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A review on the storage stability of edible vegetable oils

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Abstract
Now a day's consumers are more concern toward storage stability of edible oils and fats. Edible oils are more susceptible to oxidation due to the multiple unsaturated bonds in the predominant unsaturated fatty acids such as oleic acid, linoleic acid and linolenic acid. Oxidation of oil leads to unpleasant flavours rancid odours and discoloration. Synthetic antioxidants such as Butylated Hydroxy Toluene (BHT), Butylated Hydroxy Anisole (BHA), Tertiary Butylated Hydroxy Quinone (TBHQ) and Propyl Gallate (PG) have been used against oil rancidity but they have many health issue. Recently attention has been directed towards bioactive plant extracts which offer a unique range of applications for holistic health and wellness and also safer than synthetic ones. Therefore, objective of present review is to provide a comprehensive summary on use of various natural antioxidants to increase the stability of edible oils and the methods available for the prediction of oxidation stability. The effect of antioxidants in preventing the oxidation of oils is also discussed.

Keywords: Edible oils, linolenic acid, TBHQ, Bioactive plant extracts

Introduction
Oils and fats have wide spectrum of application. They are consumed by human beings in their foods since prehistoric times. Edible vegetable oils and fats are derived from plants. The main components of edible fats and oils are triglycerides. The minor components include mono and di-glycerides, free fatty acids, phosphatides, sterols, fat-soluble vitamins, phenollic compounds, chlorophyll, carotenoids, tocopherols, pigments, waxes, and fatty alcohols (Salunkhe and Kadam, 1998) [26]. Oil which contains high Polyunsaturated Fatty Acids (PUFAs) i.e. linoleic and linolenic acids are used in foodstuffs due to its nutritional qualities, abundance, economy, and desirable functionality (O'Brien, 2004) [22].

Oxidation is the major cause of quality loss in oils. Oil oxidation is an undesirable series of chemical reactions that involve oxygen and deteriorate the oil. Oxidation leads to rancidity with off flavours and smells. Oxidation is a complex series of reactions. When oil oxidises it produces a series of breakdown products in stages, starting with primary oxidation products (peroxides, dienes, free fatty acids), then secondary products (carbonyls, aldehydes, trienes) and finally tertiary products. Besides affecting the nutritional quality of the products, during this process, it may generates potential toxic compounds through the action of free radicals and reactive oxygen species (ROS) that are harmful on human health and is implicated in degenerative diseases such as cancer and early ageing (Krishnaiah *et al.,* 2010) [19].

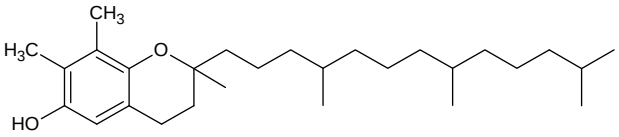
In general, antioxidants are substances present in low concentrations which significantly inhibit oxidation (Aluyor and Ori-Jesu, 2008) [2]. The radicals formed from antioxidants do not propagate the lipid oxidative chain reaction but stop the reaction by forming stable products. Synthetic antioxidants such as Butylated Hydroxy Toluene (BHT), Butylated Hydroxy Anisole (BHA), Tertiary Butylated Hydroxy Quinone (TBHQ) and Propyl Gallate (PG) are widly used as food additives to improve storage stability (Gunstone and Norris, 1983) [6]. In recent years, the toxicity of synthetic chemical antioxidants has been criticized in international market. Therefore, increasing attention has been directed towards the potential of plant products to serve as antioxidants for protection against free radicals. Phenolics, flavonoids, tannins, proanthocyanidins, and various plant and herbal extracts have been reported to be radical scavengers that inhibit lipid peroxidation. Some substances possessing antioxidant activitie such as orange peel to stabilise palm oil have been studied by Arawande *et al* 2014 [10].

Natural antioxidants in crude and refined vegetable oils

Kellens, (1997) [16] reported that lecithin is a by product of the refining processof soybean oil. During the stage of degumming (before neutralization), phospholipids must be removed because the lecithin or phospholipids (gums) increase refining losses, form precipitates in storage tanks or pipes and they make more difficult the filtrate processes.

Karabulut, *et al*., (2005) [5] reported that crude cottonseed oil contains about 1000 ppm tocopherols, but up to a third can be lost during processing. The tocopherol content decreases during each stage of processing, with the highest reductions occurring during chemical refining and deodorization. Chemical refining can reduce about 10-20 % of the tocopherols contents of vegetable oils, presumably because of their absorption on soaps formed during alkali treatments.

Kamal-Eldin & Appelqvist (1996) [13] evaluated that vegetable oils have two groups of natural antioxidants such as tocopherols, carotenoid pigments and some sterols. Other antioxidants, derived from lignans, are present only in some of oils. Tocopherols are the most important group of natural antioxidants that is present in crude and refined edible oils



Tochopherol

Swigło *et al*., (2007) [29] determined the tocopherols by reversed-phase HPLC for edible vegetable oils such as refined corn, soybean, sunflower, rapeseed, grapeseed, and peanut oils, and cold-pressed extra-virgin olive, linseed, rapeseed, and sunflower oils. Total measured tocopherol concentrations varied in the range from 121 to 829 mg/kg.

Khan *et al*., (2015) [17] evaluated total tocopherol, individual tocopherols (α-tocopherol, γ-tocopherol and δ-tocopherol), free fatty acid (FFA %) and peroxide value (PV) in different vegetable oils locally available in Bangladesh. And also evaluated their oxidative deterioration at three months interval during one year of storage. It is found that content of α-tocopherol was highest in sunflower oil which is 20.76 mg/100g while the content of γ-tocopherol was highest in soybean oil which is 59.3 mg/100g. Also, soybean oil was the richest in total tocopherol on the other hand banashpati (hydrogenated fat) had the minimum amount of total tocopherol. Furthermore, total tocopherol present in every oil decreased very slightly after one year. But the FFA% and PV were increased during storage. Thus it is confirmed that tocopherols are important natural antioxidants that are present in vegetable oils. Vegetable oils those have more tocopherol content were found to deteriorate less during storage.

Chemical characteristics and fatty acid composition of Vegetable oils

Adelola and Ndudi (2012) [1] studied the extraction and and chemical characteristics of cottons oil by solvent extraction method using hexane as solvent. Physical, chemical and proximate compositions of oil were analysed. The chemical properties determined were saponification value, iodine value, free fatty acids, acid value and peroxide value. Physical properties were specific gravity, refractive index, viscosity, color, odour, taste and pH. The results obtained are saponification value (189mgKOH/g), iodine value (94.7 g/100g), free fatty acids (5.75mgKOH/g), acid value (11.50mgKOH/g) and 1`peroxide value (9.25mEq/kg). The proximate compositions obtained are carbohydrate (57.06%), fat (13.30%), crude fibre (0.5%), ash (1.5%), moisture content (7.21%), and crude protein (15.40%). The yield of oil was 15.05%

Hamed *et al* (2012) [8] determined chemical characteristics and antioxidant capacity of Egyptian and Chinese sunflower seeds. From chemical characteristics of two seeds, it was found that protein, oil, ash, total phenolic content (TPC) and moisture increased more significantly ($P<0.05$) in Egyptian sunflower seeds Egyptian as compare to Chinese sunflower seeds after dehulling. Fatty acid composition showed that Egyptian sunflower oil contained 86% polyunsaturated fatty acids, while Chinese sunflower oil contained 80%. Major phenolic compound present in TPC was chloragenic acid measured by HPLC. Egyptian sunflower seeds had more antioxidant activity (AA %) than Chinese sunflower seeds as revealed by higher radical scavenging activity (RSA %), less degradation rate of β-carotene linoleic acid color and longer induction period. Thus, both Egyptian and Chinese sunflower seed would be a suitable source of protein and had some properties such as dispersibility, water absorption and emulsifying capacity. Egyptian sunflower oil can be also used as effective source of unsaturated fats and natural antioxidant, hence can be replaced with synthetic antioxidants in many foods to avoid health hazards.

Popa *et al*., (2012) [24] investigated the characteristics of linseed oil for edible purpose. 30% (w/w) oil was extracted using petroleum ether as a solvent. The chemical composition, moisture, total oil content, ash, iodine value, saponification value, acid value and peroxide value of obtained linseed oil was studied. The fatty acids composition was analyzed with GC-MS method. Linseed oil was found to contain high levels of linolenic (53.21%), oleic (18.51%), and linoleic (17.25%), while the dominant saturated acids were palmitic (6.58 %) and stearic (4.43%). Total saturated fatty acid was 11.01% and unsaturated fatty was 88.97%. Dominant unsaturated fatty acid was linolenic (18:3), followed by oleic acid (18:1) and linoleic acid (18:2).

Rafalowski *et al*., (2008) [25] determined fatty acid composition, tocopherols and β-carotene in Polish commercial vegetable oils. Fatty acid composition was determined by Gas Chromatography (GC), while tocopherols and β-carotene were determined by HPLC. The rapeseed oil was having highest content of α- linolenic acid, while the α-tocopherol was highest in olive oil and lowest in linseed oil. Result showed that from nutritional point of view rape seed oil is beneficial for health.

Szterk *et al*., (2010) [30] studied the chemical composition and oxidative stability of plant oils such as borage oil, *Camelina sativa* oil, linseed oil, evening primrose oil and pumpkin seed oil. Oxidative stability of oils were measured by chemiluminescence (CL) method in which light emitted during oxidation process conducted at 70°C in presence of Fe^{+2} ions. High content of tocopherol was present in some oils. It was found that rapeseed oil was most stable.

Jokic *et al*., (2013) [12] determined the fatty acid composition of soybean oil by extraction with supercritical carbon dioxide. Soybean oil fractions were obtained by collecting the extract at different time interval during supercritical carbon dioxide extraction. Fatty acid composition of soybean oil obtained from supercritical carbon dioxide compared with soybean oil extracted with n-hexane. The concentrations of linoleic and linolenic fatty acids were highest in fraction then decrease,

while oleic acid followed the opposite order. It is also an eco-friendly method can be used as alternate method.

Kostik *et al*., studied the fatty acid composition of several vegetable oils and fats by gas chromatography. The results showed that higher contents of saturated fatty acids were found in palm oil (76.0%±1.95) and coconut fat (90.5%±2.95) with predominant presence of lauric acid (12:0) and myristic acid (14:0) as compared to saturated fatty acids in linseed oil (9.65%±1.05), sunflower oil (8.8%±0.8) and safflower oil (7.2%±0.73). Sunflower, safflower and linseed oil contained highest amount of oleic acid (18:1), linoleic acid (18:2), and linolenic acid (18:3). Two varieties of canola oil contain high linolenic (44.0%±2.02) and oleic acid (59.5%±1.907). The highest P/S index (polyunsaturated/Saturated index) was found in safflower oil (10.55), while lowest in palm oil (0.016) and coconut fat (0.005). From the result it is investigated that safflower and sunflower oil contains a healthy mixture of saturated and unsaturated fatty acids which is suitable for human consumption.

Extraction of antioxidants from bioactive plant extracts

Choi *et al.,* (2007) [4] determined antioxidant activity of the methanolic extracts from some grains and to investigate relationships between antioxidant activities and antioxidant contents in the extracts. Mungbean extracts contained polyphenols 45 mg gallic acid equivalents per 100 g of grain and total carotenoid content as 102 µg β-carotene per 100g of grain for 6.1% yield. Tocopherol contents were 10.92 mg/100g of grain. Lack of strong correlation (r2 =0.6200, p > 0.05) between polyphenolic content and DPPH radical scavenging 12 activity was found due to colour interference by anthocyanin content lead to underestimation of antioxidant activity.

Schmidt and Pokorny (2005) [27] reported that oilseeds and other source of edible oils contain both less polar antioxidants soluble in the oil phase, and more polar antioxidants, better soluble in the aqueous phase. Oilseeds which are consumed directly as such or after roasting may be added to foods in order to increase their stability against oxidation. Oilseed cakes and extracted meals still contain, after the removal of non polar antioxidants, antioxidants of medium or high polarity, mainly phenolic acids, their esters, flavonoids and their glycosides, or lignin derivatives, having antioxidant activities.

Tavasalkar *et al*., (2012) [31] prepared four natural antioxidants blends with hexane and acetone extract of rosemary, green tea and other additives having synergistic effect were chosen viz. OF001, OF002, OF003 and OF004. HPLC analysis of these extracts showed that OF001contained 4.789% carnosic acid. Similarly, OF002, OF003 and OF004 contained 3.477%, 4.676% and 7.074% carnosic acid respectively. Total phenolic content of OF001, OF002, OF003 and OF004 were found as 89.64, 81.36, 28.29 and 77.68 mg GAE/g respectively. Control sunflower oil found more susceptible to oxidative deterioration as peroxide value of control sunflower found to increase rapidly. For sunflower oil, OF002 (0.1%) was found effective to reduce the peroxide value below threshold level up to 120 days of storage.

Lieu and Dang (2015) [20] found the possibility for preservation of catfish fat by using sesame cake extracts (SCE) as a source of natural antioxidants obtained by extraction of black and white sesame cakes with methanol. It was investigated that the total phenolic content (TPC) in black SCE was 1386.3± 89.6 mgGAE/100g dw (dry weight) and in white SCE 1318.5±68.7 mgGAE/100g dw. The free-radical scavenging capacities of the black and white SCE were significantly lower than that of BHT and ascorbic acid. The IC50 values of the black, white SCE samples, BHT and ascorbic acid were 817, 833, 31 and 7 mg/L, respectively. Inspite of this both the white and black SCE showed a comparable effect to BHT in preservation of catfish fat against oxidation. During the storage of 42 days at 60°C, the catfish fat samples treated with the black and white SCEs at a concentration of 400 ppm had lower free fatty acid values (FFA) and peroxide values (PV).

Player *et al*., (2006) [23] studied the decomposition of *α*-,*γ*-,or *δ*-tocopherol in soybean oil. The experiment was performed for 24 days at 50°C by high performance liquid chromatography (HPLC). The initial contents of *α*-, *γ*-, and *δ*-tocopherol in soybean oil were 53, 750, and 268 ppm, respectively. The degradation rates of *α*-, *γ*-, and *δ*-tocopherol for the first 10 days were 5.6%, 1.2, and 0.5% per day, respectively. The *α*-tocopherol was completely destroyed in 16 day. The destructions of *γ*- and *δ*-tocopherol were 28% and 17% after 24 d. The induction period of soybean oil is 8 day which is determined by headspace oxygen, conjugated diene, and peroxide value. As the degradations of *α*-, *γ*-, and *δ*-tocopherol increased, the headspace oxygen disappearance, conjugated diene formation, and peroxide value of soybean oil increased. The correlation coefficient between the degradation of tocopherols and the oxidation of soybean oil was about 0.95. The degradation of tocopherols in soybean oil during storage was due to the oxidation.

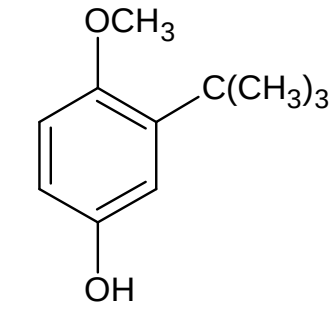
Jia *et al*., (2007) [11] studied the effect of 1.0%, 2.5% and 5.0% purified soybean oil and oxidised soybean oil on the stability of β-carotene. Different sample were stored under light intensity of 1000, 2000 and 4000lx at 20°C for two days and at 50°Cfor 16 days in absence of light. The β-carotene was determined by HPLC. As the storage period increased β-carotene content decreased. In control sample (without soybean oil) β-carotene decreased up to 60% in 12h and completely destroyed in 48h in presence of light. Result showed that purified soybean oil had more protecting effect than oxidised soybean oil. As the light intensity increased, stability of β-carotene was decreased and 100 ppm TBHQ had more significant in β-carotene stability in isooctane at α =0.05. By this study it is found that stability of fruits and vitamins in drinks can be enriched by addition of 1.0% non-oxidised soybean oil.

Wanjari and Waghmare were obtained the extracts with different antioxidant concentrations and activities by using various solvents such as Methanol, Ethanol, Ethanol:Water 1:1, Methanol:Water 1:1, Acetone, Isopropanol and Ethyl acetate from sunflower meal. Their antioxidant activity were tested by the DPPH radical scavenging method and by determination of peroxide value on Soybean oil and Sunflower oil. Ethanol extract exhibited the highest antiradical activity, followed by Methanol extract. While, the Ethyl acetate and Acetone sunflower cake extract showed the lowest antioxidant activity. Moreover, the Ethanol extract appeared to be a stronger antioxidant than BHT. The antioxidant potential decreased according to the following sequence: BHT>BHA>Ethanolic extract> Isopropanol extract >Methanolic extract > Ethanol:Water > Methanol:Water > Ethyl Acetate >Acetone.

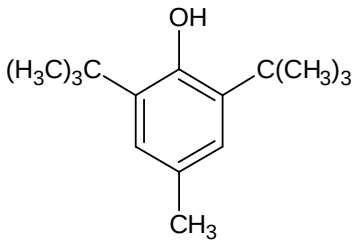
Effect of natural antioxidants on the vegetable oils stability and comparison with synthetic antioxidants

K.P. Suja *et al*., (2004) [28] studied the antioxidant efficacies of methanolic extract of sesame cake in soybean oil, sunflower

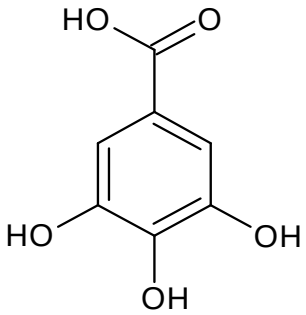
oil, and safflower oils were evaluated at 60°C. Results showed that sesame cake extract (SCE), at concentrations of 5, 10, 50 and 100 ppm in vegetable oils, could significantly ($P<0.05$) lower the peroxide value, diene value and p-anisidine value. The study also indicated that lower concentrations of sesame cake extract were effective in protecting vegetable oils and also has better antioxidant effect than BHT at 200ppm. The results also demonstrated that sesame cake extract could be used as a substitute for synthetic antioxidant to protect vegetable oils, such as soybean, sunflower and safflower oil.



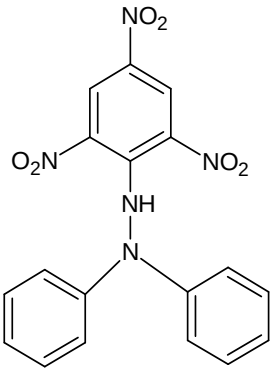
BHA



BHT



Gallic acid



DPPH

Fig. 1: Structures of synthetic antioxidants

Hamed and Abo-Elwafa (2012) [9] determined the oxidation stability of flax seed oil by blending with stable vegetable oils rich in bioactive and antioxidant compounds. FNO and FSO were formulated by blending FO with cold pressed *Nigell*a sativa (NO) and sesame (SO) oils in a ratio of 80:20 (w/w). Fatty acid composition, total phenolic content (TPC), radical scavenging activity (RSA %, DPPH* assay) and oxidation stability of pure oils (FO, NO and SO) as well as oil blends (FNO and FSO) were evaluated. Oxidation stability was evaluated by peroxide value (PV), conjugated dienes (CD), conjugated trienes (CT) and p-anisidine (p-AV) values of pure oils and oil blends. Inhibition of primary or secondary oxidation product formation was also calculated. Results indicated that blending FO with cold pressed *Nigell*a sativa oil had higher efficacy for oxidation retardation. Thus blending can substitute synthetic antioxidants.

Kapoor *et al*., (2009) [14] studied the antioxidant activity of volatile oil and oleoresins (ethanol and ethyl acetate) of black pepper (*Piper nigrum*). Extraction was done by using Clevenger and Soxhlet apparatus. 54 components were present in GC-MS analysis of piper essential oil. Major component present in both ethanol and ethyl acetate was piperine (63.9 and 39.0%), with many other component in lesser amount. Antioxidant activities of essential oil and oleoresins were found in mustard oil by evaluating peroxide value, p-anisidine value and thiobarbituric acid. Both showed strong antioxidant activity than BHA and BHT but less than propyl gallate (PG). Their inhibitory action was also determined by DPPH radical scavenging capacity.

Gutierrez *et al*., (2013) [7] studied the antioxidant activity of ethanolic extracts of onions varieties and their effects on the oxidation of edible corn oil during accelerated storage. The total polyphenolic contents of onions varieties were determined on the filtrated extracts using the Folin-Ciocalteau method. The antioxidant activity was determined by using sorbitan monooleate as emulsifying agent. The peroxide value was evaluated by using the iodometric method. Oxidation induction times were obtained from the peroxide evolution graphs, using the tangent method. The conjugated diene value was determined visible spectrophotometry. The analysis showed an increase in peroxide value and conjugated diene value of oil samples without additives and no change for the oils emulsified with onion extract. Thus, Oil samples emulsified with ethanolic onion extracts showed an extension of the induction period than the samples without additives.

Chung *et al*., (2012) [5] studied the effects of propyl gallate (PG), butylated hydroxyanisole (BHA) and garlic extract on the stability of crude Jatropha oil (CJO) at 25°C and 45°C for 12 months. Parameters measured were acid value, free fatty acid (FFA), rancimat induction period (RIP) and water content. The reading was taken after every three month. Results found that acid values, FFA and water contents of CJO increase during storage while RIP value decreased. It is found that the oxidation stability of CJO show no significant difference between synthetic antioxidants and garlic extract at different temperatures during the long-term storage.

Nyam *et al*., (2013) [21] reported the antioxidant activities of kenaf seed extract (KSE), roselle seed extracts (RSE) and roselle extracts (RE). Their activities were compared with synthetic antioxidant BHA in stabilization of refined, bleached & deodorised (RBD) sunflower oils. Peroxide values (PV), p-anisidine values (AV), TOTOX values, free fatty acids (FFA), iodine values (IV), total phenolic contents (TPC), conjugated dienes (CD) and conjugated triene (CT) were evaluated during 24 days of storage. The results showed

that antioxidant activities of KSE, RSE and RE were better than BHA and RSE is most effective. Thus KSE, RSE and RE may be used as potential source of natural antioxidants in the application of food industry to prevent lipid oxidation.

Anwar *et al*., (2007) [3] carried out assessment of oxidative deterioration of soybean oil at ambient and sunlight storage, over a period of 180 days. The magnitude of oxidative deterioration was measured by peroxide value (PV), p-anisidine value (AV), free fatty acid (FFA), conjugated dienes and trienes, color, refractive index (RI), and iodine value. After 180 days, FFA increased from 0.02 to 1.77and 2.90 % as oleic acid subjected to ambient and sunlight storage respectively, while PV increased from 0.02 to 20.52 and 41.89 meq/kg respectively. The initial value of RI (1.4630), p-anisidine (1.10), conjugated dienes (0.08) and conjugated trienes (0.04) rose to 1.4647, 1.4659; 36.13, 50.40; 23.97, 41.49; 13.81, 19.35 respectively. On the other hand, iodine value decreased from 138.00 to 126.18 and 118.04 respectively. The result demonstrated that magnitude of oxidative deterioration in soybean oil samples exposed to sunlight significantly more pronounced as compared to ambient.

Singh *et al.,* (2004) have reported antioxidant activity of Ajwain essential oil and its acetone extract on linseed oil. Results showed that Ajwain essential oil and its acetone extract at concentration 200 ppm in linseed oil, could significantly ($P<0.05$) lower the peroxide value and TBA value of linseed oil during storage at 80°C. It is found that after certain duration (21 days), BHA and BHT become less effective than acetone extract in stabilizing linseed oil. Thus the study also indicated a better antioxidant effect for acetone extract than BHA and BHT at high temperature. In terms of retardation of oxidation, the effectiveness of samples added to the concentration of 200 ppm can be put into the following order: acetone extract > oil > BHA > BHT > control.

Zhang *et al.* examined the oxidative stability of sunflower oil during storage at 60°C for 21 days on the basis of % free fatty acid (FFA), p-anisidine value (AV), thiobarbituric acid-reactive substances (TBARS) and peroxide value (PV) by the addition of different concentrations of carnosic acid (25%, 60% & 98% w/w) in comparison with synthetic antioxidants i.e. BHA, BHT and TBHQ. They concluded that carnosic acid showed better oxidative stability based on PV and %FFA but gave lower stability than TBHQ. Also, CA98 showed better inhibition to oxidation than others but lower than TBHQ.

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