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Fruit-based natural antioxidants in edible oils: a review

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Abstract

The aim of this work is to review several studies on improving the oxidative stability of vegetable oils in order to increase their resistance to high temperatures. This study aims to gather knowledge about the development of vegetable oils with an increased oxidative stability of the lipid fraction by adding natural extracts with antioxidant action. Natural antioxidants represent the healthy alternative to synthetic antioxidants. Berries, such as certain species belonging to the Rosaceae and Ericaceae families, represent important sources of bioactive compounds. The antioxidant activity of berries is due to phenolic compounds. The richest parts of a fruit in natural bioactive compounds are peels and seeds. Thus, in order to obtain potential natural antioxidant extracts, by-products resulting from the industrial processing of fruits, will be used. Incorporating antioxidant extracts obtained from by-products generated to industrial processing of some berries, in edible vegetable oils, leads to an increase of their phenolic profile and stability against the oxidation process. The available data on enhancing oxidative stability of vegetable oils by adding natural extracts with antioxidant properties obtained from fruit processing by-products, lead to the development of new technologies for obtaining oils with improved antioxidant function.

Keywords: berries, berries processing by-products, natural antioxidants, phenolic compounds, oxidative stability of edible oils

1. Introduction

Due to their sensitive and nutritional role, oils and fats are important ingredients in the human diet. They constitute the basic component in the manufacture of certain products in food industry, pharmaceutical and cosmetics industries. Lipid degradation is mainly determined by the oxidation reaction of unsaturated fatty acids [1].

Lipid degradation has an impact on the product's final quality, because oxidation causes an unpleasant taste, non-specific flavors, rancid smell and quality changes. Also, nutritional value decrease in the products containing oils and fats, thus, compromising the product's safety [1, 2]. Lipid oxidation can be accelerated by applying thermal processes [1]. Oxidized oils and fats can ultimately have a negative effect on human body, such as DNA cleavage, RNA modification, lipids and proteins modification, can cause cellular damage and may induce further promote inflammation and carcinogenesis. By adding in low quality commercial oils some antioxidant components from dry residue resulting from fruits processing, high quality oils with improved quality and features can be obtained. The procedure results not only in slowing the oils oxidation, but also in the provision of various functional components such as polyphenols [3].

Antioxidants represent a class of chemicals that help prevent and repair cell damage caused by free radicals. Antioxidants can be synthesized by chemical processes or can be of natural origin from plants and animals [4]. In recent years, numerous studies have been done to add natural plant extracts in edible oils thus, avoiding the use of synthetic additives with potentially harmful action [5, 6]. Oxidative stability during storage, exposure to light

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and thermal stability during thermal processing, are the most important aspects followed for an antioxidant added to oils and fats [7].

Many studies have demonstrated that natural additives added to edible oils have shown a higher thermal stability compared to synthetic ones [2, 8]. Synthetic additives such as butylated hydroxyl anisole (BHA), butylated hydroxyl toluene (BHT), propyl gallate (PG) and tertiary butyl hydroquinone (TBHQ) often used to increase the oxidative stability of vegetable oils, can easily be replaced by phenolic compounds which are natural additives of vegetal origin with antioxidant and antimicrobial activity [2].

Agro-industrial products can be considered a viable alternative to synthetic antioxidants used to increase oxidative stability in food, cosmetics and pharmaceuticals due to phenols that are valuable compounds with antioxidant properties. Also, this solution is a cheap source of antioxidant compounds, using by-products, residues from industrial processing [9].

In line with the above mentioned considerations, the purpose of this work is to bring in attention several studies to highlight the potential of fruit-based natural antioxidants against lipid oxidative degradation develop in foods, especially in edible oils.

2. Fruit-based natural extracts as potential antioxidants against lipid oxidative degradation in foods

Bioactive compounds in fruits and fruit processing by-products

The interest in researching various types of fruits (especially berries because they are important sources of bioactive compounds) has considerably grown in the last years. Species such as blackberries (*Rubus fruticosus*) and raspberries (*Rubus idaeus*) from *Rosaceae* family as well as bilberries (*Vaccinium myrtillus*) and cranberries (*Vaccinium vitis-idaea*) belonging to *Ericaceae* family, are the richest species in bioactive compounds [10, 11].

The bioactive compounds are important for nutritionists and food technologists because they can be valuable ingredients in functional foods. Antioxidants represented by phenolic compounds, dyes (e.g. anthocyanins and carotenoids), vitamins (such as ascorbic acid and folic acid) and minerals, belong to the category of bioactive compounds derived from berries. Bioactive compounds, individually taken or in certain combinations, exhibit high antioxidant capacity [11].

Antioxidants are components that prevent oils and fats oxidation by clearing free radicals that present an unpaired electron in the outer orbit, which results in instability and reactivity [2, 11]. The antioxidants are any substance that prevent, delay or remove the oxidative damage to a target molecule. Oxidation reactions produce free radicals that can trigger multiple chain reactions that can subsequently cause injury or even cell death. In food and pharmaceutical industries, synthetic antioxidants are used to extend the shelf life of products, but there is an increasing demand in replacing them with natural sources of efficient and available antioxidants, thus desiring to reduce the potential damage action on the body, caused by synthetic antioxidants [12]. Antioxidants show a real interest in preventing harmful effects of the free radicals on the human body and delay deterioration of fats and oils as well as other constituents from food. The most popular method in estimating efficiency of the antioxidants is based on using stable free radical 2,2-Diphenyl-1-picrylhydrazyl (DPPH). The evaluation of antioxidant capacity can be achieved by using reduction reagent Folin-Ciocalteu method and determination of antiradical capacity using ABTS radical action [13].

It is well known that phenolic compounds show multiple beneficial biologic effects including antioxidant activity. Phenolic compounds can come from edible plants and also from non-edible plants. The raw extracts coming from vegetal material, delay oxidative degradation of lipids and improve foods quality and nutritional value [14]. The phenolic compounds belong to a large and heterogeneous group of chemical components which has one or more aromatic rings with a conjugated aromatic system and one or more hydroxyl groups. They tend to donate an electron or a hydrogen atom to a free radical and turn it into an innocuous molecule [2, 11]. Phenolic compounds show benefits and well-being for human body health due to their antioxidant activity. They can be extracted from natural sources of plant origin such as fruits and berries (bilberries, blueberries, blackberries, cranberries, rosehips and grapes), vegetables (red beet, carrots, spinach and broccoli), various herbs and spices (rosemary, oregano, thyme, tea, tumeric), cereals, etc. [15]. Popularity of these compounds increased among consumers for coming from natural sources and show biological activity for health. Phenols have antimicrobial activity that can extend the shelf life of food products and can help to obtain functional foods with enhanced properties [16, 17].

Industrial waste resulting from fruits processing in order to preserve, may contain reusable substances with high nutritional and physiological potential [18]. Secondary products are important sources of sugars, minerals, organic acids, dietary fiber and phenols with extensive beneficial action including antitumor, antiviral, antibacterial, anti-mutagenic and cardio-protective activity [19]. The use of residual products is more and more common in the industrial sector for producing functional foods. By adding to various conventional foods, some by-products or extracts from them, high-quality foods with minimal costs will be obtained [18].

It has been paid a special attention in recent years to industrial waste containing residual phenols responsible for antioxidant action, especially for improving the oxidative stability of vegetable oils subjected to heating for delay the development of oxidation process of the lipid fraction. In conventional oils, these residual phenols assume the role of food lipid antioxidants [9, 20]. This has highlighted the industrial and economic potential of some secondary or by-products by many researches as is the case of the study performed by Poiana [14], where the extracts obtained from grapes seeds have contributed on the improvement of oxidative stability of sunflower oil [20].

Frying is a complex process and conditions applied most often overwhelm the capacity of endogenous or added antioxidants. Replacement of synthetic antioxidants with naturally occurring antioxidants is due both their weak performance in delaying oxidation and lower consumer acceptance of chemical synthesis stabilizers [21].

Many factors, such as antioxidant concentration, temperature, pH and processing treatment, influence antioxidant activity [2]. In order to make a comparison between the behavior of edible vegetable oils in which natural antioxidant compounds have been added and conventional oils, during thermal treatments, in order to demonstrate the inhibitory effect of polyphenol-rich extracts on lipid oxidation progress, it can be investigate the peroxide value, p-anisidine value, inhibition of oil oxidation, conjugated dienes and trienes and total oxidation value (TOTOX) [20].

Factors influencing the polyphenols extraction

In research carried out on extracts of berries growing in southern Brazil, including blackberries, Denardin *et al.* [22] evaluates the bioactive compounds as well as their antioxidant activity. After the achievement of phenolic compounds profile by high-performance liquid chromatography combined with diode array detection (HPLC-DAD) and antioxidant activity by FRAP (ferric-reducing antioxidant power) assay, DPPH (2, 2-diphenyl-2-picrylhydrazyl hydrate) assay, total reactive antioxidant potential (TRAP) assay and total antioxidant reactivity (TAR) assay, Denardin *et al.* concluded that wild fruits studied were rich sources of phenolic compounds with high antioxidant activity, among which in, the blackberry varieties present the highest values in compounds with antioxidant activity [22]. Also, after the investigation of antioxidant activity as well as bioactive components in some berries, Namiesnik *et al.* [23] concluded that bilberries are the best source of antioxidant compounds from the studied fruits [23].

In their study, Galanakis *et al.* [24] investigate the predisposition of the active coefficients of several natural phenols including gallic acid, caffeic acid, p-coumaric acid, rosmarinic acid and ferulic acid in solvents such as water, ethanol, methanol, ethyl acetate, acetone, diethyl ether and dichloromethane, at different temperatures. The obtained results indicate the superiority of alcohols and acetone to recover phenols. It has also been found that activity coefficient values increase with increasing temperature [24]. In their work on the recovery and preservation of phenols from olive residue in ethanolic extracts, Galanakis *et al.* [25] demonstrate that the extraction time between 30 and 120 minutes is not critical to the recovery/preservation process. Instead, addition of a pre-treatment step with the addition of ethanol in lower concentrations, adversely affects the recovery of phenols in the final ethanolic medium, as well as the addition of a preheating step of the residue, leading to reduction of phenol concentrations and a decrease in extract's antioxidant activity [25].

The study carried out by Burin *et al.* [26] has dealt with extraction of bioactive compounds from different grape varieties and assessing of antioxidant activity of the obtained extracts depending on the extraction methods. The results of this study have proven that among the extraction methods applied (liquid-liquid extraction, solid phase extraction, ultrasound extraction), liquid-liquid extraction was the best method for phenolic compounds extraction. Liquid-liquid extraction consisted in: an aliquot of 5 mL of grape juice was extracted twice with 10 mL of ethyl acetate by stirring with a vortex mixer for 5 minutes. Previously, type and volume of the extraction solvent as well as extraction time were determined. The organic phase of the two extractions were combined and subsequently evaporated in a rotary evaporator with a controlled temperature of 28±1°C and the residue was re-dissolved in 2 mL of methanol: water (1:1 v/v) [26].

In the research carried out by De Souza *et al.* [27] it has been investigated the chemical composition, identified the bioactive compounds and also, evaluated the antioxidant activity of different berries from Brazilian subtropical areas. Moreover, a comparison between results from subtropical fruits and results obtained from temperate zone berries has been performed. Regarding mineral content, fruits in subtropical zone showed lower concentrations in P, K, Ca, Mg and Zn and higher levels of Fe. Recorded values for bioactive compounds most closely matched the range reported in literature. The biggest difference was recorded in ascorbic acid content, fruits in the subtropical zone presenting a much higher level in ascorbic acid than those in the temperate zone. The results of this study strengthen and argue that the content of natural bioactive compounds in plant material is influenced by exogenous factors such as climate which requires certain temperature and humidity [27].

In another study, Zuorro & Lavecchia [28] investigated the recovery of phenolic antioxidants from bilberries (*Vaccinium myrtillus L.*) by-products. After separation of peels and seeds from the fruit pulp, peels and seeds were stored at -20°C in plastic bags. The frozen material was thawed at room temperature and moisture and total content of phenols and flavonoids was determined. For a complete extraction of the solid, a three-step extraction procedure was used, so that 1 g of husks were added in three stages of variable amounts of solvent (100, 50 and 20 mL) then is subsequently cast into a thermostats glass flasks at 40°C and stirred for 90 minutes. The resulting suspension was filtered and analyzed for total phenols and flavonoids. Aqueous ethanol (50% v/v) was used as an extraction medium and the total amount of phenols and flavonoids was represented by the sum of values obtained in each stage. Total amount of polyphenols was determined by Folin-Ciocalteu method. The experiment aimed to investigate the influence factors on the recovery of phenolic compounds from bilberries waste. Analyzed factors were: the liquid-solid ratio, the aqueous ethanol concentration, the extraction time and the temperature. Thus, to determine the influence of the extraction conditions on the phenolic compounds recovery, the liquid-solid ratio ranged between 20 and 40 mL g^{-1}, ethanol concentration was between 30-70% vol., the extraction time was from 90 to 210 minutes and the temperature ranged from 30 to 50°C. In the best extraction conditions (liquid-solid ratio 40 mL g^{-1}, ethanol concentration of 70% vol., extraction time of 210 min and temperature of 50°C), a recovery degree of phenolic compounds of 95.2% it has been noticed. From recorded results it has been seen that the liquid-solid ratio, ethanol concentration and temperature were the most influential factors on the phenolic compounds recovery [28].

Improving the stability and bioavailability of natural antioxidants by encapsulation

Encapsulation represents the technique by which, small particles or droplets are surrounded by a coating wall, the result being small capsules. This method also increases the stability of bioactive components to oxidation, evaporation, thermal treatments, also leading to a controlled release of encapsulated components [29].

Encapsulation of certain ingredients also has the advantage of protecting the core with the help of the capsule, against degradation or reaction with other ingredients while protecting food from any undesirable flavor of encapsulated ingredient. Encapsulated ingredients can easily be added in foods composition during processing having a homogeneous distribution. Another technological advantage that encapsulation offers, is the possibility of a delayed or controlled release of the ingredient in the capsule, release that can be triggered by factors such as time, pressure, temperature, pH and others. By applying different encapsulation materials and methods, microcapsules with specific attributes such as particle size and shape, deliberate release point of the core and others, may be obtained [30].

With the development of microencapsulation and its adaptation to food industry, a wide range of products with bioactive polyphenols have been created. Various studies have shown that polyphenols encapsulation is intended to protect their functionality and stability and increased bioavailability of these natural origin antioxidants [30].

Natural polyphenols do not have long-term stability being very sensitive to light and heat. Also, these natural compounds sometimes showed low bioavailability due to their low water solubility. In order to increase the polyphenols bioavailability and stability, delivery systems for these compounds such as encapsulation, have been developed. Encapsulation can be accomplished by physical methods (spray drying, fluidized bed coating, centrifugal extrusion, processes using supercritical fluids), physicochemical methods (cooling by spraying, melt coating, ionic gelling, extraction by solvent evaporation, simple or complex coacervation), chemical methods (interfacial or in situ polymerization, polycondensation, interfacial crosslinking) and related stabilization methods. Encapsulation aims to protect the fragile or unstable compound against environment, to capture compounds such as aromas, essential oils, organic solvents and to achieve a controlled release of the encapsulated compound [31].

Applications of natural antioxidants to inhibit lipid oxidation in foods

In the study performed by Fidaleo *et al.* [32] it has been investigated the bilberries waste, as a potential source of phenolic antioxidants. By extraction of polyphenolic compounds from bilberries by-product in a mechanical stirrer at 40ºC and 300 rpm using aqueous ethanol (60% v/v) as solvent, an extract with antioxidant activity has resulted. By incorporating bilberry extracts in different amounts, in various foods such as natural drinking yoghurt and condensed milk and following their antioxidant activity, Fidaleo *et al.* [32] demonstrates that bilberry residues can be used as a valuable source of phenolic antioxidants. These results support the affirmation that phenolic extracts can be used to produce new foods from functional foods category with a higher antioxidant activity.

In the research performed by Lorenzo Rodriguez *et al.* [33] it has been investigated the potential of berries as food additives in inhibiting lipids and proteins oxidation in meat and meat products. The results of this work highlighted that bilberries (*Vaccinium myrtillus*), blackberries (*Rubus fruticosus*), grapes (*Vitis sp.*) and cranberries (*Vaccinium vitis-idaea*) represent optimal sources of phenolic compounds, especially anthocyanins, these fruits extracts can be successfully used as natural antioxidants, in meat products [33].

Ahmad *et al.* [34] create an overview on natural fruit antioxidants and their application in meat and meat products. It highlights that different parts of the fruit, including by-products such as seeds and peel, are rich in natural antioxidants due in particular by phenolic compound. It emphasize the idea of replacing synthetic antioxidants with natural ones derived from plums, grapes, berries, pomegranates and citrus fruits, or from their by-products, as well as from extracts obtained from them [34].

The results published by Delfanian *et al.* [35], from the review of various extraction techniques such as ultrasound-assisted extraction, supercritical CO_2 extraction and solvent extraction of compounds with antioxidant properties from the *Eriobotrya japonica (Lindl.)* fruit peel, showed that solvent extraction was the most efficient method of extracting phenols and tocopherols. For solvent extraction, after harvesting and peeling the fruit, the fruit peels were dried in the sun and ground to a powder state. This powder was packed and stored at -20°C until subsequent extraction of the antioxidants. In this study, 20g of dried peel powder has been mixed with 100 mL ethanol. This mixture was protected from light and subsequently stirred at 160 rpm for 48 hours at room temperature. The extracts were filtered and solvent was evaporated at 50°C using a rotary evaporator. Concentrated extract was stored at -20°C until assayed. Total polyphenols content of extracts was determined using the Folin-Cicalteu reagent. The ability of extracts to clear the free radicals was determined by using DPPH method (2, 2-difenyl-1-picrylhydrazyl). Delfanian *et al.* [35] have also tracked the protective effects of extracts in stabilizing soybean oil against oxidation during frying. Enhanced oxidative stability effects of soybean oil during frying by adding phenol extracts were measured by determining the peroxide value, conjugated dienes and trienes, free fatty acids, total polar compounds and carbonyl value.

Results recorded by measurement of various values in soybean oil subjected to thermal treatments, to which either tertiary butyl hydroquinone (TBHQ) – a synthetic antioxidant, either loquat (*Eriobotrya japonica*) extract was added, having a control sample soybean oil without any antioxidant additives, shows that natural extract has a stronger antioxidant activity in soybean oil than chemical synthesis antioxidant [35].

Pedro *et al.* [36] investigated the ability of goji fruit extracts to protect soybean oil against oxidation as an alternative to synthetic antioxidants. The extraction parameters have been optimized. Thus, various solvents were used in the extraction process, the temperature ranged from 25 to 45°C, the extraction time has been in the range 60-180 minutes and the solid: solvent ratio has been between 1:10-1:30 w/v. The most efficient method of phenolic compounds extraction was by using an ethanol/water solution (70/30, v/v) at 45°C with extraction time of 162 min and a 1:30 solid:solvent ratio. The protection factor value of soybean oil with organic goji fruit extract was significantly higher than that of soybean oil with butylated hydroxyanisole (BHA) and butylated hydroxytoluene (BHT). The results suggested that goji extract can be recommended as a substitute of synthetic antioxidants in oil stabilization against oxidative damage, and organic goji fruit has a richer phenolic content than conventional goji berries [36].

Several research have been carried out by Oancea *et al.* [37-39] in order to enhance the oxidative stability of rapeseed oil, cod liver oil and sunflower oil by adding natural compounds with antioxidant potential.

Romojaro *et al.* [40] studied the effect of some natural compounds on sunflower oil, olive oil and soybean oil during frying. By assessing the effect of supplementing edible vegetable oils with natural extracts with antioxidant potential from by-products resulting from wild fruits processing, on lipid oxidation during thermal treatments or during storage, there was an increase in oxidative stability of oils. Peroxide value and malondialdehyde content (a biomarker that measures the cell membrane oxidative damage) significantly decrease compared with values recorded in control sample, without any antioxidant agent addition, or compared with oil sample with addition of synthetic antioxidants.

The above mentioned data highlight the potential of raw extracts obtained from berries as lipid peroxidation inhibitors in edible oils [37-40].

3. Conclusions

By-products resulting from industrial processing of berries such as bilberries (*Vaccinium myrtillus L.*) or blackberries (*Rubus fruticosus L.*) are rich sources of phenolic antioxidants. These compounds can be easily recovered up to 95% from the initial phenols content using non-invasive procedures for environment by applying solvent extraction. The berries content in natural compounds with antioxidant activity varies according to sort and variety, plant nutrition, place of origin, harvest time and stage of maturation at the time of harvesting, environmental factors, etc. Processing and storage conditions can also influence phenols content. In food industry, the replacement of synthetic antioxidants with natural antioxidants can be applied in different dairy products as well as in meat and meat products, cereal products, but the most current requirement is the replacement of synthetic antioxidants with natural ones in vegetable oils in order to increase their resistance to lipid oxidation during frying. The measurements applied on enriched oils with natural antioxidant, have shown that oxidation prevention time is higher in these oils than in oils in which synthetic antioxidants were added.

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Compliance with Ethics Requirements. Authors declare that they respect the journal's ethics requirements. Authors declare that they have no conflict of interest and all procedures involving human / or animal subjects (if exist) respect the specific regulation and standards.

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