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Review

Phenolic compounds as stabilizers of oils and antioxidative mechanisms under frying conditions: A comprehensive review

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ABSTRACT

Background: A series of chemical reactions lead to the oil degradation under frying conditions, and a number of degraded compounds compromise the safety, nutrition, health and shelf-life of frying oils and fried food.
Scope and approach: Nevertheless, the oxidative deterioration of frying oils was little protected by numerous endogenous antioxidants, including carotenoids and tocopherols, as well as synthetic antioxidants. Phenolic compounds with strong biological activities are nature antioxidants widely distributed in the plant kingdom. Under frying conditions, phenolic compounds offer good protection of oils against oxidative deterioration without decreasing sensory properties of fried food.
Key findings and conclusions: Though the antioxidant mechanisms of phenolic compounds have been reported in literature, the specific syntheses and performance evaluation of their antioxidant mechanisms under frying conditions is scarce. The purpose of this review is to provide a comprehensive understanding of the effect of phenolic compounds on the stabilization of oils and their working mechanisms to against oxidation under frying conditions, and their limitations and challenges are also included. This review will provide the potential insights on the possible application of phenolic antioxidants as the stabilizers of frying oils in food industry.

1. Introduction

Frying is one of the oldest and most prevalent food cooking methods, and has been widely used in food industry, fast food restaurant and household kitchen. Frying procedure can produce desirable and palatable foods with a crispy texture, an attractive flavor and golden color. Due to these unique organoleptic properties, deep-fat fried products are widely consumed across the world, despite the current guidelines and recommendations regarding the lower level of dietary fat. Furthermore, the sale of deep-fat fried products, such as ready-to-eat and pre-cooked products has a dramatical increase in the western countries, and is steadily and rapidly raising throughout the developing world, which indicates that fried foods have been becoming the industry chain in food market (Aladedunye, Przybylski, & Matthaus, 2017).

Three frying methods, including pan frying, shallow frying and deep-fat frying, have been commonly used, and the main difference of three frying methods is the amount of oil. For pan frying, little oil is used to lubricate the pan, while shallow frying means a small amount of pre-heated oil on a flat surface or in a shallow pan to fry food. Compared with these two methods, deep-fat frying means submerging food in enough hot oil, and more studies have focused on the deep-fat frying (Andrikopoulos, Kalogeropoulos, Falirea, & Barbagianni, 2002; Kalogeropoulos, Mylona, Chiou, Ioannou, & Andrikopoulos, 2007). During deep-fat frying, the oil is repeatedly and continuously used at high temperature and exposed to air. Under these conditions, various complex chemical reactions, including oxidation, hydrolysis and polymerization being the most common, take place to degrade the frying oil. Because of these reactions, frying oil can be converted to a number of toxic thermo-oxidative degradation compounds, including mono- and diacylglycerols, alcohols and polymeric products, which not only alter the shelf life of fried foods, but also negatively impact sensory, functional, nutritional and health quality of frying oil (Chiou & Kalogeropoulos, 2017). Numerous studies reported several diseases, such as neurodegenerative diseases and diabetes, might be induced by aldehydes, although the flavor of fried foods is partly contributed by some aldehyde species (Goicoechea, 2008; Voulgaridou, Anestopoulos, Franco, Panayiotidis, & Pappa, 2011). Consequently, there is a growing interest to enhance the oxidative stability of frying oil.

To stabilize the oil and retard oxidative degradation during frying,

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several synthetic antioxidants, including tert-butyl hydroquinone (TBHQ), butylated hydroxyanisole (BHA) and butylated hydroxytoluene (BHT), are frequently used. Although these antioxidants effectively protect the oils to prolong shelf-life or room temperatures, they have poor or no protection against thermo-oxidative degradation under deep-fat frying conditions, because of their thermal decomposition, steam distillation, and absorption by fried products (Aladedunye et al., 2017). The perceived harmful effects of synthetic antioxidants on health also limit their applications in food industry. In addition, some natural antioxidants in vegetable oils, such as tocopherols and phytosterols, could undergo thermooxidative degradation to generate various nonpolar and polar compounds during deep-fat frying (Aladedunye et al., 2017). Hence, more work has been carried out to develop the new nature antioxidants with better thermal stability and antioxidant activity, especially under deep-fat frying conditions.

The great interest in phenolic compounds has been rapidly grown in recent years, due to the increased evidence of their attractive nutritional properties on human health. Phenolic compounds have high antioxidant activity, and the consumption of food with abundant phenolic compounds might reduce the risk of coronary heart disease, certain types of cardiovascular disease and cancer (Bravo, 1998). In addition, the presence of phenolic compounds so far could effectively inhibit thermo-oxidative degradation of frying oil as well as the formation of toxic thermo-oxidative degradation compounds such as acrylamide and heterocyclic amines (Cheng et al., 2007, 2010). Therefore, understanding the role of phenolic compounds in improving the performance of frying oil during deep-fat frying contributes to the extension of fry-life of the oil and quality maintenance of both frying oil and fried food. However, the information about improving the performance of frying oil with phenolic compounds is relatively scattered, and it is necessary to support the current exploits with a systematic review. This also satisfies the increased demands to control the quality of frying oil and fried food with nature antioxidants.

In this review, the current knowledge focused on the performance of applied phenolic compounds in frying oil and antioxidative mechanisms under frying conditions are summarized. The limitations and challenges of phenolic compounds for frying applications are also included.

2. The performance of phenolic compounds in frying oil

Phenolic compounds can be mainly classified into 6 types: phenolic acids, flavonoids, lignans, lignin, phlobaphenes and tannins, and the antioxidant activities are generally determined by their chemical structure (Vermerris & Nicholson, 2008). Recently, a variety of studies indicated that compared with the efficacy of BHT, BHA and tocopherols, phenolic compounds are much less volatile and have been considered as stabilizers of frying oils over deep-frying process (Aladedunye, 2014; Aladedunye et al., 2017). Furthermore, results also demonstrated that phenolic compounds were able to effectively inhibit the oxidation of tocopherols, and the overall retention of tocopherols in the frying oil was significantly improved (Esposto et al., 2015).

2.1. Endogenous phenolic compounds

A variety of nature antioxidants, also called minor components, present in edible oils, and provide an important protection against oxidative damage under both storage and frying conditions. Phenolic compounds are one of the most significant and frequently researched endogenous antioxidants (Table 1). The higher content of total phenolic compound is presented in virgin olive oils. More than thirty-six individual olive oil phenolic compounds have been identified, and these compounds can be classified based on their chemical structure in the following five groups: phenolic acids, phenolic alcohols, hydroxy-isocromans, flavonoids and lignans.

Recently, various studies indicated phenolic compounds naturally presented in virgin olive oil also have the ability to improve its oxidative stability during frying. The possible mechanism is that the initiation and propagation stages of the oxidative chain reaction can be interrupted by phenolic compounds after reacting with free radicals to produce more stable compounds (Chiou & Kalogeropoulos, 2017; Gã³;Mez-Alonso et al., 2003). In comparison with other vegetable oils which contain higher contents of tocopherols under deep-frying conditions, phenolic compounds in the virgin olive oils have the ability to maintain oils stability. Nevertheless, when the concentrations and antioxidant activity of these phenolic extracts were rapidly decreased, the oils were easier to be oxidized during frying. All these results suggested that the amount of phenolic compounds is one of the significant parameters to predict oxidative stability of frying oils under deep-frying conditions (Gã³;Mez-Alonso et al., 2003).

Table 1
Total phenolic compound (TPC) content in some common vegetable oils.

Oil	TPC content (mg CAE/100 g)	Reference
Soybean	1.48	Siger, ogala-Kalucka & ampart-Szczapa (2008)
Sunflower	1.20	Siger, Nogala-Kalucka, and Lampart-Szczapa (2008)
Rapeseed	1.31	Siger et al. (2008)
Corn	1.26	Siger et al. (2008)
Grapeseed	0.51	Siger et al. (2008)
Hemp	2.45	Siger et al. (2008)
Flax	1.14	Siger et al. (2008)
Rice bran	1.44	Siger et al. (2008)
Pumpkin	2.46	Siger et al. (2008)
Virgin Olive	5-150[a]	Chiou and Kalogeropoulos (2017)
Sesame	711-1730[a]	Wu (2007)

[a] Express as mg/100 g

In addition, the degradation of phenolic compounds in virgin olive oils under frying conditions was also reported by other studies. The changes in α-tocopherol, hydroxytyrosol and tyrosol were analyzed during continuous frying processes of potatoes by Beltrán-Maza, Jiménez-Márquez, García-Mesa, and Frías-Ruiz (1998). Hydroxytyrosol was undetectable after the sixteenth frying processes, while tyrosol was still detectable until the end of the frying operation. Gã³Mez-Alonso, Fregapane, Salvador, and Gordon (2003) also added that almost 50% of hydroxytirosol and its derivatives in virgin olive oils was reduced only after 10 min frying sessions at 180 °C, and the rest of them were nearly completely degraded after the first six frying operation. By contrast, about 80% of tyrosol and its derivatives retained in virgin olive oils after 12 frying operation, and their degradation followed a linear degradation rate during frying. It is well known that hydroxityrosol with the comparatively higher antioxidant capacity can effectively protect lipids from oxidation during frying, whereas tyrosol with the relatively lower antioxidant activity has been therefore more kept during frying. All these results suggest that the degradation of phenolic compounds might be dependent on their antioxidant activity or chemical structure under frying conditions. On the other hand, the effect of domestic frying conditions (160–190 °C) on the contents of α-tocopherol and total phenolic compounds in virgin olive oils was investigated by Pellegrini, Visioli, Buratti and Brighenti (2001). Results showed that as the concentration of phenolic compounds increased, higher levels of α-tocopherol remained in virgin olive oils, indicating that α-tocopherol could be effectively stabilized by polyphenols during frying.

Lignans are nature compounds occurring in flaxseed oil and sesame oil. The flaxseed oil lignans mainly consist of matairesinol and secoisolariciresinol diglycoside, and the frying performance of flaxseed oil lignans was scarcely studied (Kasote, 2013). Whereas, the frying performance of vegetable oils has been significantly improved after adding sesame oil. Chung, Lee, and Choe (2010) added sesame oil to soybean oil to improve oxidative stability of soybean oil during frying, and results showed that thermo-oxidative stability of frying oil was significantly improved after adding sesame oil, probably due to the presence of lignans provided by sesame oil. The major lignans present in sesame oil are sesamin, sesamolin and sesamol, the structures of which have been reviewed by Aladedunye et al. (2017) in detail, and their influence on the thermal oxidation of methyl linoleate was studied by Jinyoung, Yoosung, and Eunok (2008). The results showed that the oxidation of methyl linoleate was significantly reduced by sesamol, sesamin, and sesamolin during heating. The degradation of sesamol was faster than sesamolin or sesamin, which was also confirmed by Hwang, Winkler-Moser, and Liu (2012), and no significant change was observed between sesamolin and sesamin. All these results suggested that sesamol has the higher antioxidant efficiency than the others, and protects oils from oxidation through its intensive loss under frying conditions. The possible reason was that sesamol could be degraded to sesamolin under frying temperature, which is the excellent radical scavenger and evidently enhances the stability of the oil (Yeo, Jeong, Park, & Lee, 2010; Yeo, Park, & Lee, 2011). Additionally, Hwang, Winkler-Moser, Bakota, Berhow and Liu (2013) also claimed that antioxidant activity of sesamol was lower than TBHQ at the same concentration (0.02% (w/w)) during deep-fat frying, but the better performance was observed when its concentration reached to 0.66%. However, thermogravimetric analysis showed that the application of sesamol in frying oil is limited by its poor thermal stability and high volatility, and this issue could be circumvented by adding sesamol during deep-fat frying.

2.2. Exogeneous phenolic compounds

Although the thermo-oxidative degradation of oils can be significantly inhibited by phenolic compounds under frying conditions, the content of endogenous phenolic compounds in common vegetable oils is relatively low (Table 1). The effectiveness of these compounds during frying are limited, largely due to their insufficient concentrations. Therefore, external fortification has been used to improve the oxidative deterioration of oil under frying conditions, and polyphenolic extracts from different sources can effectively inhibit thermo-oxidative degradation of oils (Chiou et al., 2007). The application of exogenous natural phenolic compounds from different potential sources to enhance the frying stability of oils is discussed in the following sections.

2.2.1. Phenolic compounds from spices and herbs

There is a growing interest in spices and herbs, because they have abundant phenolic compounds. Phenolic compounds of spices and herbs are also identified as effective antioxidants (Yanishlieva, Marinova, & Pokorný, 2010). Generally, spices and herbs, inducing oregano, garlic, rosemary, nutmeg, sage and thyme, have been considered as significant sources of phenolic compounds. The profile of phenolic compounds is different between spices and herbs, and a number of these compounds presented in various common spices and herbs have been identified, including caffeoyl derivatives, carnosol, epirosmanol, phenolic diterpenes (carnosic acid), gingerol, shogoal, rosmanol, 4-hydroxycinnamoylmethane, eugenol fruit, chlorogenic-acid fruit and beta-sitosterol plant. Spices and herbs extracts have been widely used to delay or inhibit the lipid oxidation of foods, such as meat and fish (Jiang, Zhang, True, Zhou, & Xiong, 2013; Sancho, de Lima, Costa, Mariutti, & Bragagnolo, 2011). Recently, there has been growing interest in their effectiveness during frying, and selected studies which employed a variety of spices and herbs extracts to enhance the oxidative deterioration of frying oil are presented in Table 2.

2.3. Rosemary

Rosemary is originally from the Mediterranean region, and has been widely used in the food industry. Generally, there is agreement in the available publications on the antioxidant efficacy of rosemary extract in oils during deep-fat frying. According to Chammem et al. (2015), the rosemary extract (0.08%) was added into mixture of soybean and sunflower oils to improve their performance under frying conditions. Compared with original oils, the addition of rosemary extract could decrease the evolution of unsaturated fatty acid composition from 25% to 5.5%. In addition, the sensory characteristics and organoleptic properties of fried potatoes could be well maintained until the 15th frying. All these results suggest that the presence of phenolic compounds in rosemary extract could enhance the oxidative stability of oils under frying conditions. In a similar study, Guo, et al. (2016) compared the antioxidant efficacy of rosemary extract with synthetic antioxidants: BHA and TBHQ in palm oil during frying, and the thermal oxidation of frying oils. Results showed the generation of secondary oxidation compounds was significantly inhabited. They also found that the alternation of primary oxidative products to secondary oxidative products started on the fourth day of frying in the oils with synthetic antioxidants, which was demonstrated by the decrease of peroxide values, whereas the oils with rosemary extract exhibited prolonged primary oxidative stage with higher peroxide values. Therefore, little antioxidant effect was provided by the synthetic antioxidants at the frying temperature, probably due to their poor stability under the extremely high temperature conditions.

Apart from improving the oxidative stability of the frying oil, the accumulation and formation of harmful substances could also be effectively inhibited by the natural spices and herbs extracts. In a recent study, Filip, Hribar, and Vidrih (2011) reported that the formation of *trans* fatty acids in sunflower vegetable oil was significantly inhabited by adding 1.55% rosemary extract during heating at frying temperature of 180 °C for 120 h. According to Urbančič et al. (2014), the acrylamide formation could be significantly reduced by adding rosemary extract. The rosemary extract was added to sunflower oil, and potato chips were fried at 180 ˚C for 10 consecutive days, and the formation of acrylamide was quantified. In comparison with tocopherols, BHA and TBHQ, rosemary extract had the better effectiveness for reduction of acrylamide formation of potato. Similarly, the acrylamide level in fried potatoes was significantly reduced by 39% and 17% after adding the extracts from cinnamon and oregano, and no significant changes were found in textural, sensorial and physicochemical properties of fried potatoes, suggesting that it is possible to inhabit the formation of harmful substances without negatively affecting the physicochemical characteristics and sensorial acceptance of fried products after treating with natural antioxidant extracts (Morales, Jimenez, Garcia, Mendoza, & Beristain, 2014).

2.4. Sage

Sage is commonly used in foods as seasoning and flavoring. As sage and rosemary belong to the Labiatae family, the similar patterns of phenolic compounds have been identified, mainly include carnosic acid and rosmarinic acid. However, the additional individual phenolic compounds have been identified in sage, such as sagerinic acid, salvianolic acid K, luteolin 7-glucoside, luteolin 7-glucuronide, luteolin 3′-glucuronide, 6-hydroxyluteolin 7-glucoside, sagecoumarin, apigenin 6,8-di-C-glucoside. Based on some workers, these compounds might contribute to the different frying performance of oils during deep-fat frying.

A further study of the effect of sage extracts on frying performance of refined, bleached and deodorized palm olein was undertaken by Man and Jaswir (2000), and the extent of oxidation was evaluated by free fatty acid and polymer content. Compared with the oils without sage extracts, the content of free fatty acid and polymer was lower in treated oils during 6-day deep-fat frying, indicating that the deterioration of the oils was significantly retarded by sage extracts. Kalantzakis and Blekas (2006) found an oil dependent activity of sage extracts. The sage extracts at a concentration of 3 g/kg oil were added to virgin olive oil, refined olive oil, sunflower oil and a commercial oil blend suitable for

Table 2
The performance of phenolic compounds from spices and herbs in frying oil under frying condition.

Phenolic compounds sources	Frying oils	Frying conditions	Results	References
Rosemary (*Rosmarinus Officinalis* L.) and sage (*Salvia fruticosa* L.)	Refined, bleached and deodorized palm olein	Frying of potato crisps at 180 °C for 5 h for 6 consecutive days	The rate of oxidation of oils was significantly reduced after adding 4000 μg/g rosemary or sage extracts.	Man and Jaswir (2000)
Rosemary (*Rosmarinus Officinalis* L.)	Rapeseed oil and the mixture of rapeseed oil with palm olein oil	Frying of French fries at 180 °C for a total of 30 frying per day for 5 consecutive days	The decomposition of polyunsaturated triacylglycerol and the formation of polymers, polar substances were significantly inhibited.	Réblová, Kudrnová, Trojáková, and Pokornya (1999)
Rosemary (*Rosmarinus Officinalis* L.)	Palm oil	Frying of French fries at 180 °C for 5 h per day for 5 consecutive days	The stability of palm oil was significantly enhanced after adding extract at 120 μg/g as determined by PV, AnV, IV and FFA.	Guo et al. (2016)
Rosemary (*Rosmarinus Officinalis* L.)	Sunflower oil	Frying of potato chips at 180 °C for two batches for 10 consecutive days	Extract showed a significant effectiveness for inhibition of oil oxidation as determined by TPC, FFA, VTV and CDV.	Urbančič, Kolar, Dimitrijević, Demšar and Vidrih (2014)
Rosemary (*Rosmarinus Officinalis* L.)	Mixture of soybean and sunflower oils	Frying of potato chips at 180 °C for 6 h for 5 consecutive days	The thermo-oxidation was significantly improved by adding 800 mg/g extract as determined by PV.	Chammem et al. (2015)
Rosemary (*Rosmarinus officinalis*) and thyme (*Thymus capitatus*)	Soybean oil	Frying of potato crisps at 180 °C for 6 h for 4 consecutive days	Both extracts showed a more pronounced effect against the prevention of oxidation as determined by TPC.	Saoudi et al. (2016)
Thyme (*Thymus Vulgaris* L.) and Oregano (*Origanum Vulgare* L.)	Soybean oil	Heated at frying temperature (180 °C) for 10 h per day for 3 consecutive days	The formation of polar compounds was significantly inhabited by oregano and thyme extracts at 3000 μg/g as determined by TPC and PV.	Jorge et al. (2015)
Oregano (*Origanum Vulgare* L.)	Refined cottonseed oil	Frying of potato chips at 185 °C for 12 h	The oxidative deterioration of oil was significantly reduced after adding extract at 2000 μg/g as determined by PV, polymerized triacylglycerols and dimeric triacylglycerols.	Houhoula et al. (2004)
Oregano (*Origanum Vulgare* L.)	Soybean oil	Heated at frying temperature (180 °C) for 10 h per day for 2 consecutive days	The oxidation of oil was significantly prevented by extract at 3000 μg/g as determined by TPC.	Pereira Moura Aranha and Jorge (2012)
Greek sage (*Salvia fruticosa*) and summer savory (*Satureja hortensis* L.)	Virgin olive oil, a commercial oil blend suitable for frying, sunflower oil, refined olive oil	Heated at frying temperature (180 °C) for 5 h per day for 2 consecutive days	Extract could significantly reduce thermal oxidation of frying oil, and summer savory extract showed a better effect than the Greek sage extract.	Kalantzakis and Blekas (2006)
Garlic	Sunflower oil	Heated at frying temperature (180 °C) for 3 h for 5 days	The lipid oxidation was effectively inhibited by extract as determined by PV, AV, CDV and CTV.	El-Hamidi and El-Shami (2015)
Nigella sativa L. seed	Sunflower oil and refined, bleached and deodorized palm olein	Frying of potato chips at 180 °C for 3.5 h per day for 5 consecutive days	The oxidative stability of both frying oils has been significantly improved after adding extract as determined by TPC, PV, AV and TV.	Solati and Baharin (2015)
Urtica dioica (Nettle)	Rapeseed oil	Frying of French fries at 180 °C for 8 h per day for 6 consecutive days	Extract at 100 μg/g had a lower inhibitory effect against thermal oxidation as determined by PV, AV, IV and AnV.	Riyazi and Asefi (2015)
Curcuma longa Linn. (*C. longa*)	Refined, bleached and deodorized palm olein	Frying of French fries at 180 °C for 8 h for 5 consecutive days	The oil oxidation and deterioration was significantly retarded after adding extract at 2000 μg/g as determined by PV.	Mohd Nor et al., 2009a,b
Thymus capitatus	Corn oil	Frying of potato chips at 180 °C	The thermal oxidation was effective against by extract incorporation as characterized by PV and FFA	Karoui, Dhifi, Ben Jemia, and Marzouk (2011)
Murraya koenigii leaf	Refined, bleached and deodorized palm olein	Frying of French fries at 180 °C for 8 h for 5 consecutive days	Extract at 2000 μg/g significantly retarded oil oxidation and deterioration as determined by PV, AnV, IV and FFA.	Mohd Nor et al., 2009a,b
Pandanus amaryllifolius leaf	Refined, bleached and deodorized palm olein	Frying of French fries at 180 °C for 8 h for 5 consecutive days	Extract at 2000 μg/g significantly retarded oil oxidation and deterioration as determined by PV, AnV, IV, FFA, polar and polymer compound contents.	Nor et al. (2008)
Inca muña (*Clinopodium bolivianum*) leaf	Soybean oil	Frying of potato chips at 185 °C for 7.5 h	Extract at 600 μg/g had the highest efficacy against oil oxidation as determined by TPC and AnV.	Chirinos, Huamán, Betalleluz-Pallardel, Pedreschi and Campos (2011)
Mashua (*Tropaeolum tuberosum*)	Soybean oil	Frying of potato pieces at 170 °C for 4 h	Extract had the stronger protective effects against oil oxidation as determined by PV, FFA, CDV and CTV.	Betalleluz-Pallardel, ete al. (2012)
Malvaceae (*Althea rosea* L), Chenopodiaceae (*Chenopodium album* L), Asteraceae (*Cichorium intybus* L) and Fumiraceae (*Fumaria indica* L)	Sunflower oil	Heated at frying temperature (180 °C) for 1 h	Only Fumiraceae extract at 400 mg/g was able to extensively inhibit lipid oxidation and improve oxidative stability of oil as determined PV, IV, CDV and CTV.	Raza, Rashid, William, and Razzaq (2014)
Caralluma fimbriata	Sunflower oil	Frying of French fries at 190 °C for 1 h	The oxidative stability was better after treating extract at 200 μg/g.	Babu et al. (2017)

(*continued on next page*)

Table 2 (*continued*)

Phenolic compounds sources	Frying oils	Frying conditions	Results	References
Citrus hystrix peel	Refined, bleached and deodorized palm olein	Frying of fish crackers at 180 °C for 5 h per day for 4 consecutive days	Extract at 2000 μg/g can significantly retard the thermal oxidation reactions as determined by PV, AnV, TV, IV and FFA.	Jamilah, MAN, and CHING (1998)

Abbreviations: FFA = free fatty acids, TPC = total polar content (TPC), PV = peroxide value, AnV = *p*-anisidine value, TV = totox value, IV = iodine value, CV = carbonyl value, CDV = conjugated dienes value, CTV = conjugated trienes value.

frying. Vegetable oils were heated for 5 h per day at frying temperature (180 °C) for two consecutive days, and the thermal stability assessed by the content of total polar materials and *p*-anisidine values. The authors reported that the added ethanol extract obtained from sage only exhibit a significant inhibitory effect on a commercial oil blend suitable for frying; however, in virgin olive oil, refined olive oil and sunflower oil thermal oxidation was not significantly improved. The possible reason might be that the sage extracts offered a concentration dependent protection for different vegetable oils. In addition, they also observed that the ethanol extract obtained from sage had a less pronounced inhibitory effect than the respective acetone extract against thermal oxidation of vegetable oils during deep frying, although the total polyphenol content was relatively higher in the ethanol extract. The differences between these two extracts could be explained by their polyphenolic composition. Rosmarinic acid was presented in both extracts, but flavonoids were only identified in the acetone extract. Therefore, the performance of phenolic compounds in frying oil should consider both the profile and content of phenolic compounds.

In a recent study, Taha, et al. (2014) added different concentrations of sage extracts in the refined rapeseed oil. The oil was heated up to 175°C for frying French fries for 8 h per day for 4 consecutive days, and the formation of oligomer TAGs was quantified. However, the content of oligomer TAGs did not exceed the limitation (12%) until 25 h of frying in the original oil, while the treated oil reached this value after 20 h of frying, suggesting that the formation of oligomer TAGs in oils treated with sage extracts was faster than control oils despite higher phenolic content and oxidative stability. The possible reason might be related to the extraction method, by which higher amounts of volatile monoterpenes and the majority of oligomer precursors, including mono- and di-acylglycerols, free fatty acids and other oxidized compounds, were co-extracted with polyphenols. Hence, it might be necessary to fractionate phenolic extracts with C18 SPE cartridge and Sephadex column before mixing with vegetable oils.

2.5. Oregano

Oreganum vulgare L., commonly known as oregano, is one of the most favorable spices commonly used in foods all over the world. The oregano extracts have the ability to retard lipid oxidation. For instance, the antioxidant effectiveness of oregano extracts in soybean oils under heating conditions was evaluated by Jorge, Veronezi, and Del Ré (2015). After 20 h of heating at a temperature of 180 °C, the value of total polar compounds in soybean oils without oregano extracts was 28%, and reduced to 12.4% after adding oregano extracts, indicating that oregano extracts effectively inhibited the formation of polar compounds in the soybean oil. Compared with TBHQ, they found that the synthetic antioxidant had poorer oxidative protection than oregano extracts according to the amount of total polar contents, which was also confirmed by Pereira Moura Aranha and Jorge (2012).

A number of solvents (e.g., petroleum ether, diethyl ether, and ethanol)have been widely used to extract the antioxidative compounds from oregano, and their antioxidant efficiency on the oxidative stability of cottonseed oil during potato chips frying was evaluated by Houhoula, Oreopoulou, and Tzia (2004). Comparing the antioxidant ability of oregano extracts prepared by different organic solvents (e.g., petroleum ether, dietyl ether, and ethanol), ethanol extracts effectively prevent cottonseed oil against oxidative deterioration under frying condition as measured by the accumulation of polymerized and dimeric triacylglycerols. This difference can be related to the solvents with different polarities. Although the majority of polyphenols could be extracted by organic solvents, some extremely polar polyphenols, such as cinnamic and benzoic acids, could be effectively extracted with higher polar organic solvents (Stalikas, 2007). As ethanol and petroleum ether have the highest and lowest polarities, respectively, more polar polyphenols could be present in ethanol extracts, which could have the better frying performance. As discussed previously by Yanishlieva et al. (2010), the

Table 3
The performance of phenolic compounds from berries, fruits and other vegetable sources in frying oil under frying condition.

Phenolic compounds sources	Frying oils	Frying conditions	Results	References
Eriobotrya japonica (Lindl.) fruit skin	Soybean oil	Frying of potato pieces at 185 °C for 24 h	Extract at 400 µg/g had a better protective effect against thermal oxidation of frying oil as determined by TPC, PV, FFA, CV, CDV and CTV.	Delfanian, Esmaeilzadeh Kenari, and Sahari (2016a)
Pomposia (Syzygium cumini) fruit	Sunflower oil	Frying of French fries at 180 °C for 12 h	Oil with extract at both 800 µg/g and 1200 ug/g was able to increase the stability of oil as determined by AV, PV and TPC.	Ah (2010)
Jujube Fruit (*Ziziphus mauritiana* Lam.)	Soybean oil	Frying of potato pieces at 185 °C for 24 h	Stability of oil was significantly improved by extract at 600 mg/g as determined by FFA, CV, CTV, CDV, PV and TPC.	Delfanian, Esmaeilzadeh Kenari, and Sahari (2016b)
Mango peels and kernels	Frying oil (75% sunflower and 25% soybean)	Frying of potato pieces at 180 °C for 2 h	Bothe extracts showed a more pronounced inhibitory effect against thermal oxidation as determined by PV.	Mostafa (2013)
loquat fruit (*Eriobotrya japonica* lindl.) skin	Soybean oil	Frying of potato pieces at 180 °C for 24 h	Extract at 400 mg/ug exhibited stronger protective effects in stabilization of oil as determined by FFA, PV, CDV and CTV.	Delfanian et al. (2016)
Pomegranate (*Punica granatum* L.) Peel	White Coconut Oil	Frying of potato chips at 180 °C for one frying cycle per day for 3 consecutive days	The oxidation of oil was effectively suppressed after adding extract at 20 mg/g as determined by PV, CDV and CTV.	Bopitiya and Madhujith (2015)
Canadian rowanberry (*Sorbus aucuparia*) and crabapple (*Malus baccata*) berries	Rapeseed oil	Frying of French fries at 180 °C for 8 h per day for 2 days	Both extracts effectively lowed the thermo-oxidative degradation, with the Canadian rowanberry extract being more pronounced as determined by TPC, AnV, IV and PV.	Aladedunye and Matthäus (2014)
Morden Hawthorn (*Crataegus mordenensis* Boom.) and Chokecherry (*Prunus virginiana*)	Sunflower oil	Frying of French fries at 180 °C for 8 h	The stability of oil was significantly improved after adding both extracts as determined by AnV, IV, PV and TPC.	Aladedunye, Kersting, et al. (2014) and Aladedunye, Przybylski, et al. (2014)
Grape seed (*Vitis labrusca* L.)	Soybean oil	Heated at frying temperature (180 °C) for 20 h	The higher levels of essential fatty acids and total tocopherols were retained after adding 100 µg/g extract.	Freitas et al. (2017)
Grape seed (*Vitis labrusca* L.)	Sunflower oil	Heated at temperature (200 °C) for 20 h with convective and microwave ovens	Extracts at 600–800 µg/g could significantly inhibited the lipid oxidation with convective and microwave heating as determined by PV, AnV, CDV and CTV.	Poiana (2012)

Abbreviations: FFA = free fatty acids, TPC = total polar content (TPC), PV = peroxide value, AnV = *p*-anisidine value, TV = totox value, IV = iodine value, CV = carbonyl value, CDV = conjugated dienes value, CTV = conjugated trienes value.

majority of phenolic acids and flavonoids, including caffeic acid, protocatechuic acid, dihydrokaempherol, dihydroquercetin, eriodictyol and apigenin, could be extracted by ethanol, while other solvents mainly extract tocopherols. In this purpose, Aladedunye, et al. (2017) reported that tocopherols are thermally unstable and easily to be evaporated or distilled during frying, while polyphenols could have the better ability to improve the oxidative stability of frying oils.

2.6. Thyme

Thyme, originally from the Mediterranean regions, is one of the aromatic perennial evergreen herb spices with ornamental, medicinal and culinary uses. The antioxidant activity of thyme is mainly attributed to phenolic compounds, including caffeic acid, rosmarinic acid, luteolin, lithospermic, luteolin-7-*O*-β-glucuronide, luteolin 7-*O*-glucoside, apigenin 7-*O*-glucuronide, (Vergara-Salinas, Pérez-Jiménez, Torres, Agosin, & Pérez-Correa, 2012). Literature reports on the antioxidant effect of thyme extracts are rather insufficient, and one of the recent available studies by Jorge et al. (2015) reported that the formation of polar compounds was effectively inhibited by thyme extracts. They found the antioxidant activity of TBHQ was lost after heating 5 h in oil, whereas the thyme extract was still active.

In addition, Saoudi, et al. (2016) found that thyme extracts not only extended the fry-life and inhibited thermo-oxidative degradation of the soybean oil, but also highly preserved tocopherols during the frying. The possible reason is that some typical phenolic structural compounds, such as thymol and *p*-cumene-2,3-diol, could act as free radical scavengers to protect tocopherols against thermal loss under frying conditions. They also found that the sensory attributes, including taste, crispness, color, odor and overall acceptance, of potatoes crisps could be well maintained throughout 15 h of frying.

2.7. Other spices and herbs

Besides the above four types of spices and herbs, extracts from other spices and herbs can also effectivity inhibit, prevent or delay the thermo-oxidative degradation and improve the performance of the frying oils. Studies on the activity of spices and herbs extracts during frying are numerous, in a study of El-Hamidi and El-Shami (2015), which compared the antioxidant effect of garlic extract with BHT in the sunflower oil heated at frying temperature (180 °C) for 3 h per day for 5 days. The stability of sunflower oil was estimated by changes in conjugated trienes, conjugated dienes, acid and peroxide values. They found the oil treated with garlic extract was more stable than the samples with BHT or without additives. In a similar study, Nor, Mohamed, Idris, and Ismail (2008) observed that when both *Pandanus amaryllifolius* leaf extract and BHT were added to refined, bleached and deodorized palm olein that was used for deep frying at 180 °C from 0 to 40 h, the level of oxidative degradation was lower in oils treated with *Pandanus amaryllifolius* leaf extract based on the lower content of polar and polymer compound, free fatty acid changes and anisidine value. The sensory property of different batches of French fries was also evaluated, and result showed that the taste, crispiness, oiliness and overall acceptability were not significant influenced by *Pandanus amaryllifolius* leaf extract throughout the 40-h frying study.

A further study of the effect of phenolic compounds of Inca muña (*Clinopodium bolivianum*) leaves on the oxidative stability of soybean oil during frying was undertaken by Chirinos et al. (2011). Inca muña purified extract, ethyl acetate and aqueous fractions were added to the soybean oil, and the samples fried potato chips at 185 °C for 7.5 h. As determined by the content of polar compounds, conjugated dienes and trienes, and *p*-anisidine values during deep-fat frying process, ethyl acetate fraction provided a better protection against thermo-oxidative degradation of the oil. The authors explained that more phenolic compounds have been extracted by ethyl acetate, and ethyl acetate fraction had higher antioxidant activity than other fractions.

The addition of *Curcuma longa* leaf extract in refined, bleached and deodorized palm olein significantly reduced the polar and polymer compounds contents during frying at 180 °C up to 40 h, and the sensory quality of French fries were acceptable and not significantly different until the 40th hour of frying (Mohd Nor et al., 2009a,b). In a similar study, the extracts from Murraya koenigii leaf also effectively inhabited the thermo-oxidative oxidation of palm olein under deep-fat frying conditions as demonstrated by lower anisidine value, peroxide value, iodine value, oxidative stability index and the content of polar and polymer compounds (Mohd Nor et al., 2009a,b).

2.8. Phenolic compounds from agriculture and processing by-products

Nowadays, a number of agricultural and by-products are considered rich sources of phenolic compounds, which also have been demonstrated as effective antioxidants, due to their high antioxidant activity. For example, avocado and olive leaves are agricultural by-products from annual pruning of avocado and olive trees. A number of phenolic compounds has been identified in both by-products, including phenolic acids (e.g., vanillic, caffeic, *p*-coumaric, chlorogenic, sinapic, syringic and gallic acids) and flavonoids (e.g., oleuropein, luteolin-4-glucoside, apigenin-7-rutinoside, luteolin-7-glucoside, kaempferol 3-*O*-β-glucopyranoside, catechin and epicatechin) (Chiou et al., 2007). The oxidative stability of frying oils could be significantly increased by adding agricultural by-products extracts.

2.9. Agricultural leaves

Generally, extracts from agricultural and processing by-products studied so far showed that they can effectively inhibit thermal oxidation and extent the usage life of frying oils (Table 4). For example, According to the study of Jiménez, García, Bustamante, Barriga, and Robert (2017), the extracts of avocado and olive leaves have been added to the canola oil and high oleic sunflower oil to improve their frying permeance. The main constituents of phenolic compounds in avocado leaf extract were B-type trimer procyanidins, while oleuropein was the major phenolic compound presented in olive leave extract. Results showed that the retention of tocopherols in frying oils was significantly increased by adding both extracts. The possible reason was that polyphenols might reduce the degradation rate of tocopherols. However, compared with avocado leaf extract, olive leave extract could significantly reduce the formation of polar compounds and triacylglycerol polymers in high oleic sunflower oil and canola oil, indicating that the inhibitory effect against thermo-oxidative degradation depended on polyphenol structural features, and oleuropein had a more pronounced effect, which was in agreement with a previous study by Chiou, Kalogeropoulos, Efstathiou, Papoutsi, and Andrikopoulos (2013).

The powerful nature phenolic compounds have been identified in tea leaf extracts, mainly including epicatechin, epicatechin gallate, epigallocatechin and epigallo-catechin gallate, and several studies have investigated the ability of tea leaf extracts to protect oils during frying. According to Naz, Siddiqi, Sheikh, and Sayeed (2005), the addition of crude tea leaf extract to soybean, corn and olive oils resulted in a significant reduction in the iodine value, *p*-anisidine value and peroxide value during deep frying of French fries at 180 °C. In a similar study, the effect of old tea leaf extract on the performance of rapeseed oil during deep-fat frying of potato crisps at 180 °C was reported by Zandi and Gordon (1999). The similar results were also reported by Kmiecik, Gramza-Michałowska, and Korczak (2018).

2.10. Grape seeds

Grape seeds are by-products from grape, and some environmental problems can be aggravated by these by-products. Recently, a number of research studies have indicated that grape seeds are rich sources of bioactive compounds, especially phenolic compounds, mainly including

Table 4
The performance of phenolic compounds from agricultural and processing by-products in frying oil under frying condition.

Phenolic compounds sources	Frying oils	Frying conditions	Results	References
Avocado (*Persea americana cv.* Hass) or olive (*Olea europaea* cv. Arbequina) leaf	High oleic sunflower oil and canola oil	Frying of potato chips at 180 °C for 8 h for 7 consecutive days	The olive extract could significantly increase the thermal stability of oils, while avocado extract only enhanced he thermal stability of high oleic sunflower oil as determined by TPC.	Jiménez et al. (2017)
Olive Waste Cake	Sunflower oil	Heated at frying temperature (180 °C) for 4 h per day for 5 consecutive days	Extract could significantly increase the oxidative stability as determined by PV.	Abd-ElGhany et al. (2010)
Oat	Soybean and cottonseed oils	Frying of Bread cubes at 180 °C for 10 h per day for 10 consecutive days	The amounts of polar compounds with high molecular weight were significantly reduced after adding extract 50 μg/g in soybean oil and 70 μg/g in cottonseed oil.	Tian and White (1994)
Canola oil deodistillates	Refined canola oil	Frying of French fries at 185 °C for 6 h for 5 consecutive days	Extract at 200 μg/g showed a better protection against oil deterioration as determined by AnV.	Aachary et al. (2014)
Olive vegetation water	Refined oil	Heated at frying temperature (180 °C) for 12 h	Extract at 400 μg/g could inhabit the emission of low-molecular-weight aldehydes and oxidation of oil.	Esposto et al. (2015)
Olive leaf, hazelnut leaf and hazelnut green leafy cover	Refined canola oil	Frying of dough at 180 °C for 5 h per day for 7 consecutive days	Compared to Olive leaf and hazelnut leaf extracts, hazelnut green leafy cover extract could extend the usage life of oil as determined by TPC.	Aydeniz and Yilmaz (2012)
Jujube (*Ziziphus mauritiana* Lam.) leaf	Sunflower oil	Frying of potato pieces at 180 °C for 24 h	The stability of sunflower oil was significantly improved after treating extract as determined by TPC, CV, PV, FFA, CDV and CTV.	Delfanian, Esmaeilzadeh Kenari, and Sahari (2015)
Black tea leaves	Corn oil	Frying of French fries at 180 °C for 90 min	The deterioration of oil was significantly decreased by adding extract.	Naz et al. (2005)
Old tea leaves	Rapeseed oil	Frying of potato crisps at 180 °C for 12 successive batches per day for 3 consecutive days	Extract at 2500 μg/g could effectively retarded the deterioration of oil.	Zandi and Gordon (1999)
Canola meal	Canola oil	Frying of French fries at 185 °C for 6 h per day for 5 days	The frying performance was effectively improved after treating canola meal extract as determined by TPC, AnV, IV.	Matthäus, Pudel, Chen, Achary and Thiyam-Holländer (2014)
Olive pomace	Sunflower oil	Heated at frying temperature (180 °C) for 100 min	The degradation of lipidic components was significantly decreased to improve stability of oil by adding extract.	Orozco, Priego-Capote, and Luque de Castro (2011)
Rose hip with seed	Canola oil	Frying of French fries at 185 °C for 16 h	The presence of extract could significantly increase the oxidative stability of oil as determined by PV and TPC.	Aladedunye, Kersting, and Matthäus (2014)

Abbreviations: FFA = free fatty acids, TPC = total polar content (TPC), PV = peroxide value, AnV = *p*-anisidine value, IV = iodine value, CV = carbonyl value, CDV = conjugated dienes value, CTV = conjugated trienes value.

catechin, gallic acid, epicatechin and various proanthocyanidins (Nawaz, Shi, Mittal, & Kakuda, 2006; Shi, Yu, Pohorly, & Kakuda, 2003). Poiana (2012) investigated the effectiveness of grape seed extract on the inhibition of lipid oxidation of sunflower oil at frying temperature (180 °C). The lipid oxidation was significantly retarded by adding grape seed extract based on peroxide value, *p*-anisidine value, conjugated dienes and trienes values. Freitas, Cattelan, Rodrigues, Luzia, and Jorge (2017) added that the formation of decomposition products was remarkably prevented after adding grape seed extract when heating soybean oil at frying temperature (180 °C) for 20 h. The addition of grape seed extracts also resulted in higher retention of total tocopherols as well as essential fatty acids.

2.11. Oil seed meals

Oil seed meals are by-products of the oil crushing industry, such as linseed meal, soybean oil meal and canola meal. They are commonly used to provide vegetable proteins for poultry and pigs feeding. The majority of phenolic compounds are kept in the meal when extracting oils from oil seeds, due to their relatively poor dispersal in oils. A number of studies showed that thermal oxidation of frying oil could be improved by oil seed meals extracts. For example, in the study by Aachary, Chen, Eskin, and Thiyam-Hollander (2015), the formation of primary and secondary oxidation products in canola oil was significantly inhibited after adding canola meal extract during deep-fat frying when comparing with the oil without adding extract. In a similar study, thermo-oxidative degradation in the sunflower oil was significantly decreased in the presence of olive cake extracts (Abd-ElGhany, Ammar, & Hegazy, 2010).

2.12. Phenolic compounds from berries, fruits and their by-products

A variety of berries, fruits and their by-products are also common sources of phenolic compounds. Although the accurate global data on the average dietary intake of berries and fruits are not available, the consumption of them has been closely associated with the lower risk of some developing chronic diseases, including obesity, type 2 diabetes, coronary heart disease, chronic obstructive pulmonary disease and cancer, largely due to the great source of phytochemicals. The common phenolic compounds identified in berries and fruits mainly include ellagitannins, flavonols, anthocyanins, proanthocyanidins, flavan3-ols, hydroxycinnamic and hydroxybenzoic acids derivatives. Recently, a number of studies indicated that phenolic extracts of berries, fruits and their by-products could improve the frying performance of vegetable oils (Table 3).

Studies on the effect of berry extracts on the performance of oils under frying conditions are scarce. It was reported that the addition of chokecherry extracts to sunflower oil resulted in the reduction of hydroperoxide up to 50% (Aladedunye, Kersting, et al., 2014) during deep-fat frying. In a similar study, the effect of rowanberry and crabapples extracts on the performance of rapeseed oil during frying was evaluated by Aladedunye and Matthäus (2014). In comparison with oils with or without BHT, the level of thermo-oxidative degradation of rapeseed oil was significantly reduced in samples fortified with crabapples and rowanberry extracts, with the later extracts being more effective. This might be due to the less volatile of phenolic compounds from berry extracts in response to frying temperature. They also added that the amounts of dimerized and polymerized triacylglycerols were also lower in oils with berry extracts, while no protection has been provided by BHT. It is well known that the operation of BHT mainly related to radical scavenging activity. On the contrary, polyphenols with the anti-polymerization activity might have the ability to hydrolyze the glycosides catalyzed by acids, which improved the frying stability of oils under frying conditions.

In addition, the ability of fruits or their by-products extracts to protect oils under frying conditions has been evaluated by numerous studies. According to Ah (2010), different concentrations of pomposia extracts were added to sunflower oil to prepare French fries for 12 h, and the quality of sunflower oil has been significantly improved when comparing with samples without the extract. In a recent study, Delfanian, Esmaeilzadeh and Sahari (2016a) compared the antioxidant activity of TBHQ and loquat fruit skin extract using soybean oil without antioxidant addition during frying of potato pieces at 185 °C for 24 h. Samples treated with loquat fruit skin extract were more stable than oils with TBHQ or without additives. Delfanian, Esmaeilzadeh and Sahari (2016b) also added jujube extract to soybean oil during potato chips frying at 185 °C for 24 h, and the stabilization efficiency of soybean oil with jujube extract was higher than that of the oil with three common synthetic antioxidants. Besides, extracts from a number of related fruits or their by-products, including hawthorn, lemon, loquat fruit skin, pomegranate peel, mango peels and kernels, have been shown to improve the thermo-oxidative stability of oils under frying conditions (Aladedunye, Kersting, et al., 2014; Bopitiya & Madhujith, 2015; Delfanian, Kenari, & Sahari, 2016; Kmiecik et al., 2018; Mostafa, 2013).

Furthermore, the effect of nature phenolic compounds extracted from fruits on the formation of acrylamide during frying has been evaluated by Cheng et al. (2010). Six fruit extracts were added to peanut oil, and frying was carried out at 170 °C. The authors reported that acrylamide formation could only be significantly inhibited by apple extract, whereas dragon fruit extracts demonstrated the potent increase acrylamide formation. For longan, mangosteen and blueberry extracts, no significant influence on the formation of acrylamide. The possible reason was that only apple extracts were rich in proanthocyanidins, including monomers, dimers, trimers, and tetramers, and the reactive carbonyls could be effectively scavenged by certain proanthocyanidins, which might contribute to the inhibition of acrylamide formation (Granvogl & Schieberle, 2006).

3. The possible antioxidant mechanisms of phenolic compounds under frying conditions

The chemistry of lipid oxidation is much more complicated during deep-fat frying, mainly due to the involvement of oxidative and thermal reactions simultaneously. Although the solubility of oxygen is reduced at the high temperatures, oxidation reactions between oil and oxygen are accelerated. It has been suggested that the thermal oxidation mechanism is the same as the chemical mechanism of autoxidation, which includes the initiation, propagation, and termination (Choe & Min, 2007) (Fig. 1). For example, the formed radicals have been identified and quantified with electron spin resonance spectroscopy technique during deep-fat frying by Liu, Wang, Cao, and Liu (2018a). The authors observed that a number of free radicals were formed during the prolongation of frying time. However, the lipid oxidation under frying

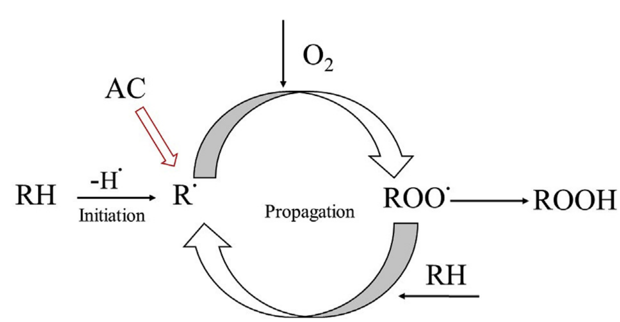

Fig. 1. Mechanism of lipid oxidation.

conditions is different from that formed under heating condition without food or that taking place at room temperature, because various chemical changes happened in deep-frying system, including, dehydration of food, denaturation of protein and gelatinization of starch (Parkash Kochhar & Gertz, 2004). Although numerous studies have proved that the frying performance of oils can be improved by phenolic compounds, the detailed and specific scientific information on the antioxidant mechanism of phenolic compounds under frying conditions is rather scarce. Three possible mechanisms have been reviewed in the following section.

3.1. Scavenging free radicals

The movement of free radical plays a key role to accelerate lipid oxidation, and the predominant mechanism of antioxidative compounds includes scavenging free radicals (Fig. 1). Under frying conditions, the alkyl radical reacts with triplet state oxygen to produce a peroxyl radical, which has high energy to promote the abstraction of hydrogen from another unsaturated fatty acid. Therefore, the addition of hydrogen on the peroxyl radical produces hydroperoxide. Numerous free radicals are continually formed at the propagation step, and the oxidation will be terminated as soon as non-radical products are formed. Thus, the effective antioxidant should have the ability to sacrificially provide the hydrogen radicals to protect labile hydrogen from lipid molecule which is abstracted at the initiation step, or scavenge radicals produced at the propagation (Aladedunye, 2015).

Phenolic compounds have been considered as the stronger free radical scavengers. Free radicals could be terminated by phenolic antioxidants, which can react with the oxidation of lipids or other molecules being rapidly donated the hydrogen atom (s) to radicals. The formed phenoxy radical intermediates are much stabler, and it is not easy to initiate the new chain reaction. The phenoxy radical intermediates can also react with other free radicals to terminate the propagation route (Bravo, 1998). In addition, their antioxidant activity depended on the structure and temperature. For example, Elhamirad and Zamanipoor (2012) observed that the antioxidant activity of gallic and caffeic acids were significantly higher than catechin and quercetin at 120 °C, whereas the most effective was quercetin, followed by catechin and gallic acid, which were markedly higher than caffeic acid. Therefore, the effectiveness of phenolic compounds could be explained by the radical-mediated mechanisms.

Although some common chemical methods, including DPPH, ORAC, ABTS and FRAP radical scavenging assays, have been used to evaluate their antioxidant activity under frying conditions, the results have a poor correlation with the performance in the real frying system. Besides, the Oxidation Stability Index or the Rancimate test, which have been used to evaluate the antioxidant activity of oils under heating or room temperature, are not suitable to measure the behaviors of oils under frying condition (Parkash & Gertz, 2004). For example, the oxidative stability of sunflower and olive oils after deep-fat frying was determined by electron paramagnetic resonance (EPR) (Castejón, Herrera, Heras, Cambero, & Mateos-Aparicio, 2017). The oxidative stability of fried oils could be measured by the remaining stable galvinoxyl radicals, and the aldehyde formation determined by nuclear magnetic resonance was highly correlated with oxidative stability determined by EPR. Quiles, Ramírez-Tortosa, Gomez, Huertas, and Mataix (2002) determined the role of lipid profile, polyphenol and vitamin E content in the antioxidant capacity, with EPR, of sunflower, virgin olive and olive oils after deep-fat frying, results showed that the higher antioxidant activity was in virgin olive and sunflower oils through measuring free radicals with EPR. All results suggested that EPR can be a sensitive and rapid assay to determine the antioxidant capacity of frying oils. The free radical changes of lipid oxidation under deep-fat frying could be detected by EPR, and the mechanisms of some formed oxidative compounds have been proposed according to free radicals (Liu et al., 2018a; 2018b). In order to better understand the radical-mediated mechanism of phenolic compounds improving the performance of oils at frying temperature, EPR can be used in the further studies.

3.2. Carbonyl scavengers

The active carbonyls are commonly formed through lipid oxidation during frying, and potentially damage the quality and safety of food products. All these reactions are induced by a number of complex interrelated changes in the presence of oxygen and initiators, and reaction rates are much higher under frying temperatures (Martínez-Yusta, Goicoechea, & Guillén, 2014). As a consequence of thermal degradation of oils under frying conditions, the formed compounds presented in frying oils have been studied for a long time. Some of oxidative compounds, such as 2-propenal, (*E*)-2-butenal and (*E*)-2-pentenal, are toxicologically relevant, and have been found in both fried foods and frying oils (Zamora, Aguilar, Granvogl, & Hidalgo, 2016).

Phenolic compounds are considered as carbonyl scavengers to trap toxicologically relevant aldehydes formed during the frying process (Zamora et al., 2016). Three main aldehyde-phenol adducts have been formed and identified in fried onions. The protective role of phenolic compounds is related to scavenging carbonyl compounds formed in the lipid oxidation pathway, except for the chelating or scavenging of free radicals. In similar studies, the ability of phenolic compounds to trap lipid oxidation products, including 4-oxo-2-alkenals, 2,4-alkadienals, 4,5-epoxy-2-alkenals, alkanals and 2-alkenals was determined (Hidalgo, Aguilar, & Zamora, 2018; Hidalgo & Zamora, 2018). A number of carbonyl − phenol adducts have been isolated and characterized, and all results suggested that phenolic compounds could act as carbonyl scavengers to prevent the lipid oxidation. Hidalgo, Aguilar, and Zamora (2017) also added that the formed carbonyl − phenol adducts was influenced by the structure of phenolic compounds. They observed that phenolic compounds with groups increasing the nucleophilicity of phenolic carbons have the higher ability to scavenge carbonyls.

3.3. Non-radical reaction

Only a small amount of dimerized triacylglycerols are formed from carbon to oxygen linkages, and most of dimers and oligomers of acylglycerols are formed by carbon-carbon linkage. It is suggested that compounds linked by C-O-C bonds are mainly produced by oxygen during low temperatures, while compounds linked by C-C bonds are mainly formed at high temperatures. As the availability of oxygen is limited under the actual frying system, non-radical mechanism for the formation of dimerized, polymerized and cyclic triacylglycerols has been proposed and might be more significant than the radical scavenging mechanism under the prevailing conditions (Fig. 2). According to the non-radical mechanism, the less activation energy is required to form dimerized, polymerized or cyclic triacylglycerols for the acid catalysts when comparing with the free-radical mechanism. For example, the formation of dimerized triacylglycerols occurs at 220 °C, while this reaction takes place at temperatures of lower than 140 °C undergo acid catalyzation (Parkash & Gertz, 2004).

Consequently, with the non-radical mechanism, the protective action of phenolic compounds could be explained by the proposed acid-catalyzed polymerization and dimerization. For example, despite sesamolin with lower radical scavenging activity, it can undergo acid catalyzed reactions to increase the frying performance of oils under frying conditions (Gertz, 2004). Although some antioxidant behaviors of phenolic compounds could be convincingly explained by the non-radical mechanism, further studies are still needed to deeply discuss the antioxidant mechanism of phenolic compounds under frying conditions.

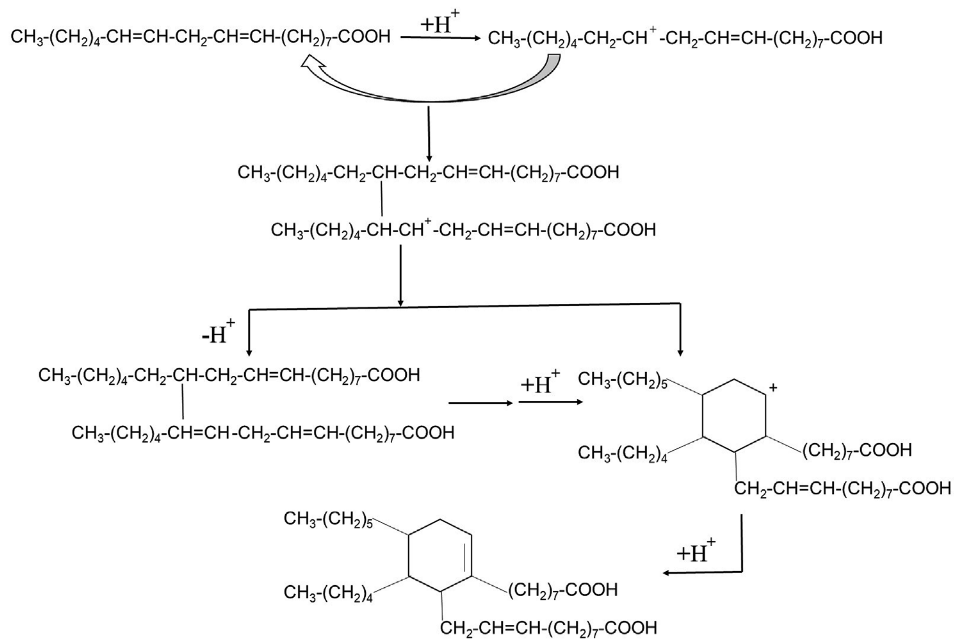

Fig. 2. The proposed mechanistic pathway of non-radical acid cyclisation and dimerization of fatty acids (Parkash Kochhar and Gertz (2004)).

4. Limitations and challenges

Although the performance of frying oils can be improved by phenolic compounds, the majority of these compounds with hydroxy groups exhibit poor solubility in oils. It is observed that the efficiency of phenolic compounds increases with temperatures, probably due to their higher solubility or dispersal under high temperature conditions. Therefore, the applications of these compounds have been generally limited under hydrophobic conditions, and it is significant to increase their solubility and dispersal in the substrate. Lipophilization of phenolic compounds has been studied in recent years, and chemical or enzymatic acylation is an effective method to improve their lipophilization (Aladedunye et al., 2017). Chemical synthesis belongs to a traditional assay, and some of polyphenols, including sinapic, caffeic and ferulic acids, have been lipophilised to enhance their antioxidant capacity and lipophilic solubility (Liu, Jin, & Zhang, 2014). Although some partial needs could be met to a certain extent with chemical synthesis of lipophilic polyphenols, chemical synthesis is commonly reacted under the drastic condition and accompanied by various purification steps to eliminate catalyst residues or by-products. Whereas, there are numerous advantages offered by enzyme-catalyzed acylation, including minimization of by-products formation and side reactions, milder reaction conditions, fewer purification steps, a selective specificity as well as the more environmentally safer and friendly process. Even though the majority reactions of chemical reagents could be cheaper than enzymes, enzyme synthesis is a well-mastered method to selectively modify lipophilic polyphenols, to which the high degree of conversion can be achieved under the optimal reaction conditions (Liu et al., 2014).

Lipophilic phenolic compounds have been reported to reduce the thermo-oxidation of oils, under frying conditions. Aladedunye et al. (2015) evaluated the ability of the lipophilized phenolic extracts to inhibit thermos-oxidation of rapeseed oil during frying, and the level of polar components and polymerization of triacylglycerol was reduced up to 48%, suggesting that thermo-oxidative degradation could be better prevented by adding the lipophilized phenolic extracts under frying conditions. According to the non-radical mechanism, the protection of lipophilized phenolic extracts against polymerization is related to the acid-catalyzed hydrolysis their ester linkage of phloridzyl octadecenoate, and the phloridzin was released as a secondary antioxidant to prolong the overall antipolymerization or antioxidant activity of the lipophilized phenolic extracts after the acid-catalyzed hydrolytic reaction (Aladedunye & Matthäus, 2016). Since thousands of phenolic compounds have been identified from plants, only a few studies are available on the performance of lipophilicity of phenolic compounds in frying oils. Hence, investigations into applications of lipophilized phenolic compounds to improve the performance of oils under frying conditions are warranted.

5. Conclusion

Frying is a relatively common method to prepare foods, but the chemical reactions taking place during frying are rather complicated, leading to decrease the nutritional quality of the both frying oils and fried food. Although the common endogenous antioxidants, such as tocochromanols, could reduce the oxidation of oils under storage, their antioxidant activity was less effective under frying. Phenolic compounds have the ability to inhibit the oxidation of oils under frying conditions. Compared with other common endogenous antioxidants, the amounts of phenolic compounds are limited. Frying oils have been fortified with the exogeneous phenolic extracts from spices and herbs, agriculture, berries, fruits and their by-products, to improve their oxidation without the influence on sensory qualities of fried food. Three possible antioxidant mechanisms of phenolic compounds are also proposed, including: scavenging free radicals, carbonyl scavengers and non-radical reaction, under frying conditions. However, their applications are limited in oils, due to their poor solubility and dispersal. Therefore, the evaluation of lipophilized phenolic compounds on the performance of frying oils will be necessary in future.

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