

Review

Innovative and Sustainable Technologies to Enhance the Oxidative Stability of Vegetable Oils

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Abstract: To meet consumers' demand for natural foods, edible oil producers and food processing industries are searching for alternatives to synthetic antioxidants to protect oils against oxidation. Antioxidant compounds extracted from different plant parts (e.g., flowers, leaves, roots, and seeds) or sourced from agri-food industries, including residues left after food processing, attract consumers for their health properties and natural origins. This review, starting from a literature research analysis, highlights the role of natural antioxidants in the protection of edible oils against oxidation, with an emphasis on the emerging and sustainable strategies to preserve oils against oxidative damage. Sustainability and health are the main concerns of food processing industries. In this context, the aim of this review is to highlight the emerging strategies for the enrichment of edible oils with biomolecules or extracts recovered from plant sources. The use of extracts obtained from vegetable wastes and by-products and the blending with oils extracted from various oil-bearing seeds is also pointed out as a sustainable approach. The safety concerns linked to the use of natural antioxidants for human health are also discussed. This review, using a multidisciplinary approach, provides an updated overview of the chemical, technological, sustainability, and safety aspects linked to oil protection.

Keywords: natural antioxidants; lipids oxidation; food waste; oil blending; sustainability; oxidative stability; peroxidation

1. Introduction

During storage, handling, or cooking, edible oils may undergo deterioration processes due to lipid oxidation. Oxidation is mainly responsible for the loss of oil quality, causing the decrease in an oil's nutritional value and giving rise to undesirable off-flavors, which make oils containing food less acceptable to consumers. Moreover, lipid oxidation produces some toxic by-products like reactive carbonyl compounds (RCCs), which could generate

advanced lipid peroxidation end products (ALEs), which are potentially harmful to human health [1]. The extent of lipid oxidation depends upon the oil's internal factors, like the unsaturation degree, the presence of antioxidant compounds, or metals like copper and iron, and the oil's external factors, such as oxygen availability and temperature. The contact of oils with oxygen gives rise to chain reactions that progress faster with higher temperatures as well as with an increased unsaturation degree of the lipids.

Lipid peroxidation is a major concern for food industries due to the rising trend toward the replacement of hydrogenated oils containing saturated fatty acids (SFAs) with monounsaturated (MUFAs) and polyunsaturated fatty acids (PUFAs), which are considered healthier by consumers but are much more susceptible to oxidation than SFAs. In addition, oils rich in MUFAs and PUFAs, when exposed to deep frying or cooking temperatures, may develop RCCs, which originate ALEs, posing serious health risks. Even if the consumption of oils rich in MUFAs and PUFAs is strongly encouraged, these oils need to be "protected" before being used for cooking. Some oils (e.g., olive oil) are naturally protected by their high content of endogenous antioxidants (polyphenols and tocopherols), while some others (e.g., soybean, sunflower, and peanut oils), due to the refining process they undergo [2], need to be enriched with exogenous antioxidants during their production to make them more resistant to oxidation.

Thus far, the control of oxidation processes is achieved by synthetic antioxidants like butylated hydroxyanisole (BHA), butylated hydroxytoluene (BHT), *tert*-butylhydroquinone (TBHQ), and propyl gallate (PG), which are able to extend oils' storage time by delaying or hindering the degradation of lipids [3–5].

The safety concerns associated with the use of synthetic antioxidants and the consumers' increasing demand for natural food fostered the development of methods based on the employment of natural molecules to delay oil oxidation [6]. In this context, natural antioxidants have been suggested as an alternative to synthetic additives in the prevention of oil oxidative degradation. Plants are the main source of natural antioxidants, and the ability of plant antioxidants to protect oils from lipid degradation has been the topic of several investigations [7–14].

Make it green and make it healthy are the issues of the Next Generation EU program [15], which aims at making food production more sustainable for human health and for the environment. The strategy of the European commission in the future is to move toward a circular economy and sustainability, making better use of food waste, side streams, and by-products. Recent estimates report that about 88 million tons of food is wasted every year in Europe. Much of this waste comes from households, while food processing waste accounts for 30.6 million of tons annually, and fruits and vegetables contribute 35% [16]. These products still contain large amounts of bioactive compounds like flavonoids, anthocyanins, carotenoids, and phenolic acids, which can be valuable for the oils industry to enhance their oxidative stability [16].

Following the strategy adopted by the EU, this review focuses on the use of biomolecules and crude extracts obtained from food by-products and side streams to protect edible oils from oxidation. Starting from a qualitative and quantitative analysis of the literature published so far on the use of antioxidants to enhance oil oxidative stability, this work highlights the role of antioxidants in oils' oxidative stability, focusing on their mode of action as well as on the conventional and emerging analytical methodologies used to assess oxidative stability and antioxidants' efficacy. The role of natural antioxidants coming from food side streams and by-products and the innovative technologies recently developed, for oil enrichment, such as antioxidant encapsulation and blending with unconventional oils, are also discussed. Following a multidisciplinary approach, this review emphasizes the safety concerns linked to the use of natural antioxidants for human health.

2. Antioxidant, Oil Oxidative Stability, Waste, Biodiversity, and Sustainability: Literature Research Review

To give a current snapshot of the interest raised in the international research context, a literature overview of the relationships among oxidative stability, oil, and antioxidants was performed on the Scopus database based on the following search string: (“oxidative stability*” AND “oil*” OR “antioxidant*”) (accessed on 24 October 2021). Bibliographic data (e.g., publication year, publication count, document type, countries or territories of origin, and institutions) were recorded. The functions of the Scopus web online platform named “Analyze” and “Create Citation Report” were utilized for carrying out basic analyses.

The search resulted in 2419 publications from 1953 to 2022. The most cited paper (1043 times) was that of Choe and Min [17], published in 2006 in *Comprehensive Reviews in Food Science and Food Safety*, addressing the mechanisms and factors behind edible oil oxidation.

The most productive author was Decker, E.A. with 48 documents. His most cited paper (336 times) dealt with lipid oxidation in corn oil-in-water emulsions stabilized by casein, whey protein isolate, and soy protein isolate [18]. Among the most cited works, the work of Faraji et al. [19] described the role of continuous phase protein in the oxidative stability of fish oil-in-water emulsions. Another one is focused on the antioxidant activity of cysteine, tryptophan, and methionine residues in continuous phase β -lactoglobulin in oil-in-water emulsions [20]. Figures 1 and 2 report the most productive countries or territories and institutions, respectively. Regarding countries and territories, the United States ($n = 353$) was the most productive country, followed by China ($n = 256$) and Brazil ($n = 249$). The main keywords covered for documents reported from the United States were antioxidant(s), oxidation, oxidative stability, chemistry, lipid oxidation, oxidation inhibition, and emulsion(s).

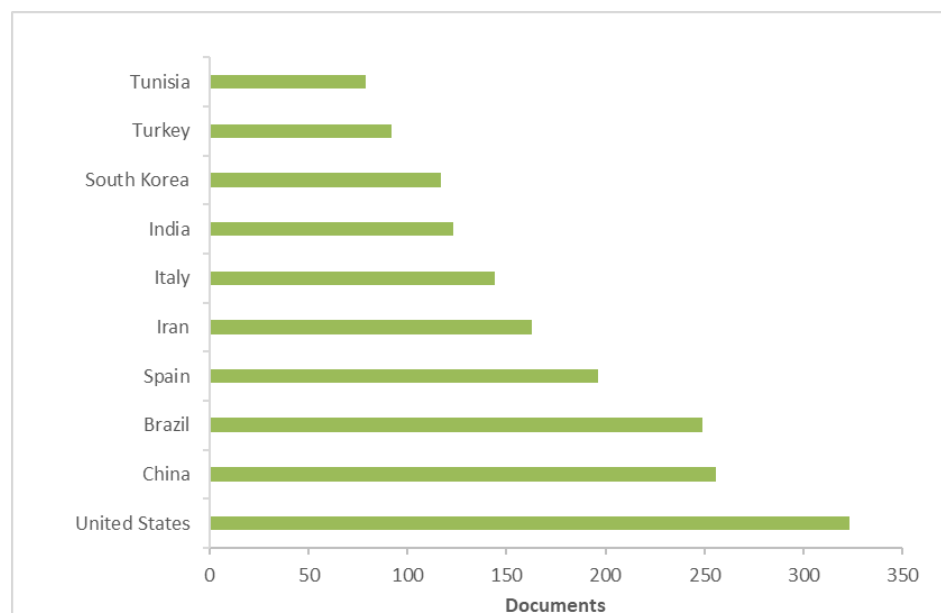


Figure 1. Most productive countries and territories (based on data from Scopus).

The most productive institution was the University of Massachusetts Amherst with 59 documents. All the top 10 institutions contributed at least 33 publications or more. The most cited paper was the paper of Hu, McClements, and Decker [18], which was previously described, whereas the most recent work by Inchingolo et al. [21] described the ability of sodium dodecyl sulfate (SDS) micelles to increase the antioxidant activity of α -tocopherol.

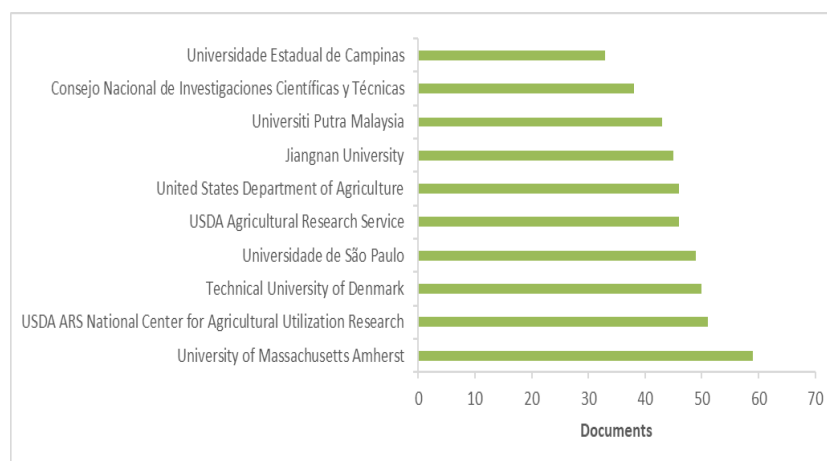


Figure 2. Most productive institutions (based on data from Scopus).

The search string TITLE-ABS-KEY (“oxidative stability*” AND “oil*” OR “antioxidant*” AND “waste*” OR “by-product*” OR “byproduct*”) was also used to address the literature review search toward the utilization of waste and by-products. The search returned 172 publications covering the period from 1989 to 2021. “Full records and cited references” were exported to VOSviewer software (version 1.6.16, www.vosviewer.com, accessed on 24 October 2021) for further bibliometric study and additional processing. The VOS viewer software analyzes the terms or words used in the titles and abstracts of publications by breaking down the paragraphs into words and phrases, linking them with the citation data of the publications, and visualizing the results in the form of a bubble map by using a term map with the default settings [22–24].

The most cited paper (238 times) addressed the radical scavenging activity of black cumin (*Nigella sativa* L.), coriander (*Coriandrum sativum* L.), and niger (*Guizotia abyssinica* Cass.) crude seed oils and oil fractions [25]. Then, the paper of Fernández-Bolaños et al. [26] (160 times) dealt with the production in large quantities of highly purified hydroxytyrosol from the liquid-solid waste of two-phase olive oil processing or “alperujo”. Iqbal, Haleem, Akhtar, Zia-ul-Haq, and Akbar [14] (138 times) described the efficiency of pomegranate peel extracts in the stabilization of sunflower oil under accelerated conditions.

A total of 105 terms were identified and visualized as a term map (Figure 3). The top recurring terms on oxidative stability, oil, antioxidant and waste, and by-product and byproduct research are the following: antioxidant(s), oxidative stability, chemistry, oxidation, fatty acids, oxidation-reduction, oxidation-reduction reaction, phenols, oils and fats, fruits, plant extract(s), antioxidant activity, olive oil, vegetable oil, plant oils, and biodiesel.

When narrowing the inquiry considering the sustainability and circular economy by using the search string TITLE-ABS-KEY: (“oxidative stability*” AND “oil*” OR “antioxidant*” AND “waste*” OR “by-product*” OR “byproduct*” AND “sustainability*” OR “circular economy*”, the search returned eight publications. Among the “Review” category, Difonzo et al. [27] described the functional compounds from olive pomace to obtain high added value foods. On the other hand, Blasi and Cossignani [7] gave an overview of natural extracts with antioxidant activity for the improvement of the oxidative stability and shelf life of edible oils, highlighting new approaches in the field of health-promoting foods. Angelovič et al. [28] focused on the review of the oxidative stability of fatty acid alkyl esters.

Among the “Article” category, Cisneros-Yupanqui et al. [29] gave a preliminary characterization of wastes from the rapeseed and sunflower protein isolation process and their valorization in delaying oil oxidation. The rapeseed and sunflower ethanol wash solutes showed great potential, and they could be used as a source of natural antioxidants in the field of the food industry, replacing the synthetic ones and promoting the circular economy since they are agro-food wastes. Luzi et al. [30] reported how hydroxytyrosol and

Palm oil is the most frequently used edible oil in the world [37]. Two types of oils with different fatty acid compositions can be obtained from palm fruit: palm oil and palm kernel oil [38]. The former is extracted from the mesocarp (rich in palmitic acid and oleic acid), and the latter is extracted from the kernel (rich in lauric acid) [37].

Olive oil is among the most popular gourmet and health-promoting specialty oils, and it is produced predominantly in the Mediterranean region [39]. Adherence to the Mediterranean diet, in which olive oil is a staple ingredient consumed in high amounts, is reported to be associated with various health benefits including a reduction in mortality due to coronary heart diseases [40].

Today, due to the increasing health awareness of individuals and rising living standards, eating habits are also changing. Heart diseases and cancer, which are closely related to diet, especially the quality and amount of dietary fat, are major causes of death in developed societies [41,42]. The functional properties of vegetable oils are mainly due to their fatty acid compositions (essential fatty acids such as ω -3 and ω -6 and the ω -3: ω -6 ratio) and bioactive components (e.g., tocopherols, sterols, squalenes, carotenoids, antioxidants, and phenolics).

In recent years, the interest in natural and reliable oils have been increasing. Therefore, consumer preferences have changed rapidly from refined oils toward virgin oils due to the increasing awareness of healthy nutrition. Consumers demand foods that are proven to have positive effects on health and contain health-promoting constituents. To meet consumers' demands, the bulk production of virgin oils should be accomplished from oilseeds or fruits at reasonable prices. For this purpose, there are major challenges that have to be addressed, including sustainable production of high-quality oil seed, the production of virgin oils with a higher yield, and higher bioactive components such as antioxidants.

3.2. Oil's Oxidative Stability: Main Features, Mechanisms, and Analytical Methods for Its Determination

A vegetable oil's quality is defined by both its compositional and organoleptic properties. These also determine consumer acceptance and agro-industrial preferences. One of the main quality attributes in vegetable oils is their oxidative stability. Unfortunately, all vegetable oils can undergo a degradation phenomenon known as oxidation. This is a complex series of reactions leading to rancidity, smells, and off-flavors [43,44]. Likewise, a vegetable oil's shelf life depends on its oxidative stability [45,46]. It is noteworthy that during the oxidation process, some toxic compounds such as RCCs can result in advanced lipid peroxidation end products, which are potentially harmful to human health [47]. Many factors are involved in oil oxidation. The main important factors are the storage conditions, a high temperature, high oxygen availability, the level of polyunsaturated fatty acids, and their composition, as well as the presence of prooxidants such as chlorophylls, metal ions (Fe^{3+} and Cu^{2+}), and heavy metals [17,48].

The oil oxidation reactions are explained by the conventional free radical chain phenomenon. This process begins with radical reactions on unsaturated fatty acids [49]. These reactions include three phases, namely initiation, propagation, and termination, involving several reactions and mechanisms that are well documented in the literature [50–53].

To assess an oil's oxidative stability, a set of routinely measured parameters is adopted. Among them, the peroxide value (PV) and K_{232} specific extinction coefficient, measured at $\lambda = 232$ nm, are very important indicators of the degree of degradation of an oil at the initial stage (i.e., formation of primary oxidation products) [54]. Therefore, the determination of PV and K_{232} is a reliable method for predicting subsequent deterioration of an oil's organoleptic quality. However, an oil may be rancid or have mediocre organoleptic properties without having a high PV. In fact, the hydroperoxides or peroxides and conjugated dienes are unstable products, and they are quickly transformed into ketones and aldehydes as secondary oxidation products [55].

The content of the secondary oxidation products is usually evaluated by examining the p-anisidine value [56]. This is considered to be a more reliable indicator of oxidative

rancidity compared with the PV [57]. The K_{270} specific extinction coefficient is another important index used to evaluate the secondary oxidation products. However, this parameter is not completely similar to the analysis of p-anisidine, because the ketones are not detected in anisidine determination [58].

Likewise, a better picture of vegetable oil oxidation, the total oxidation (TOTOX), can be calculated. This index merges the PV (primary oxidation products) and p-anisidine value (secondary oxidation products). Total oxidation is better compared with the peroxide and p-anisidine values since the hydroperoxides (peroxides) are unstable and do not offer a reliable picture for an oil's oxidative stability [59]. In general, a TOTOX less than 10 indicates better quality [59]. Other measurements such as the headspace volatiles (smell), free fatty acids (FFA), thiobarbituric acid value (TBA), iodine value (IV), the Rancimat test, and the Shaal Oven Test can also be tested for controlling the stages of the oxidative stability of vegetable oils [46,60]. The Rancimat test determines the resistance time of oil to oxidation, achieved via a conductimetric measurement [61–64]. Recently, electron paramagnetic resonance (EPR) spectroscopy coupled with the spin trapping techniques has gained great interest due to its reliability and specificity in the detection of the radicals produced during lipid oxidation. This technique has been used to determine the oxidative stability of food lipids, bulk oils, oil emulsions, or fried oils by measuring the resistance to the formation of radical species [65–67]. Peroxyl and alkoxyl radicals formed during oil oxidation are trapped by N-tert-butyl- α -phenylnitrone (PBN), the most widely used open-chain nitron spin trap, forming a PBN radical adduct detected by EPR. The presence of antioxidants hinders the propagation step of lipid peroxidation, slowing down the formation of the radical adducts which, being relatively stable, are easily detected by EPR in different vegetable oils [65,68–72].

4. Focus on Synthetic and Natural Antioxidants for the Stabilization of Edible Oils

Vegetable oils are widely used in food preparation. Their oxidative stability is a key quality parameter, which depends upon the balance of different intrinsic and extrinsic factors. The main important factors are fatty acid unsaturation, the composition of minor components, environment conditions, delivery techniques, and the use of antioxidants (Figure 4). Lipid oxidation leads to negative effects on both the food quality and human health. In such a context, minimizing oxidation and improving the oxidative stability of lipid-based products become imperative [73].

Vegetable oils are endowed with a wide variety of endogenous antioxidants (e.g., pigments, vitamins, tocopherols, and phenols). However, the use of exogenous antioxidants is widely practiced to improve oxidative stability. In this optic, synthetic antioxidants such as butylated hydroxyanisole (BHA), butylated hydroxytoluene (BHT), tert-butylhydroquinone (TBHQ), and propyl gallate (PG) are commercially used to extend an oil's shelf life by delaying or even hindering lipid degradation. Such molecules were considered "generally recognized as safe" (GRAS) [5]. However, some reports associated these molecules with health risks such as carcinogenesis, and their use was reduced [5,6]. Natural antioxidants are a good alternative to replace synthetic ones in the preservation of vegetable oils [3–5].

Natural extracts from various plant parts (e.g., peel, fruit, leaf, flower, and root) belonging to different herbs, agri-food residues, and by-products were investigated for their antioxidant power as well as their use for the enrichment of edible oils, with an emphasis on enhancing oxidative stability. Such natural extracts were proven to possess a wide range of bioactive compounds, which were identified as belonging mainly to carotenoids and phenols [7,74]. Promising results were obtained regarding the improvement of oxidative stability and shelf life of vegetable oils. The effect of natural extracts on an oil's oxidative stability will be discussed deeply in Section 5 of this review. With respect to the antioxidant activity of synthetic and natural additives, several mechanisms are involved. They act as free radical scavengers, inactivators of peroxides as well as other ROS, singlet oxygen quenchers, metal ion chelators, quenchers of secondary oxidation compounds, and inhibitors of pro-oxidative enzymes, among other functions [73]. According to these authors,

antioxidants may be classified, based on their mode of action, into primary antioxidants, which break the oxidation chain reaction via scavenging free radical intermediates, and secondary antioxidants, which prevent or delay oxidation through the suppression of oxidation initiators, accelerators, or the regeneration of primary antioxidants.

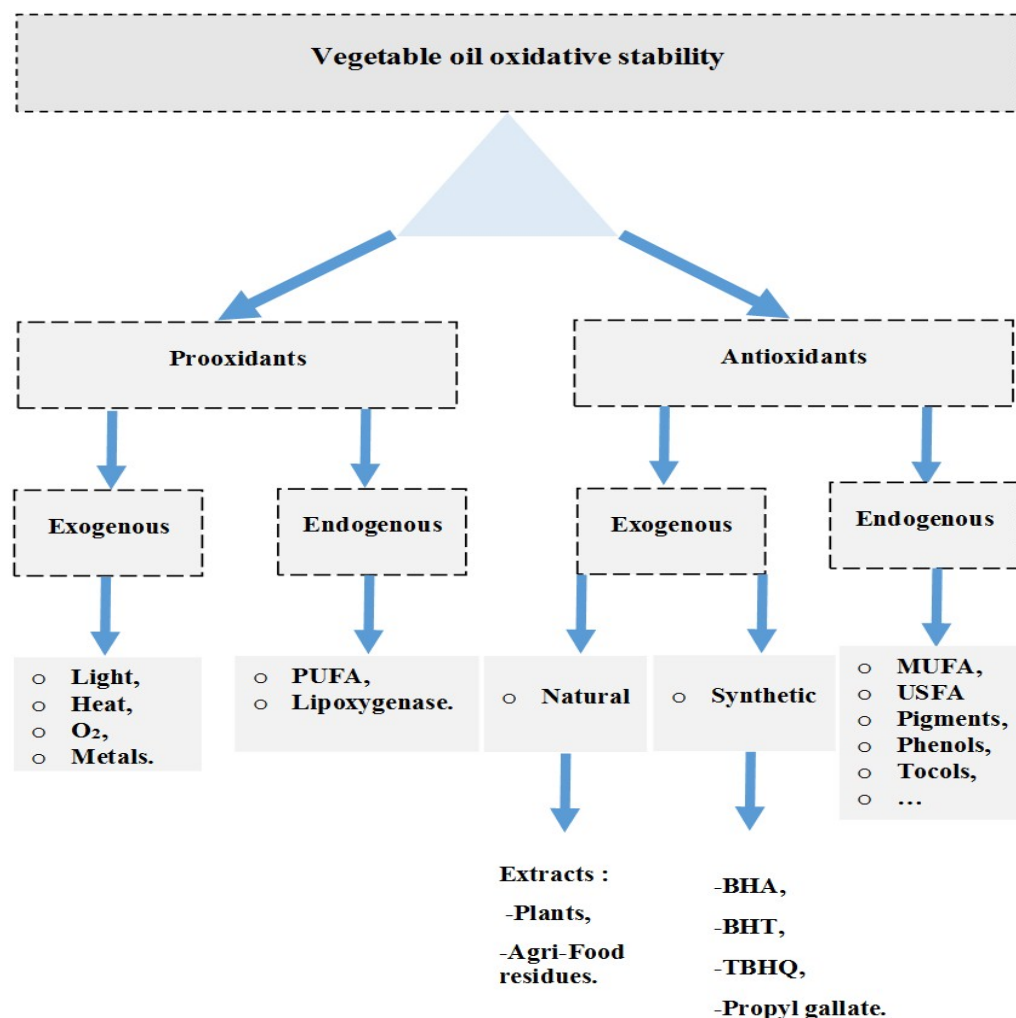


Figure 4. An overview of factors involved in vegetable oil oxidative stability.

4.1. Synthetic and Natural Antioxidants

The most commonly used synthetic antioxidants added to oils are BHA, BHT, TBHQ, and PG (Figure 5). The amount which can be added depends on national regulations, being around 100–200 mg kg⁻¹ [3,12,75–78]. These compounds share the common structural feature of having at least one phenolic group. TBHQ and PG are more effective than BHT and BHA because they can be oxidized to semiquinones and quinones, but PG can also chelate redox active metal ions like iron(II/III).

Redox active transition metal ions (Fe^{II}/Fe^{III} or Cu^I/Cu^{II}) can influence the initiation step of a lipid peroxidation reaction. In fact, if these metal ions are present in reduced form (Fe^{II} or Cu^I), they can react with peroxides, generating hydroxyl radicals ([•]OH) with a Fenton reaction, which in their turn can start new radical chain reactions. In the literature, it has been reported that chelating agents exert their antioxidant activity by blocking the cycling of redox-active metal ions between the two redox states (i.e., Fe^{II}/Fe^{III}) [79]. This is not strictly true because the chelating ligands can coordinate both the oxidation states of the metal ions. What is really changed is the redox potential of the couple Fe^{II}/Fe^{III} [80]. If the chelating ligands stabilize the oxidized form (Fe^{III}), Fe^{II} will oxidize more easily to Fe^{III}, its concentration will be decreased, and this will not be able to give the Fenton reaction, thus

preventing the start of new radical chain reactions [81]. The more active natural or synthetic antioxidants have in their structure di- or tri-hydroxybenzene (o-catechol) moieties, and these are strong chelating agents with a high affinity for Fe^{III} . When these ligand molecules are present in oils together with trace amounts of metal ions (Fe or Cu), the latter have more of a tendency to exist in oxidized form, thus preventing the Fenton reaction and therefore protecting oils from further oxidation.

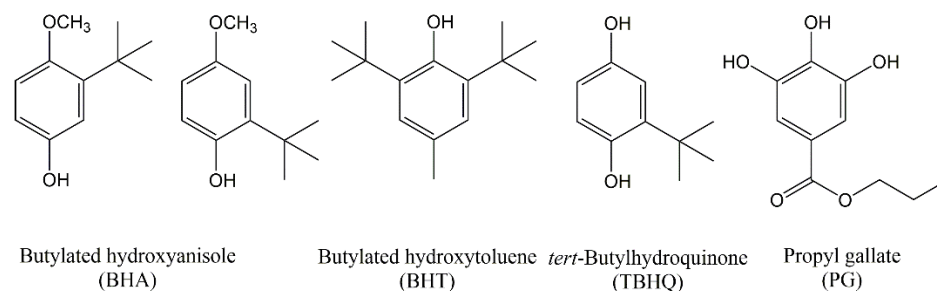


Figure 5. Synthetic antioxidants.

The mechanism of action of antioxidants entails the formal abstraction of a hydrogen atom (H^\bullet) from the phenol group (Ph-OH) to form a phenoxyl radical (Ph-O^\bullet), which being stabilized by resonance and/or sterically hindered is less reactive than the alkyl/peroxyl radicals involved in the lipid peroxidation chain reaction. In this way, the propagation step is hindered or slowed down, contributing to the increased oxidative stability of oils.

Natural and synthetic antioxidants, depending on their concentrations, can behave as, aside from beneficial antioxidants, pro-oxidants. This has been verified with purified olive oil (containing only triacylglycerols without mono- or di-acylglycerols or other minor components) containing 100, 500, and 1000 ppm of α -tocopherol [82]. The greater protective effect against the formation of peroxides during storage at 40 °C was obtained with 100 ppm of tocopherol, while at a higher concentration, the protection decreased. This effect was due to the auto-oxidation of tocopherol, which generated free radicals that could start new chain reactions. Vegetable oils contain ca. 95% of triacylglycerols, with some di- and mono-acylglycerols and free fatty acids. Some of the remaining 5% comprises compounds that can be classified as antioxidants, like tocopherols, tocotrienols, and polyphenolic compounds [83]. Tocopherols and tocotrienols exist as four different compounds each, indicated by the letters α , β , γ , and δ and differing by the number and position of methyl substituents on the chromane ring (Figure 6A). There is no agreement on the relative antioxidant activity of tocopherols in vitro [84]. The results depend on what is being measured. For example, γ is more effective than α at trapping radical species because it has no substituent on C-5, while if we consider the stability of the tocopherol radical, α is the more stable one, because all the H atoms in the aromatic ring are replaced by CH_3 groups. The three methyl groups on the aromatic chromane ring stabilize the phenoxyl radical by inductive and steric effects. Olive oil contains tocopherols in the range 100–300 mg kg^{-1} [85], while sunflower oil contains between 440 and 1520 mg kg^{-1} [86]. The tocopherol content in argan oil can be up to 900 mg kg^{-1} and is never below 600 mg kg^{-1} [54].

Polyphenolics comprise a large variety of compounds, among which phenolic acids (o- and p-coumaric, vanillic, homovanillic, sinapic, syringic, p-hydroxybenzoic, p-hydroxyphenylacetic, ferulic acids, all containing only one phenolic $-\text{OH}$ group and one or more $-\text{OCH}_3$, as well as caffeic, protocatechuic, and gallic acids containing two or three adjacent OH groups), tyrosol, hydroxytyrosol, and their ester derivatives (oleuropein, liginoside, oleacein, and oleocanthal).

Most of these compounds, as their names suggest, contain one or more phenolic groups, and when more than one is present, some can exist as $-\text{OCH}_3$. These latter groups are less effective in comparison with phenolic $-\text{OH}$ groups at scavenging radical species because they cannot donate a hydrogen atom to quench radical species, but they

Biorefinery represents an integrated approach and strategy to tackle food waste valorization in the perspective of a circular economy by addressing the agro-food chain and the lifestyle toward a “zero waste” model [94]. Sustainable food management is based on a circular economy and requires efforts and changes in agricultural and industrial organization [95].

4.3. Antioxidants' Extraction Methods

Nowadays, different extraction methodologies have been introduced, such as green and sustainable techniques for the separation of natural products from waste, in order to replace the conventional methods of extraction based on the use of organic solvents [96]. Examples of this include enzymatic treatment, microwave-assisted extraction, ultrasound-assisted extraction, supercritical and subcritical fluid extraction, and others.

For instance, Gligor et al. gave a comprehensive review of enzyme-assisted extractions of polyphenols. The use of specific enzymes or enzyme mixtures as well as the set-up of the process parameters (e.g., temperature, time, concentration, and pH) are key elements [97].

Kumar et al. described the ultrasound-assisted extraction (UAE) of bioactive compounds from fruit and vegetable processing by-products [98]. Ultrasound waves, by inducing cavitation with erosion of the plant tissues, fragmentation of the cells, and the release of bioactive compounds, can increase the extraction yield with respect to conventional techniques [98,99]. Microwave-assisted extraction is described as an efficient method, with short extraction times, high extraction rates, selectivity, and yields, and low cost [100].

The very recent review of Villacis-Chiriboga [101] gives an overview of the applications and economic and environmental aspects of biorefinery via supercritical fluid extraction. The subcritical water extraction of natural products is currently described by Cheng et al. [102] under ecology, economy, and safety aspects. Its major advantage is that water is nontoxic, and so it is more suitable for the extraction of herbs, vegetables, and fruits.

A current and green alternative to study bioactive components in foods and food wastes is given by the combined use of spectroscopic techniques and chemometrics, which has the advantage of a simple sample preparation and extraction procedure and a short time for data collection and analysis [103,104].

Generally, the current approach is the application of sustainable and integrated methods to food wastes and by-products, particularly toward the construction of a new suitable and industrial platform under the perspective of the biorefinery approach [105–108].

5. Enrichment of Edible Oils with Antioxidant Rich Extracts

5.1. Application and Beneficial Outcomes: Case Studies

Because lipid peroxidation is a drawback for the food industry, some studies focused on finding solutions to overcome this issue (Figure 7). In this way, researchers put their efforts toward finding the best way to increase an oil's stability. Rubio et al. [109] revealed that although virgin olive oil has phenolic compounds in its composition, an enriched olive oil with thyme phenolic extract showed increased oxidative stability and also a higher level of bioactive compounds, such as flavonoids, which have the ability to promote human health. Furthermore, the resulting product comprises the requirements of functional food, which gives phenol extract enrichment relevant applicability regarding an oil's stability [109].

Suarez et al. [110] managed to develop three prototypes of enriched virgin olive oils with phenols extracted from olive cake, which was a by-product. Their results were promising from different points of view. First of all, their article shows that the oxidative stability was increased proportionally with the addition of phenolic extract from olive cake, and the quality parameters remained within the range that made them edible. Second of all, the stability of the phenolic compounds during digestion was higher than under normal conditions, which can be an important premise for future studies regarding the bioavailability and metabolism of phenols [110]. Further studies were made in order to improve

the stability of the enriched olive oil and enhance the bioavailability of phenolic compounds, which are known to be susceptible to degradation throughout the gastrointestinal tract. Thus, the scientists tested the effects of lecithin as an emulsifier that could stabilize the phenols. The results showed that not only did the lecithin increase the stability levels of the compounds, but the shelf life of the oils was also extended, delaying the initiation of peroxidation, which was in line with the results of Delgado-Adamez et al. [111,112]. Moreover, the same team of researchers highlighted that a higher amount of phenolic compounds in olive oil could increase the overall bioavailability of these substances in an in vivo study, showing that the concentration in plasma was higher in the case of enriched olive oil compared with plain oil [113].

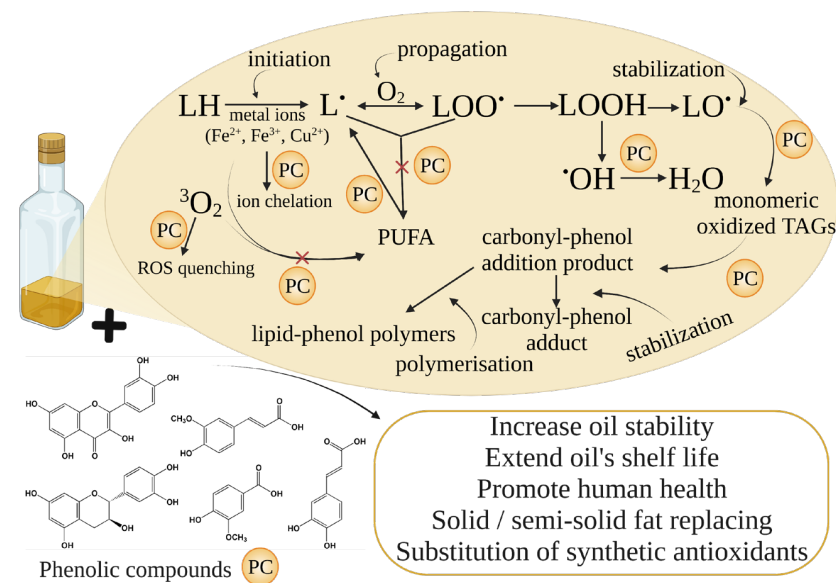


Figure 7. An overview of the oxidation reactions.

As is well known, today's trend is to substitute synthetic compounds from foods with natural substances that play the same role without changing the properties of the products. In this way, Chougui et al. [114] frameworked the industrial exploitation of by-products of the *Opuntia ficus-indica* (prickly pear) extract in the substitution of Tocoblend (vitamin E), a synthetic antioxidant used as a preservative in margarine production. The results showed that the extract rich in phenolic compounds (1512.58 mg GAE/100g DM) managed to extend the shelf life of margarin without any effect on its physicochemical and microbiological properties. Although it was tested at different concentrations, the extract exerted better effects compared with vitamin E, even at half the concentration of the latest [114]. When analyzing this, an important application of enriched vegetable oils with phenolics is the ability to substitute synthetic compounds with natural ones obtained from by-products.

Another study focused its attention on the ability of tea polyphenol palmitate to stabilize camellia oil-based W/O (water-in-oil) emulsions (Figure 6C). Because of its rich content in unsaturated fatty acids (especially oleic acid) and other functional compounds, camellia oil was cataloged as a potential substitute for solid or semi-solid fats. The scientists used tea polyphenol palmitate to stabilize the emulsion and, in this way, proposed a method by which the fat could be replaced with a more stable and surfactant-free emulsion. Moreover, their final product had increased storage stability and a better freeze–thaw stability. Taking into consideration the visual appearance and the rheological characterization, it was proven that enriching a camellia oil-based emulsion with tea polyphenol palmitate can be a promising solid fat replacer and a better one compared with margarine because of its higher slip melting point (SMP) [115].

Şahin et al. [116] recently conducted a study regarding the enrichment of hazelnut oil with several polyphenols as an alternative approach to a novel functional food. The oil sample was supplemented with six compounds, among which gallic acid showed enhanced stability against oxidation three times greater than the pure sample itself. This study strengthened the premise that enriching edible oils with polyphenols increases oxidative stability [116].

Additionally, one study analyzed the effect of a phenolic extract obtained from walnut press cake on oxidative stability of walnut oil. The press cake is a valuable source of phenolic compounds which shows a reduction of the formation of oxidation products in the sample that contains it compared with the control. The results showed that 200 mg/kg of oil phenolic extract from a walnut press cake successfully stabilized the tested oil, making the enrichment a potential natural antioxidant alternative [117].

In 2021, Abenoza et al. [118] implemented a strategy to increase the shelf life of Empeltre virgin olive oil by incorporating flavoring agents rich in antioxidant compounds such as garlic and rosemary. The enrichment increased the total phenolic content, and as a consequence, the antioxidant capacity and oxidative stability were enhanced, especially when the incorporation was carried out during malaxation. From the sensory point of view, garlic aromatization achieved a better score compared with rosemary, which was more effective in increasing the phenolic content. Further studies are necessary in order to obtain the best flavoring agent, but it is important to obtain a high phenolic content in virgin olive oil, as European regulations recognized it as a related health claim [118].

Aside from improving the stability, nowadays, there is a growing interest in studying new formulations to also improve the functional properties of foods. Recently, many natural antioxidants have been obtained from agro-industrial waste and used in the food industry. Cocoa bean husk, which has long been considered a waste, showed in several studies its biological and potential health benefits due to its phenolic content, making it susceptible for incorporation in food products. The chosen product was extra virgin olive oil jam, as in its case, the prevention of oxidation of both phases is crucial, as is a heterogeneous matrix formed by oil and water. The study analyzed the efficiency of both encapsulated and non-encapsulated extract from cacao bean husk and observed that for foods with higher contents of oil, it would be recommended to not use encapsulation, and in the case of a higher aqueous content, encapsulation is more appropriate. The results showed that the enrichment improved the stability and antioxidant activity of the tested product [119].

Approaching the sustainability point of view, olive mill wastewater contains high amounts of phenols and their derivatives, which are valuable, but its disposal is toxic, and it is difficult to biodegrade. In this way, the recovery of phenolic compounds represents a promising approach to obtaining suitable compounds for food enrichment. Romeo et al. [120] conducted a study aimed to evaluate the efficiency of the enrichment of sunflower oil with a phenolic extract from olive mill wastewater. In order to evaluate the resistance of oil to oxidation, the samples were subjected to a high oxidative stress environment, and increased stability was observed, which was associated with the addition of phenolic extract. Thus, the researchers successfully valorized a by-product from the oils industry by converting it from waste to resource [120].

Aside from the oil itself, there is also a focus on the prevention of oxidation in the case of blends. Edible oils such as olive, soybean, and rice bran oils are considered long-chain triacylglycerols (LCTs) and possess monounsaturated and polyunsaturated fatty acids with tocopherols, and their consumption requires several steps for digestion. The excess is likely to be stored as fat, causing different health-related issues. In order to reduce this risk, LCTs are blended with MCTs (oils with medium-chain fatty acids), which are rapidly digested, representing a quick source of energy. Thus, preventing the oxidation of edible oil blends was the purpose of More et al.'s [121] study. They incorporated moringa leaf extract and pomegranate peel extract in an edible oil blend, and the rate of oxidation was

clearly retarded. They concluded that the obtained results were comparable or at times better than the effect of chemical antioxidants such as BHA and BHT [121].

As presented, the oxidative stability of edible oils can be enhanced using innovative and sustainable technologies, but aside from this principal aim, the enrichment gives the final product different beneficial properties, making it a functional food in most cases.

5.2. Conventional and Emerging Strategies for Edible Oil Enrichment

Spices, herbs, vegetables, and fruit are the main sources of natural antioxidants suggested as alternatives to synthetic molecules to counteract, hinder, or delay the peroxidation of edible oils. The comparison of natural antioxidants with BHT or BHA was the topic of several studies aimed at identifying the most effective and proper concentrations of natural extracts to maintain oil stability. The extracts obtained from vegetable sources are rich in flavonoids, phenolic acids, and tocopherols which, as highlighted in the previous paragraphs, play a role in oil's protection by slowing down the radical chain reactions occurring during lipid peroxidation.

Several papers reported the use of plant extracts as stabilizers of oils subjected to accelerated storage conditions or thermal treatments due to cooking. Rosemary, sage, oregano, olive leaves, pomegranate, mangosteen peels, and myrtle leaves are some examples of the crude extracts studied and proven to be as effective as BHT or BHA in protection against lipid peroxidation during the shelf life or frying. Mangosteen peel extracts at 200 ppm delayed sunflower oil lipid oxidation like BHA [77]. Similarly, sesame cake extracts provided an effective inhibition of the thermal degradation of sunflower and soybean oils [122]. The enrichment of extra virgin olive oil (EVOO) with *Myrtus communis* L. phenolic compounds prevented the consumption of endogenous antioxidants in EVOO oil subjected to flame, oven, and microwave heating [78]. Recently, the methanolic extracts from the petals of 23 edible flower species have been used to enhance the oxidative stability of cold-pressed chia seed and flax oil. The flower species differed in their chemical compositions and their protective effects against oil oxidation, with the flowers with the highest concentration of flavonoids and carotenoids being the most effective. The authors associated the highest protection with the possible involvement of synergy between both flavonoids and tocopherols as well as tocopherols and carotenoids [123]. The effectiveness of the natural antioxidants on the oxidative stability of edible oils and the proper extracts' amounts have been recently reviewed [2,7,124]. In this paper, we focus on the emerging and sustainable strategies employed to meet the consumer's demand for natural and safe food additives. In addition, we target the technologies used to increase the solubility of the extracts in the oil's matrix.

In the last few decades, side streams and by-products from the food and beverage processing industries have become the most interesting sources of antioxidant compounds. Food wastes or food side streams and by-products, despite the processing they undergo, still retain a considerable amount of bioactive compounds, with health-promoting properties that may be also able to protect edible oils from oxidation.

Table 1 summarizes some of the studies dealing with oil's enrichment with extracts recovered from food wastes or by-products.

Carotenoids extracted from tomato peel by-product stabilized refined olive and sunflower oil against oxidation occurring during long storage periods at a concentration of 250 and 2000 $\mu\text{g g}^{-1}$, respectively. The tomato by-products' protective effect was correlated with the amount of lycopene [125]. Pomegranate peel extract containing ellagic and gallic acid, punicalina, and punicalagina had a positive role in the stabilization of pomegranate seed oil stored under accelerated aging conditions. When applied at a concentration of 0.01%, it effectively decreased the concentration of the secondary oxidation products, while when applied in combination with reduced doses of BHT, it inhibited the formation of the primary oxidation products without affecting the concentration of fatty acids [10]. Grape seed extract enhanced the oxidative stability of sunflower oil subjected to convective and microwave heating [126]. Flavonoids and flavanols recovered from sesame oil extraction by-products inhibited thermal-induced lipid peroxidation by inhibiting double-bond con-

jugation and decreasing the loss of PUFA [122]. Water extracts obtained from olive mill wastewaters rich in hydroxytyrosol-, verbascoside-, and tyrosol-protected sunflower oil stored for 90 days at 25 °C [120].

The supplementation of sunflower oil with freeze-dried extracts (rich in pyrocatechol, rutin, and phenolic acids) obtained from blueberry processing by-products inhibited the formation of primary and secondary oxidation products in oils subjected to convective heating at 180 °C for 12 h to simulate frying conditions.

The analysis of the literature highlights the growing interest in the recovery of natural antioxidants from by-products and their use as a sustainable method for the protection of oils from oxidation. However, many times, the sustainability does not entail the whole extraction process. As can be observed in Table 1 (please see the type of extract column), green solvents or sustainable extraction methods have not always been used to recover natural antioxidants. In many cases methanol or hexane have been used, which cannot be considered sustainable solvents.

The stabilization of oils with phenolic compounds may be affected by several factors, like their low stability during processing, their poor solubility, and their interaction with fatty acids and with the minor components of the oil. The thermal stability and solubility are important characteristics to consider in the selection of the proper extract in oil oxidative stability. When added to bulk oils, most antioxidant extracts are poorly soluble, and being thermolabile in heated oils, they may lose their antioxidant activity. In a recent paper, Tinello and Lante [127] studied the protective effect of ginger and turmeric extracts on soybean oils stored for 28 days at 62 °C, ascribing their strong effectiveness to their high solubility in oil and the resistance to thermal degradation. Phenolic compounds may have different thermal stabilities, depending on their chemical structure, with the cleavage of double covalent bonds being the main thermal degrading mechanism described [128]. For example, (–)-galocatechin, the main component of litchi juice, disappeared after storage at 45 °C for 72 h [129]. Regarding the solubility of natural antioxidants added to the oils, it depends on the extraction solvent and on the phenolic composition of the extracts. Nwuha et al. [130] observed that the direct solubilization of a green tea extract in edible oils (oleic acid, rapeseed, sunflower, and coconut oils) was not successful at the applied temperatures (20, 40, and 62 °C), while a sunflower oil–ethanol mixture increased the extract solubility. It must be pointed out, however, that solubility, although very important, is not the only factor to consider. Mohanan et al. [131] compared the protective activity of natural compounds with a different degree of hydrophobicity in flaxseed oil oxidation. They observed that ascorbyl palmitate, a compound with intermediate polarity, was as effective as TBHQ, while tannic acid, a hydrophilic compound, and α -tocopherol, which is highly soluble in oil, were not able to prevent oil oxidation. On the contrary, α -tocopherol has shown pro-oxidant properties. This study highlights that aside from the natural antioxidant's polarity, the interaction with the minor compounds present in the oils determines the oxidative protection of the oils. Recently, the encapsulation technique has been suggested to protect the extracts during thermal treatments and to increase their solubility in oils [124]. Ethyl cellulose particles were loaded with tocopherol to stabilize this antioxidant at high temperatures. Ethyl cellulose particles with different viscosities were prepared, showing that the antioxidant effect increased with the viscosity [132].

Table 1. Extracts obtained from food wastes and by-products used to enhance the oxidative stability of edible oils.

Type of Waste or By-Product	Type of Extract	Extract Concentration in the Oil	Oil-Enriched and Oxidative Treatment Conditions	Analytical Approach	Main Results	References
Olive mill wastewaters	Water extract (WE)	50 mg hydroxytyrosol L ⁻¹ of oil	Sunflower oil (SFO) stored for 90 days at 10 or 25 °C	Refractive index, peroxide value, oxitest oxidation test reactor, DPPH, ABTS, and ORAC	WE increased the oxidative stability of SFO and decreased the peroxide value	[120]
Tomato peel and seed by-product	Oleoresin extracted with hexane	250, 500, 1000, and 2000 µg g ⁻¹ of oil	Refined olive oil (ROO) and SFO Storage for 19 weeks at 50 °C	Free acidity, peroxide value, k ₂₃₂ , k ₂₇₀	250 and 2000 µg g ⁻¹ increased the stability of ROO and SFO, respectively	[125]
Olive mill wastewaters	Water extract	500, 1000, 2000, and 3000 mg L ⁻¹ of oil	Extra-virgin and refined kernel olive oil Heating at 100 °C for 30 min Heating at 160 °C for 120 min	DPPH, peroxide value, k ₂₇₀ , Totox index	Olive polyphenols were efficient against oxidation of both oil types at 1000–3000 mg L ⁻¹ . Oils treated with olive polyphenols and heated had similar levels of PV than fresh oils.	[133]
Mango peel by-product of mango industrial processing	supercritical CO ₂ extract	200, 500, and 1000 mg extract kg ⁻¹ oil	Sunflower oil (SFO)	Rancimat method	Mango peel extracts protected sunflower oil against lipid oxidation when evaluated between 200 and 1000 mg extract kg ⁻¹ oil	[134]
Sesame cake by-product of oil extraction	Methanolic sesame cake extract (SCE)	5, 10, 50, 100, and 200 mg kg ⁻¹ of oil	SFO and soybean oil (SBO) Stored for 72 h at 70 °C	Peroxide value, <i>p</i> -anisidine, k ₂₃₂ , k ₂₇₀ , Schaal oven test	SCE at 200 mg kg ⁻¹ inhibited SBO oxidation in SFO SCE controlled peroxide and <i>p</i> -anisidine formation	[122]
Olive mill wastewaters (OMWW) and olive pomace (PO)	Ethyl acetate extract	1 mg g ⁻¹ of oil	SBO subjected to accelerated oxidation at 60 °C for 21 days	Schaal oven test, k ₂₃₂ , and k ₂₇₀	Under accelerated oxidation conditions, PO and OMWW extracts were more effective than BHT. They decreased K ₂₃₂ but had no effect on K ₂₇₀ .	[135]
Blueberry juice by-product (BBE)	Hydro-alcoholic extract,	200, 500, and 800 mg kg ⁻¹ of oil	SFO Simulated frying conditions at 180 °C for 12 h	Peroxide value, <i>p</i> -anisidine, Totox, thiobarbituric acid test	BBE 800 mg kg ⁻¹ inhibited the formation of primary oxidation products, BBE significantly inhibited lipid thermo-oxidation, and BBE at 500 mg kg ⁻¹ was as effective as BHT 200 ppm.	[3]

Table 1. Cont.

Type of Waste or By-Product	Type of Extract	Extract Concentration in the Oil	Oil-Enriched and Oxidative Treatment Conditions	Analytical Approach	Main Results	References
Pumpkin peel	Water extracts obtained with subcritical water extraction (SWE) Ethanol or water extracts obtained with supercritical CO ₂ extraction (SFE)	400 mg kg ⁻¹ of oil	Canola oil Storage at 30 °C for 60 days	Peroxide value, carbonyl value, acid value	Both SWE and SFE inhibited the formation of peroxides and carbonyl compounds during storage. The mix of SFE and SWE was more effective than the extracts alone. Oils with SFE had lower acidity than SWE-enriched oils.	[136]
Peel (AP) and seeds (AS) from avocado waste	Freeze dried ethanolic extract	0.5% (<i>w/w</i>)	Avocado oil and water emulsions and nanoemulsions stored for 50 days at room temperature, oxidation was induced by the addition of Fe ²⁺ solution	Peroxide value and thiobarbituric acid test	AP and AS had no effect on the inhibition of peroxide production but accelerated their degradation. Ap and As limited the formation of secondary oxidation products.	[137]
Rice bran, rice hull, wheat bran, wheat husk, peels of citrus, banana, apple, pomegranate	Methanolic extracts	600 mg kg ⁻¹ of oil	Corn oil heating at 60 °C (8 h of heating per day) for 30 days	Peroxide value (PV), <i>p</i> -anisidine, conjugated dienes (<i>k</i> ₂₃₂), conjugated trienes (<i>k</i> ₂₆₈)	Methanolic extracts decreased the rise in peroxide value, conjugated dienes, conjugated trienes, and <i>p</i> -anisidine compared with the control. Pomegranate peel extract was the most effective, while rice hull extract was the least effective.	[138]

Nano-encapsulation of olive leaf extracts with pectin-WPC (wood plastic composite) increased the extracts' antioxidant activity and protected against the oxidation of soybean oil at a level comparable to that of TBHQ and higher than the extract alone, but it decreased the thermal stability of the extract [139]. Phospholipid-quercetin formulations increased the oxidative stability of camelina oil subjected to accelerated storage conditions [140]. The loading of α -tocopherol in ethyl cellulose particles significantly improved the thermal stability of α -tocopherol and hindered the oxidation of soybean oil heated at 180 °C for 25 h [141].

Methanolic extracts of *Trifolium resupinatum* were used in combination with lecithin to produce nanoliposomal structures which effectively protected the soybean oil against oxidation processes, as shown by the PV and TBA [142].

Other authors investigated the stabilization of fish oil-loaded electrosprayed capsules with dextran or glucose as the main biopolymer wall material, with an ethanol extract from the brown alga *Fucus vesiculosus*, or with a combination of δ -tocopherol and rosemary extract [143]. The seaweed extract enhanced the stability of the fish oil when the main biopolymer was dextran, while δ -tocopherol in combination with rosemary extract contained in glucose capsules stabilized the fish oil during 21 days of storage.

Extracts of green coffee fruits, either of the Arabica and Robusta variety, were microencapsulated with maltodextrin and gum Arabic and were tested to delay sunflower oil oxidation [144]. The microencapsulated extracts of green coffee fruits were more effective in delaying sunflower oil oxidation than the free extracts and BHT.

Microwave-assisted olive leaf extracts encapsulated by gum Arabic and maltodextrin were used to improve the thermal stability of soybean oil [145]. Encapsulation increased the thermal stability during 20 days of storage at 55 °C in comparison with the free extracts, but the encapsulation decreased the antioxidant activity of the extracts.

Encapsulation has the advantage of improving the bioactivity and bioavailability of the extracts poorly soluble in oils, protecting them from thermal degradation. It also decreases possible pro-oxidant activities and provides their controlled release. However, as seen in these examples, the results depend on the treatment to which the oils are subjected, being very different when comparing prolonged storage with short heating at high temperatures.

5.3. Safety Concerns Associated with the Use of Natural Antioxidants

Studies have shown that phenols are a good choice when it comes to improving the quality of vegetable oils, managing the performance by increasing the shelf life, stability, and their nutritional qualities. Aside from being known as effective natural antioxidants, phenols managed to impress with their antimicrobial, anti-inflammatory, and anti-cancer properties, as demonstrated by numerous researchers who have been studying these compounds for several decades. However, in some cases, certain polyphenols showed cytotoxic effects on the body. A relevant example is gossypol, a polyphenol found in cottonseed oil known for its toxicity but also for its ability to induce infertility in men [146].

This double-edged blade has led researchers to study these compounds in more detail to conclude whether phenols could actually be used *in vivo* or whether all the qualities they provide are theoretical, and the results can only be obtained from *in vitro* studies. The reason why some researchers might consider the use of polyphenols only for *in vitro* experiments is the low concentration used, because a high concentration of polyphenols can lead to toxicity due to their hormetic nature. The hormetic phenomenon refers to the ability of certain compounds to behave as stimulants when administered in low doses and as toxic agents when administered in high doses [147]. On the other hand, a study reported by Martin [148] revealed that caffeic acid, a polyphenol frequently used in oil enrichment, can be a tumor suppressor at a 0.5–5 g kg⁻¹ diet, a tumor promoter at a 5–10 g kg⁻¹ diet, and a carcinogenic compound at a >20 g kg⁻¹ diet. The same study showed that ferulic acid, another main antioxidant used in the food industry, exerted both antitumoral activities on the tongue, skin, and colon and pro-tumoral activity on the liver when administered to rats at a 500 mg kg⁻¹ diet [148]. Islam et al. [149] also pointed out that vanillic acid can induce acute toxicity in mice and rats at LD50 with a 2.691 and 5.020 mg kg⁻¹ diet, syringic

acid in rats at LD50 with a $>2000 \text{ mg kg}^{-1}$ diet, and gallic acid in BEAS-2B (transformed or non-tumorigenic cells) at $25 \mu\text{g mL}^{-1}$ [149].

Even if all these results may seem alarming, it should be noted that these results are related to mice and rats. Cladis et al. [150] performed transformation of the data related to rats in order to find out the human polyphenol intake in a novel study. The team publicized that the no-observed-adverse-effect level (NOAEL) for blueberry polyphenols is $\geq 1000 \text{ mg kg}^{-1} \text{ bw/d}$ for OVX-SD rats, translating to a 70-kg human consuming $\sim 10 \text{ g}$ of blueberry polyphenols per day. The obtained amount is higher compared with the current available dietary supplements, so the researchers concluded that blueberry polyphenols are safe to consume [150]. However, more studies are needed to ascertain whether a particular extract of a specific by-product or food waste containing well-identified molecules exhibits or does not exhibit toxic effects toward human health and in which range of concentrations its use can be considered safe.

6. Oil Blending as a Tool to Improve an Oil's Oxidative Stability

The blending of two or more seed oils is an old approach employed to modify the fatty acid profile and to improve an oil's nutritional quality. The edible oil industry often uses the blending practice to produce blended oils with enhanced oxidative stability and improved functional characteristics [151]. This practice has gained renewed interest after the introduction in the market of new oils extracted from unconventional oil-bearing seeds. The blending with this kind of oils has economic and functional advantages, changing the physico-chemical and nutritional properties of the blends [151]. The main effect of blending is the modification of the fatty acid profile, particularly the ratio of saturated, mono-, and poly-unsaturated fatty acids and the ratio of essential fatty acids ($\omega 3:\omega 6$). Blending with an oil with a high unsaturation degree increases the nutritional properties and the health effects of the oils, but the drawback is that the use of a less stable oil may decrease the blend's oxidative stability during storage or cooking. Careful blending should be modulated based on the final use of the blended oil. In a diet rich in essential fatty acids, a healthy $\omega 3:\omega 6$ ratio, important in the prevention of many diseases, may be achieved by mixing oils to obtain a balanced fatty acid composition [152,153]. The mixture of sunflower oil with 20% cold-pressed flaxseed oil produced an oil with an optimal $\omega 3:\omega 6$ ratio and with an enhanced level of bioactive compounds like tocopherols and carotenoids [152].

Table 2 reports some of the studies dealing with the effect of oil blending on the oils' oxidative stability. The blending of linseed oil with corn, sesame, and bitter almond oils at different ratios enhanced the linseed oxidative stability as determined by the Rancimat method at 353, 373, and 393 K [154]. The blending of *Moringa oleifera* oil with sunflower and soybean oil, by changing the proportion of MUFAs and PUFAs, determined an enhancement in the oxidative stability of the oils during storage at room temperature for 180 days and increased the oils' stability during heat treatment at $180 \text{ }^\circ\text{C}$ for 42 h, with a 6-h heating cycle per day [155]. Chia oil is rich in $\omega 3$ fatty acids that own beneficial properties for human health but are highly susceptible to oxidative damage. The blends of chia oil with walnut, almond, or sesame oils increased the oxidative stability of the oils subjected to accelerated storage conditions while keeping a high level of $\omega 3$ fatty acids [156]. The blending of sunflower oil with pomegranate seed oil (85:15) determined an increase in the oxidative stability during an accelerated storage test at $60 \text{ }^\circ\text{C}$ for 20 days with respect to sunflower oil alone [157]. The addition of 15% pomegranate seed oil to the sunflower oil changed the fatty acid profile by increasing the concentrations of the primary saturated fatty acids like palmitic and stearic acids and enhanced the levels of the phenolic compounds, carotenoids, and α -tocopherol.

Table 2. Oil blends used to enhance the oxidative stability of edible oils.

Oil Blend (First + Second Component)	Concentration of the Second Blend Component	Analytical Approach	Main Results	References
Sunflower oil + Sclerocarya oil	10, 20, 30, and 40%	Oxidative stability (Rancimat 120C) and stability at 70C using peroxide value.	The oxidative stability in the Rancimat test was improved from 47 to 147% in oil blends compared with sunflower oil alone. Storage of the blends at 70 °C showed that peroxide value of the blends was remarkably lower than sunflower alone.	[158]
Sunflower oil (SFO) + <i>Moringa oleifera</i> oil (MOO)Soybean oil (SBO) + MOO	20, 40, 60, and 80%	Storage test: oils stored at room temperature for 180 days. Peroxide value (PV), iodine value (IV), and induction period (IP, Rancimat test) were evaluated monthly. Thermal stability test: heating at 180 °C for 42 h (6-h heating cycles each day).	The storage test showed an improvement of the oxidative stability of blends with increasing MOO concentration. Each 20% addition of MOO resulted in decreases of PV and an increases of IP. The heating performance test revealed the SBO:MOO (20:80 <i>w/w</i>) blend was the most stable.	[155]
Coconut oil + palm oil Coconut oil + sunflower oil Coconut oil + soybean oil	20, 50, and 80%	Storage test: 6 weeks at 37 °C. Peroxide value (PV), fatty acid composition, and antioxidant activity.	Blending coconut oil with other vegetable oils increased the medium chain fatty acid concentration and the oxidative stability to the blends.	[159]
Rapeseed oil + rice bran oil Rapeseed oil + black cumin oil	5, 10, and 20%	Oxidative stability index (Rancimat test), phytosterols, fatty acids, and tochochromanols.	The blends with black cumin seed oil had higher level of α - and γ -tocopherols and all isomers of tocotrienols. Blends with rice bran oil had high tocotrienol, β -sitosterol, and squalene levels. Blending resulted in lowering ratio of PUFA:SFA and improved the stability of these oils.	[160]
Sunflower oil (SFO) + cold-pressed black cumin oil (BCO)	5, 10, and 20%	Thermally accelerated oxidation conditions with Rancimat method (110C) and Schaal oven test (60C). Peroxide value (PV), conjugated dienes (CD), and conjugated trienes (CT). Volatile oxidation compounds.	BCO increased the stability of SFO at high temperatures according to induction period times. SFO:BCO (80:20) blends had lower PV at the end of storage at 60 °C (Schaal oven test). Stability of blends was better than SO, most likely due to changes in the levels of thymoquinone and tocopherols found in BCO.	[161]
Butter oil + crude mango kernel oil (MKO)	2.5, 5, 7.5, and 10%	Storage at 25 °C and 55 °C for 6 months. Induction period (rancimat methd) peroxide values, anisidine values, and iodine values were measured at 0, 45, and 90 days.	Supplementation of MKO oil in butter oil inhibited oil oxidation at ambient and accelerated oxidation, as showed by the low PV and anisidine values of blends after 90 days of storage. Induction period of all the treatments increased as a function of the addition of MKO.	[162]

Table 2. Cont.

Oil Blend (First + Second Component)	Concentration of the Second Blend Component	Analytical Approach	Main Results	References
Chia seed (<i>Salvia hispanica</i> L.) oil (CO) + walnut (WO), almond (AO), virgin and roasted sesame oils (VSO, RSO)	60, 70, and 80%	Accelerated storage test (Schaal oven test (SOT) 40 ± 1 °C, 12 days). Peroxide value (PV), conjugated dienes (CD), conjugated trienes (CT), p-anisidine value (PAV), free fatty acid content, antioxidant compounds, and induction time (Rancimat method).	The blends presented a higher induction period than chia oil alone. Sesame oil blends (VSO and RSO) were the most stable, followed by CO:AO blends.	[156]
Perilla seed oil (PO) + extra virgin olive oil (EVOO)	0, 55, 60, 65, 70, 75, and 100%	Free acidity, peroxide value, conjugated dienes (CD), conjugated trienes (CT), fatty acid composition, sterols, tocopherols, biophenols content, and oxidation stability.	Blends were rich in ω -3 and ω -6 fatty acids, biophenols, tocopherols, and sterols. Blends showed higher oxidation stability than PO alone.	[163]
Linseed oil + corn, canola, sesame, and bitter almond oils	33, 66%	Oxidative stability index (OSI) as determined by the Rancimat method at 353, 373, and 393 K.	Blending linseed oil with 66% corn, bitter almond, sesame, and canola oil increased the OSI value of linseed oil by 67.05%, 61.68%, 60.12%, and 59.13%.	[154]
Chia oil + sesame oil	84.5, 89.6, 92.3, and 93.9%	Oxidative stability index (OSI), with the Rancimat method, and shelf life (oxidative stability index at 25 °C, OSI ₂₅). Acidity, peroxide value (PV), p-anisidine value (p-AV), and the total oxidation value (TotOx).	The supplementation of chia oil with sesame oil increased the OSI value and the shelf life (oxidative stability index at 25 °C, OSI ₂₅).	[164]
Soybean oil (SBO) + sesame seed oil (SSO)	40, 50, and 60%	3 frying cycles. At the end of each cycle, the free fatty acid (FFA), peroxide value, and acid value were determined. Characterization of fresh and fried oil blends by Fourier transform infrared spectroscopy (FTIR).	The blend with the ratio of 60% had the lowest peroxide value. The blending of SBO with SSO improved the thermal stability, as sesame seed oil contains high levels of antioxidants such as sesamin, sesamol, and tocopherol.	[165]

Table 2. Cont.

Oil Blend (First + Second Component)	Concentration of the Second Blend Component	Analytical Approach	Main Results	References
Virgin olive oil (VOO) + sesame (SSO), flaxseed (VFO), hazelnut (VHO), and pistachio (VPO) oils	SSO 5, 15% VFO 3, 10% VPO or VHO 50, 75%	Oxidative stability by Rancimat method. Shelf life testing at room temperature for 24 days (25 °C) and under accelerated shelf-life conditions (55 °C) (ASLT).	The oxidative stability test showed that the oil blends maintained similar stabilities of VOO under normal and accelerated storage conditions.	[166]
Sunflower oil (SFO) + pomegranate seed oil (PSO)	10, 15, 20% (<i>w/w</i>)	Accelerated storage test was conducted using the best blending ratio (85:15) at 60 ± 2 °C for 20 days following the evolution of the peroxide value, ρ -anisidine value, and total oxidation value.	Blending of SO with PSO improved total phenols, total carotenoids, and tocopherol content. Blended oils exhibited better oxidative stability than SO, as shown by the low peroxide and ρ -anisidine values. Blending SO with PSO decreased the rate of hydroperoxides formation during the accelerated storage period.	[157]

The resistance to oxidation of the oil blends is strongly affected by the chemical features and the relative percentage of the starting oils in the blends. An oil with a high unsaturation degree is more susceptible to oxidation, and thus its percentage in the blend should be tailored while considering its fatty acid composition and the presence of natural antioxidants.

7. Conclusions

The stabilization of edible oils and their protection from oxidation would be favorable for oil producers and food processing industries, allowing a better exploitation of the resources and supplying safer products to consumers. Natural antioxidants have been suggested as alternatives to synthetic additives for the protection of edible oils against oxidative damage. The use of natural antioxidants recovered with sustainable methodologies from food wastes or by-products originating from food processing industries is an emerging approach in the perspective of a circular economy and to meet consumers' demand for natural and safer foods. The reuse and exploitation of new unconventional and alternative sources, such as the wastes of food processing industries, represent key issues in this direction, following the green transition and the principles of a circular economy.

The literature survey conducted in this review highlighted a huge amount of papers explaining the role of antioxidants in the protection of edible oils against autoxidation during storage or thermal treatments due to cooking. However, the sustainability of the biomolecule extraction processes and the design of proper oil enrichments in terms of quality and the amount of extracts to be added are moving through their first steps, and further studies are needed.

Furthermore, the enrichment methods should be further developed both to protect the extracts from thermal damage during high-temperature treatments occurring in frying and cooking conditions and to provide a controlled release of the active ingredients. The development of nanotechnologies for the encapsulation of non-lipophilic antioxidants are a step in this direction. This review has also considered the safety concerns for human health associated with the use of natural antioxidants, pointing out a double effect of polyphenols. This highlights the need for a multidisciplinary and integrated approach in the studies of stabilization of edible oils with natural antioxidants.

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