidation (Jie et al., 2012).

Light-sensitive substances like riboflavin as well as chlorophyll interact with triplet oxygen to produce singlet oxygen or a superoxide anion reactive that combines with fats to produce reactive oxygen species (Orsavova, 2015). Combining primary and secondary antioxidants has proven more effective than the effect of a single antioxidant due to their synergistic effect which increases the length of the induction period (Bente et al., 2002; Fernandes et al., 2019).

2.3.5. Synthetic antioxidants

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Due to their efficacy BHT, BHA and TBHQ are still the most extensively utilized antioxidants in the culinary oil business. Several studies have looked into the effects of synthetic antioxidants added to edible plant oils. Bente et al., (2002), studied the effect TBHQ, BHT and a merge of TBHQ and BHT on the antioxidant potential of palm oil, soybean oil and linseed oils at ambient temperature and at 70°C for 168 hours and found that TBHQ greatly improved the antioxidant properties of palm oil at 70°C, while the combination of TBHQ and BHT had a synergetic effect on the stability of soybean oil at room temperature and Linseed oil at 70°C.

According to Xiu-qin et al., (2009), at frying temperatures tocopherol, tocopherol esters and BHA have a decreased antioxidant impact, whereas ascorbic acid-6 palmitate and phytosterol fractions have a higher antioxidant effect; this has therefore contributed to the significant movement toward using natural antioxidants instead of synthetic antioxidants to improve the antioxidant capacity of fats and oils (Diamanti et al., 2017; Ke-Zheng et al., 2016).

2.3.6 Natural antioxidants

Plant sources of natural antioxidants include grains, spices, nuts, fruits, vegetables as well as seeds (Rather et al., 2016; Zahid et al 2024). Flavonoids (quercetin, kaempferol, myricetin), catechins or phenols (carnosol, rosmanol, rosamaridiphenol) and phenolic acids (carnosic acid, rosmarinic acid) are the constituents responsible for the antioxidative effect in plants (Diamanti et al., 2017; Embuscado, 2015).

Tocols are the natural antioxidants found in plant-based oils. The α-tocopherol is the most active biological isomer while γ-tocopherol is regarded as the best antioxidant (Taghvaei &Jafari, 2015). Among all types of tocopherols, α-tocopherol is the most unstable and therefore it is easily destroyed at elevated temperatures (Boschin & Arnoldi, 2011).

Edible oils such as peanut, corn, sesame, sunflower and soybean contain high levels of polyunsaturated fatty acids which are rapidly decomposed during continual frying (Xiu-qin et al., 2009). Natural compounds like tocopherols, oryzanol, sterol fraction and squalene on the other hand, improve their durability at extremely high temperatures. Several studies have been conducted on the use of natural plant isolates as antioxidants. (Bansal et al., 2013 and Lourenço et al., 2019) found out that natural plant extracts had greater antioxidant activity and heat resistance, which is the most critical criterion for an antioxidant to be employed for edible oils.

Pomegranate peel, green tea, olive waste, sesame cake, sesame seed, rosemary, Eucalyptus leaf, celery, oregano (*Origanum vulgare*) and cinnamon have all been employed as natural sources of antioxidants (Bouaziz et al., 2008; Fernandes et al., 2019; Orsavova et al., 2015; Zhang et al., 2020). The antioxidative effects of tocopherol (vitamin E); a fat-soluble carotenoid, citric acid and rosemary extract have all been studied extensively (Embuscado, 2015). The rosemary extract exhibited the highest antioxidative activity of all of them. When compared to BHA and BHT, sesame cake extract has greater antioxidant activity, however not as much as TBHQ (Carvalho et al., 2012). Sesame seeds' antioxidant activity is a result of its natural antioxidant components sesamolin, sesamin and sesaminol (Ruslan et al., 2018). Green tea, a powerful antioxidant exhibits excellent antioxidant activity at a concentration of 200 ppm and above in both sunflower and soybean oils; its antioxidant activity is higher than that of BHA and BHT but still lower that of TBHQ. (Casarotti & Jorge, 2014), studied the thermoxidative stability of soybean oil with rosemary extract at 3000 ppm and TBHQ at 50 ppm, from this study it was conclusive that natural antioxidants can only be more effective than TBHQ when used in higher concentrations compared to synthetic ones.

Moringa oleifera too has gained great recognition in the recent past as a natural antioxidant (Nadeem & Imran, 2016). *Moringa oleifera* contains bioactive compounds which have antioxidant properties; the compounds attributed to *Moringa oleifera's* antioxidant capability include tocopherols, catechins, quercetin, ferulic acid and zeatin. In assessing the physicochemical and antioxidant properties of *Moringa oleifera* (Ogbunugafo et al., 2011), reported that the presence of flavonoids in *Moringa oleifera* play a great role in its antioxidant action. The antioxidant potential of *Moringa oleifera* was further demonstrated by the ability to quench free DPPH radicals (Wright et al., 2017). Decreasing absorbance of DPPH in *Moringa* oil mixture in this experiment which measured the extent of radical scavenging ability in the oils also confirmed the findings as reported by Ogbunugafo et al., (2011): that flavonoids in *Moringa oleifera* play a role in carrying out antioxidant action through chelation or scavenging.

In the food industry and specifically in edible oils, several studies have been done to test the antioxidant capacity of *Moringa oleifera*. Nadeem et al., (2015), studied the effect of *Moringa* leaf extract as an antioxidant in vegetable oil blends, the leaf extracts were incorporated at three concentrations; 300, 600 and 900 ppm in comparison with 100 TBHQ as a control then stored at ambient temperatures. From the study, the free radical scavenging effect of the leaf extracts was comparable with that of TBHQ with the antioxidant activity increasing with increase in concentration of the leaf extracts. Both mature and tender leaves of *Moringa oleifera* have closely similar antioxidant effect on vegetable oils (Sreelatha & Padma, 2009).

Moringa oleifera blended in canola, sunflower and soybean oils at varying concentrations had a physicochemical effect on the oils. Shelf-life studies revealed that the oil had lesser oxidation byproducts with improved fatty acid composition. This evidence is proof that *Moringa oleifera* can be used to enhance the oxidative stability of edible oils. A fat blend of Moringa at 2.5%-10%

and butter oil was prepared then stored at room temperature, accelerated storage studies indicated that the blend had better oxidation resistance compared to butter oil alone (Nadeem et al., 2012). The concentration of alkenes were also greatly reduced in the blends as compared to butter oil (Nadeem et al., 2014).At 10% the free radical scavenging activity of the blend was 31.65% compared to 5.22% in butter oil.

Further to the findings involving edible oils, (Anwar & Bhanger, 2003) reported that, Moringa oleifera has beta-sitosterol and zeatin which are the most active antioxidants in the plant with 36 antioxidants naturally present in *Moringa oleifera* (Lalas & Tsaknis, 2002; Tsaknis et al., 1998).Conclusively, *Moringa oleifera* can replace the synthetic antioxidant for long term protection of edible oils against autoxidation. Other natural antioxidants that have been extensively studied are summerized in **Table 2**.

Table 2: Effects of natural antioxidants on edible oils

Natural sources of antioxidants	**Product stabilized**	**Outcome**	**References**
Peanut skin extract	Soy bean oil	Peanut shell extract ability to stabilize soybean oil was similar to the ability of BHT when oil is stored for 16 days	(Franco et al., 2018)
Ginger extract	Canola oil	Ginger extracts (6-gingerol, 8-gingerol, 10-gingerol, 10-gingerol and 6-shogaol exhibited antioxidant properties	(Si et al., 2018)
Sea weed	Canola oil	Aqueous sea weed extract ability of inhibiting oxidation of canola oil was similar to that of BHT. A threefold concentration of the aqueous extract had significantly higher antioxidant activity as compared to BHT.	(Agregán et al., 2017)
Tomato peel extract	Cotton seed oil	Cotton seed oil supplemented with tomato peel extract exhibited similar antioxidant activity against DPPH with cotton seed oil supplemented with BHT	(Elbadrawy & Sello, 2016)
Potato peel extracts	Soy oil	Stability of soy bean oil supplemented with potato peel extract was significantly higher than stability of soy bean oil without any antioxidant added. It is worth noting that stability of soybean oil supplemented with potato peel extract had was significantly lower than that of oil supplemented with BHT at a concentration of 200 ppm	(Franco et al., 2016)
Orange peel extract	Ghee (butter oil)	Ghee supplemented with orange peel extract was more stable at 6 and 32°C as compared to 60 °C. In ghee supplemented with orange peel extract there was significantly lower levels of peroxides, TBA and AGL as compared to ghee supplemented with BHA	(Asha et al., 2015)

Banana peel extract	Refined sunflower and soybean oils	Addition of banana peel extract to sunflower and soybean oil at various concentrations increased the stability of the oils. The highest stability was observed at a concentration of 0.4% which was comparable to the antioxidant capacity of BHA	(Kurhade & Waghmare, 2015)
Roasted rice hull extracts	Corn oil	Ethanolic roasted rice hull extracts had significantly high antioxidant activity as compared to aqueous extracts	(Park et al., 2019)
Oregano extract	Virgin olive oil	Oregano leaf extract added at a concentration of 5g/100ml significantly inhibited lipoperoxidation of virgin olive oil	(Peñalvo et al., 2016)
Basil extract	Sunflower oil	Addition of basil extract to sunflower oil at a concentration of 200-500mg/kg exhibited oxidative stability comparable to stability of BHT added to sunflower oil	(Ben-Ali et al., 2014)

Subsequently, with respect to circular economy the trend of use natural as source of antioxidants has increased the interest of researchers for new raw materials with antioxidant power (such as by-products from the agricultural-food industry), without affecting the consumers' perceptions and the quality of the final products (Lourenço et al., 2019). For instance, Mulberry Silkworm pupae, a by-product of yarn reeling, can be a great natural anti-oxidant.

Mulberry Silkworm (**Fig.2**) is an insect under the order Lepidoptera, commercially reared for silk production. Although, the other silkworm varieties *tasar, eri, and muga* are also used for silk production (**Fig. 2**), most of the world's silk is produced from the Mulberry Silkworm (Sheikh et al., 2018). The latter accounts for 90% of commercial silk production in the world (Patil et al., 2019). At the pupal stage of its lifecycle, silkworms build a shielding cocoon made up of raw silk. After pupation, the cocoons are chemically or heat-treated so as to release an enzyme that breaks open the cocoon to release the pupae (**Fig.2**). The spent pupae from yarn reeling are then discarded as a waste product.

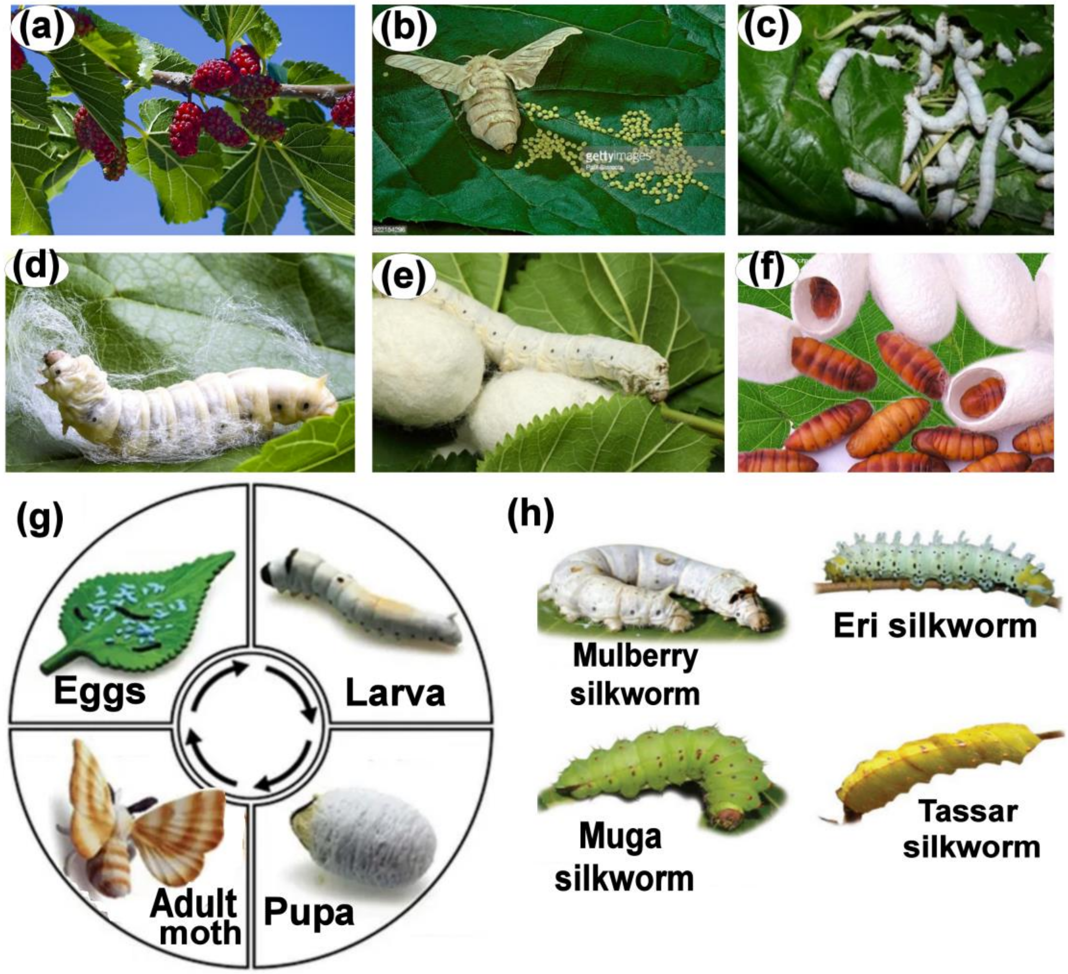

Figure 2: a) Mulberry plant, **b)** Adult mulberry moth lying eggs, **c)** Mulberry larva, **d)** Mulberry larva making silk, **e)** Mulberry silkworm cocoon with pupae, **f)** Mulberry pupae, **g)** Mulberry silkworm lifecycle and **h)** Various types of silkworms. (International Sericultural Commission, 2013).

A lot of silkworm pupae is produced from yarn reeling yet little has been done to find its utilization. Apart from some of it being used as livestock and poultry feed, most of it ends up in dump sites. Despite the chemical composition of disliked silkworm pupae having attracted great attention worldwide and as an outstanding reservoir of an enormous number of bioactive compounds, little research has been done on them (Mahesh et al., 2015).

Mulberry Silkworms have a distinct pattern of redox components including phenolics and proteins (Butkhup et al., 2012), these chemical species can protect against oxidative damage in both lipophilic and hydrophilic conditions. According to Kotake-Nara et al., (2002) and Ravinder et al., (2016), the silkworm pupae contain antioxidant tocopherol; about 180 micrograms per gram of extract. Carotenoids, lutein, Violaxanthin, and neoxanthin approximately 5.12, 0.28, and 0.88 (ug/g) pupae (wet basis) respectively, are also present in Mulberry Silkworms (Tabunoki et al., 2004). In addition to their radical scavenging impact, these compounds have other varied functions, such as antiallergy, antityrosinase, and anti-inflammatory activities (Park et al., 2008; Yokohira et al., 2008).

An investigation by Pachiappan et al., (2016) looked into the antioxidant potential of silkworm pupal products such as pupal powder, chitosan, and pupal oil by measuring their radical scavenging activity. There was no significant difference between the scavenging activity of silkworm pupae by-products and ascorbic acid, a well-known antioxidant. At 10 g/ml, the by-products had a radical scavenging ability of 60 to 76 %, compared to 64 % for ascorbic acid of the same quantity. Winitchai et al., (2011) reported the free radical scavenging activity in these by-products to be attributed to phospholipids and tocopherol, which play an important role in protecting the lipids against oxidation. Investigation of the antioxidant activity of silkworm by the determination of the phenolic and flavonoid content also revealed that they also have an average of 15.5 mg catechin/g and 5.4 mg catechin/g phenolic and flavonoid contents respectively (Deori et al., 2014).

The two main kinds of pigments responsible for cocoon color in silkworms are ether-soluble yellowish carotenoids and water-soluble green flavonoids (Kurioka & Yamazaki, 2002; Tamura et al., 2002). The coloring content is dependent on the Mulberry silkworm strain. Flavonoid compounds, in addition to fibroin and sericin proteins in mulberry silkworms are also accountable for the antioxidant elements (Kato et al., 1998; Kurioka & Yamazaki, 2002).

The Mulberry silkworm pupae are widely utilized as food and feed in Asian countries and other parts of the world due to its high-quality nutrient profile; they are high in protein and include monounsaturated and polyunsaturated fatty acids in their fat (Zhou & Han, 2006). Mulberry Silkworm pupae on average contain 40% and 5% omega-3 and omega-6 fatty acids respectively of total fatty acids by weight respectively (Kwon et al., 2012). Omega-3 (docosahexaenoic acid) which is the most dominant is an essential fatty acid also found naturally in some seafood and maternal breast milk. It is responsible for organ development in premature babies. The pupae, therefore, have a great potential in human diets. In China, for instance, the silkworm pupae have been approved as a novel food source by the Ministry of Health, (Patil et al., 2019).

While Mulberry silkworm pupae have been explored for food and other uses, the functionality of their biologically active compounds remain under-explored (Di Mattia et al., 2019; Kotake-Nara et al., 2002). The nutritional makeup of edible oils is significantly influenced by the processing of edible oils (Matthäus, 2010). Crude oils consist of triacylglycerides, free fatty acids, phospholipids, phytosterols, waxes, color compounds and aroma components. Some of these components impair the stability of the oil since some of them undergo oxidative deterioration during processing therefore they have to be removed by refining. The refining process eliminates even the vital constituents such as free fatty acids, tocopherol, phenolic compounds and carotenoids (Matthäus, 2010). These compounds are not only important for shelf stability but also nutritionally hence they have to be added back through synthetic fortification which has sometimes proven inefficient since some of the fortificants are lost during storage (Diosady & Krishnaswamy, 2018). The use of pupae as an antioxidant source, a high-value product, will not only boost yarn reeler returns and pupae use, but will also lessen environmental concerns. Synthetic antioxidants also need more energy to metabolize than natural antioxidants. Since silkworm pupae are a good source of high quality unsaturated fatty acids, functional pigments such as lutein, neoxanthin, carotenoids and phenolic compounds (Longvah et al., 2011; Tomotake et al., 2010); the functional and nutrient profile of the oils can be restored without fortification hence making the process more sustainable and economical.

Conclusion

Shelf life of edible oils is mainly affected by oxidation of the oils. Synthetic antioxidants are currently in use to improve the shelf stability. However, they are strictly regulated due to the possible risks associated with food safety and human health. Among other undesirable effects, skin allergies, gastrointestinal tract problems and the possibility of development of cancerous cells are common problems associated with artificial antioxidant in edible oils. To this extent, natural antioxidants seem to hold the key to prolonged shelf-life of edible oils. For instance, the antioxidant activity of silkworm pupae and *Moringa oleifera* are unmatched and can be explored as an antioxidant in vegetable cooking oils. However, more research is needed to ascertain the use of natural antioxidants including toxicological tests.

Data availability

The authors confirm that the data supporting the findings of the study are available within the article. Raw data that support the findings of the study are available from corresponding author upon request.

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