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An overview of the prevention of oxidative rancidity in butter and butter products using natural antioxidants treatment: a review

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ABSTRACT

Butter and butter products are considered the main part of human diet widely used as table-spread and for cooking purpose due to special taste and aroma. These dairy products contain vital macro- and micro-nutrients including essential fatty acids and vitamins. When these fat rich products exposed to oxygen, heat, light, water and enzymes lead to chemical deterioration and eventually induces undesirable taste and color to them during storage time. The rancidity developments degrade the quality and shelf life of the products, which ultimately reduced consumer acceptability. Moreover, the chemical by-products formed by oxidation reaction trigger chronic illnesses like cancer and heart diseases in consumers. Antioxidants have such molecules, which vigorously react with free radicals formed within the initial phase of autoxidation and remove them from reacting with oxygen. This paper overviewed the function of natural antioxidants as effective and safe method for the oxidation stability of butter and butter products.

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1. Introduction

Butter is known as desirable dairy products with unique flavor in form of water in oil emulsion made by churning fresh or fermented cream or milk. Butter and butter products are widely used as table-spread and for cooking purposes due to high content of essential fatty acids and fat soluble vitamins including A, E, D and K (1–5). Butter constitutes of many volatile compounds, which caused special aroma characteristics. More than 230 volatile compounds have been detected as natural flavor in milk fat (2,6). The aroma profile of butter considerably depends on several main factors including animal diets, milk type,

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season of lactation and butter production, manufacturing process and storage situation (2,3,7,8). Butter contains at least 80% fat, incline to chemical and microbial deterioration when exposed to environmental factors. These factors are oxygen, water, heat, light, enzymes and microorganisms which induced rancid taste and off-flavor to butter during extended storage time (9,10). Rancidity is one of the major corruptions developed in butter and butter products (11,12). The beginning of reactions with unsaturated fatty acid oxygen in double bonds lead to formation of peroxide and hydroperoxide and the fat oxidation occurs in butter (13). Fat oxidation not only have detrimental effect on nutritional value of butter and its sensory properties, but also bring about chronic illnesses like cancer and cardiovascular diseases and early aging in consumers (14–16). The degradation products of hydroperoxides such as aldehydes, alcohols, ketones and hydrocarbons lead to off-flavors. This chemical deterioration also decrease economic value of butter products due to market unacceptability and short shelf life extension (17–19). So the proper solution should be considered for the prevention of lipid oxidation of butter and butter products.

Antioxidant is one the best solutions, which could inhibit lipid oxidation within the long storage period and consequently, avert from butter off-flavor and quality degradation (20,21). Antioxidant defined as any substances which delaying or preventing from rancidity development and other off-flavor in food products in order to oxidation (22). These molecules extremely incline to react with free radicals produced from the initiation phase of autoxidation. So these molecules removed free radicals and inhibit them from reaction with oxygen to produce hydroperoxides (13). These substances could not improve rancid food and do not exacerbate the oxidation reaction (1). Antioxidants divided to two groups involving synthetic (artificial) antioxidants and natural antioxidants. Synthetic antioxidants including propyl gallate (PG), butylatedhydroxytoluene (BHT), butylatedhydroxyanisole (BHA) and tertiary butylhydroquinone (TBHQ), which mostly applied as additives in food industry may have adverse effect on human health (23–25). Whereas natural antioxidants known as antibacterial and anticancer agents could neutralize free radicals in human body and induction desirable aroma to food products without any detrimental effect (26). So consumer's demand is the use of natural food ingredients to food products (27). Various plants, waste plant by-products, herbs, skins, peels, seeds and natural oils are good sources of natural antioxidants such as flavonoids, alkaloids, phenolic compounds, lignins, tannins, quinines and other metabolites. Each of these components has several health benefits. For instance, phenolic compounds showed anti-inflammatory, anti-allergic, cardio-protective, anti-bacterial and anti-oxidant effect (28–30). These natural components added to fresh fat rich food before the beginning of oxidation process to retard lipid oxidation and prolong their shelf life (28,31,32). The efficiency of these antioxidants depends on their concentration, food matrices, temperature, etc. The application and selection of different antioxidants for butter formulation rely on some factors including, safety, antioxidant potential, accessibility, cost and not adverse effect on sensory characterizations (33).

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In this review the application of natural extracts as safe antioxidants were discussed for the prevention of, rancidity taste, stabilization of butter and butter products during storage period and prolong the shelf life of these products without undesirable effect on sensory properties. The natural antioxidants, their concentration, and special conditions in butter and butter products is also reported in Table 1.

2. Natural antioxidant treatment

2.1. Butter

One of the studies developed functional yogurt and butter fortified with microencapsulated α-tocopherol (vitamin E) using canola (α-CAN) or coconut oil (α-COC) as core and ora-pro-nobis mucilage/whey protein as wall material. Over 28 days, physical attributes remained stable, antioxidant activity increased significantly (yogurt: 15.99% to 34.84%; butter: 28.94% to 67.03%), but α-tocopherol degraded similarly in free and microencapsulated forms ($p > 0.05$). Degradation was greater in yogurt due to hydrophilic matrix-wall interaction. Fortification proved more effective in butter, especially with coconut oil core, highlighting potential for vitamin E-enriched dairy products (34). The effect of cinnamon (Cinnamomum verum) extract on butter rancidity as the main indicator of chemical off-flavor was studied. The purpose of this experiment was to ameliorate the butter quality made from cream with cinnamon extraction treatment. Milk cream with 36% fat was churned at 10°C with low moisture amount. The 1, 3 and 5% (w/w) as three levels of cinnamon extracts were added to butter formulation. Results revealed that cinnamon extract could use in butter formulation up to 3% (w/w) without adverse effect on sensory characteristics. Samples treated with cinnamon extract indicated low level of peroxide value, free fatty acids compared to usual butter samples. Cinnamon extracts could impressively scavenge hydroxyl radicals and superoxide anions and eventually prolonged butter shelf life. The 3% of cinnamon extract could applied as a natural preservative for butter and effectively prevent from rancidity (35). In the other investigation, 20 ppm lycopene were added to butter and its sensory properties were analyzed within 4 months of storage. Lycopene-treated samples indicated lower peroxides and free fatty acids compared with control samples. Lycopene as safe antioxidants slowed off-odors, off-flavors and color alteration in treated samples in this storage duration (36). The alcoholic extract of rosemary also had highest antioxidant potential for the prevention of 1,1-Diphenyl-2-picrylhydrazyl (DPPH) radical in butter. In this research, the highest oxidative stability of butter was received with the 400 mg of phenolic compounds per kg of butter (37). Black cumin (*Nigella staiva* L.) seeds and essential oils widely employed as spices and medicinal purposes. The major components detected in black cumin were 5.8%-11.6% carvacrol, 1.0%-8.0% longifoline, 27.8%-57.0% thymoquinone, 7.1%-15.5% *P*-cymene, 2.0%-6.6% 4-terpineol and 0.25%-2.3% *t*-anethole. Butter enriched with 0.05, 0.1 and 0.2 wt-% of this essential oil for the assessment oxidation stability. The antioxidant potential of black cumin was compared to 100 ppm BHT. All samples were stored at 4°C for 90 days. Peroxide values and thiobarbituric acid of treated samples were diminished relying on essential oil concentrations. 0.2% of this essential oil represented vigorous antioxidant activity, which was approximately equal to BHT as artificial antioxidant (38).

The extraction of olive oil generated a large volume of waste by-products, which is a rich origin of natural antioxidants including polyphenols. The olive by products involving olive mill waste water and pomace were incorporated to commercial butter samples in 25°C and 60°C for 3 months of storage. The consequences confirmed that incorporation of 80 mg of olive mill waste water or olive pomace to 1 kg of butter samples indicated lower oxidative deterioration at 25°C and 60°C for 3 months. The achievements showed that the acidity and peroxide value of enriched samples were lower than control samples in this experiment (39). Different parts of walnuts such as leaves, kernel and septum are rich sources of potential antioxidants including, melatonin, tannins ellagic acid and gallic acid (40,41). Herein the traditional butter formulated with 0.05%, 0.1% and 0.5% of walnut kernel septum membranes hydroalcohol extract (WHE) for the evaluation of oxidative stability. These treated samples compared with control sample and samples contained 200 mg tocopherol per kilogram. Traditional butters formulated with 0.5% WHE was found to be more capable for the prevention of peroxides development than samples treated with tocopherol. The antioxidant potential of samples enrich with 0.5% WHE and 0.05% WHE in Schaal and Anisidine test, respectively was higher than tocopherol-treated products. Sensory evaluations also demonstrated that 0.05% WHE was the most suitable concentration for sensory attributes of traditional butter (42). In one of the investigations, the effect of antioxidants from tomato processing by-product (TPB) was evaluated on traditional Tunisian butter (TTB) locally named (*zebda beldi*). This type of butter is straightly provided after milk fermentation followed by churning process (43). Oxidation and lipolysis of TTB trigger rancidity, which decrease quality and butter shelf life. The storage durability of TTB treated with TPB antioxidant was assessed within 60 days of storage at 4°C. TPB contains high level of phenolics and lycopene. Lycopene as liposoluble carotenoid obtained from tomato by products used as strong antioxidants for rancidity prevention without off-flavor induction. Scientific evidences confirmed that lycopene could reduce cancer, cardiovascular and osteoporosis disease (44,45). TTB treated with 400 mg of TPB extract/kg of TTB exhibited lowest peroxide value. This amount of TPB extract did not indicated detrimental effect on lactic bacteria, which are responsible for aroma development in TTB. Control samples and sample enriched with 800 mg of TPB extract/kg of TTB showed higher lipid peroxidation. Consequences disclosed that TPB could postpone lipid peroxidation in TTB and prolong its shelf life up to two months (46).

2.2. Ghee (Clarified butter fat or butter oil)

One of the investigations studied the encapsulation of *Ferulago angulata* essential oil (FEO) into zein nanofibers using electrospinning and evaluated their effectiveness in preventing lipid oxidation in butter oil during accelerated storage. The key components of FEO were α-pinene (35.08%), limonene (21.85%), and γ-terpinene (8.03%). Scanning electron microscopy showed that the resulting nanofibers were cylindrical, smooth-surfaced, continuous, and formed a uniform, randomly oriented network. The consequences showed that nanofiber mats containing 1% and 1.5% FEO (zein + FEO 1% and zein + FEO 1.5%) showed remarkably stronger protection against oxidative deterioration in butter oil over 24 days of accelerated storage compared to butter oil treated with 100 mg/kg butylated hydroxytoluene (BHT) ($P < 0.05$). At the end of the storage duration, the butter oil samples treated with the 1% and 1.5% FEO nanofiber mats demonstrates peroxide value: 0.79–1.03 meq O_2/kg, thiobarbituric acid-reactive substances (TBARS): 0.35–0.45 mg MDA/kg, p-Anisidine value: 1.36–1.66, and acid value: 0.53–0.65 mg KOH/g. These findings highlight that zein/FEO nanofiber, especially at 1% and 1.5% FEO concentrations, outperform BHT as natural antioxidants for extending the oxidative stability of butter oil (47). Another interesting studies assessed the oxidative stability of iron-fortified butter oil (30 mg iron sulfate) supplemented with natural antioxidants, including sesame oil and turmeric powder. Treatments included T0 (control, no antioxidants), T1 (5% sesame oil + 0.10% turmeric), T2 (10% sesame oil + 0.15% turmeric), and T3 (15% sesame oil + 0.20% turmeric). Samples were stored at 40°C for 90 days, with free fatty acids, peroxide value, thiobarbituric acid value, and sensory attributes evaluated after 30 days. Consequences demonstrated that T3 exhibited the lowest free fatty acid and peroxide values, while T_0 and T1 had the highest. Higher antioxidant levels improved appearance, aroma, and overall acceptability, though T3 received slightly lower sensory scores due to turmeric's dark yellow color. Treatments T1 and T3 scored significantly higher than T0 in antioxidant efficacy and sensory quality (T1: 7.76±0.25; T3: 7.99±0.29 vs. T0: 7.50±0.22). The study recommends T3 (15% sesame oil + 0.20% turmeric powder) for effective oxidative stabilization of iron-fortified butter oil with suitable sensory properties for use in fat-based dairy products (48). Curry leaves is medicinal plant and native to India. The antioxidant activity of this plant was analyzed to extent the shelf life of ghee. It was reported that curry leaves had 1913.33±57.35 mg GAE/100g total phenolic compounds and exerted 84.07±0.93% DPPH radical scavenging activity. It was also pointed that curry leaves addition at final phase of heat clarification were more efficacious than at initial phase of this process. It was found that curry leaves could postpone rancidity in ghee. This antioxidant was less effective than BHA as artificial antioxidant. The carbozole alkaloids, which isolated from curry leaves, are koenigine and mahanimbine represented higher antioxidant potential (49). The taro peels extract incorporated to ghee formulation with three various levels of 200, 400 and 600 ppm for the evaluation of its antioxidant activity. 200 ppm of TBHQ as artificial antioxidant was added to ghee for comparison purpose. All of treated and control samples were incubated at 63°C for 21 days in order to accelerating oxidation. Consequences demonstrated that taro peels extract contain high concentration of flavonoids (75.94±9.90mg QE/100g DW) and phenolic compounds (214.58±20.76 mg GAE/100g DW). It was also declared that antioxidant activity of taro peels extract was 91.04±1.55% compared to 94.96±0.67% for TBHQ. Samples containing 600 ppm taro peels extracts exhibited the lowest peroxide value compared to control samples. TBHQ represented higher antioxidant potential than taro peels extract.

However, taro peels extract could be acceptable as a natural antioxidant for ghee shelf life extension (50). The impact of three natural antioxidants such as peanut skins (PS), olive pomace (OP) and pomegranate peels (PP) were studied on ghee oxidation stability.

Therefore, bioactive compounds of abovementioned antioxidants were extracted using ethyl acetate, ethanol (80%) and n-hexane. Observations proved that ethanol extract had better antioxidant properties compared to n-hexan and ethyl acetate extracts. Three different concentrations (200, 400, 600 ppm) of ethanol extracts of PS, PP and OP were added to ghee. 200 ppm of each ethanolic extracts showed proper antioxidant potential within 21 days of ghee storage (51). Asha et al in 2015 studied the antioxidant potential of orange peel extract and BHA in ghee stored at 6, 32 and 60°C as three various storage temperatures within 21 days. In this study the effect of storage condition and antioxidant capability were compared to each other for assessing the oxidative deterioration. Results showed that thiobarbituric acid, peroxide value and free fatty acid of samples remarkably increased whereas radical scavenging activity reduced at 60°C. The effect of storage temperature in the increment of peroxide value and thiobarbituric acid was considerably higher than the effect of storage duration and treatment. Treatment indicated more impact on free fatty acid and radical scavenging activity compared to storage duration and temperature. The capability of orange peel extract for radical scavenging and rancidity reduction was higher than BHA and control samples. Therefore, this survey proved that orange peel extract could effectively control the oxidative degradation of ghee (52).

Dihydroquercetin (DHQ) known as a flavonoid compound, which extracted from Siberian larch (*Larix sibirica*) and have strong antioxidant activity. DHQ showed strong radical scavenging in butter oil and retarded oxidative deterioration (53). Four kinds of essential oils extracted from native plants including, *Mentha longifolia* (*ML*), *Cuminum cyminum* (*CC*), *Salvia officinalis* (*SO*) and *Mentha spicata* (*MS*) were analyzed for their antioxidant capability on ghee at 65°C. Experiment showed that *SO* had 70.06±0.72 mg gallic acid as the highest amount of phenolic components. Among the other essential oils *SO* showed the highest antioxidant potential. Ghee samples treated with 20 ppm of this essential oil had the acceptable sensory attributes (54). The methanolic rice bran extract at 63°C could strongly repress the formation of oxidative by-products in treated ghee samples. The increasing the dose of this antioxidant gives rise to higher inhibition of lipid oxidation in ghee products (55). Mango kernel oil mixed with butter oil at 25 and 55°C for the increasing of oxidation stability. High performance liquid chromatography determined the amount of quercetin 52 mg/100g, chlorogenic acid 837 mg/100g, mangiferin 1257 mg/100g and catechin 436 mg/100g. This natural oil considered a proper alternative for synthetic antioxidants and could prevent the oxidation of C18:1 and C18:2 within ambient and high temperature (56).

One of the studies investigated on clove extracts in the preventing of the oxidative deterioration of ghee products within accelerated and deep fat frying conditions. Direct relations were received among total phenolic compounds, antioxidant activity and free radical quenching for clove extracts. It was also found that BHA exhibited better antioxidant potential at accelerated storage situations while it showed lower antioxidant activity within deep-frying compared to clove extracts (14). According to another survey tomato skin as a rich source of natural antioxidants like lycopene, could augment shelf life of ghee (clarified butterfat or butter oil) (57) without affecting physicochemical and sensory characteristics (58).

Buffalo butter oil formulated with 0.5% coriander extract depicted the lowest peroxide value (0.80 ±0.04 meq.O2 / g fat) and protected the buffalo butter oil form the earliest chemical deterioration (59).

The effect of antioxidant activity of some nonconventional plant sources was assessed on ghee shelf life and oxidative stability. The samples treated with harde (*Terminalia chebula*), nagkesar (*Mesua ferrea*), catechu (*Acacia catechu*), and tamarind (*Tamarindus indica*) seed exhibited considerably lower peroxide values compared to other plant sources as well as the control samples. This group of samples also showed acceptable color and flavor in the sensory evaluation. The DPPH radical scavenging activities of harde, catechu, nagkesar, and tamarind seed were 86.15%, 88.37%, 71.41%, and 89.02%, respectively. These natural antioxidants noticeably restrained oxidative deterioration and prolonged the shelf life of the ghee samples (60). In the other studies, 0.1% of rosemary (*Rosmarinus officinalis Linn*) extract induced remarkable thermal stability to ghee and postponed the beginning of cholesterol oxidation during frying. The shelf life of ghee samples formulated with rosemary extracts at 37 and 60°C was reported 210 and 36 days as compared to 90 and 45 days at 37°C and 12 and 4 days at 60°C in BHA treated samples and control ones (61). The impact of coriander extract was also analyzed for the quenching of oxidative activity of ghee during long time storage and deep frying condition. The observations disclosed that oleoresin as well as steam distilled extract of coriander were more impressive for the control of ghee oxidation whereas were less efficient than BHA. The steam distilled coriander extract demonstrated better antioxidant activity during deep frying compared to its oleoresin and BHA due to higher thermal stable antioxidant concentration in the extract (62). Various concentration of lycopene (30, 60, 90,120 and 150 ppm) as natural additive and 200 ppm BHA were formulated in cow ghee products in order to shelf life enhancement. Four main parameter including peroxide value, free fatty acids, thiobarbituric acid value and color value were measured within 12 months of storage at 30°C for the assessment of antioxidant activity of lycopene. These parameters demonstrated that all concentration of lycopene (30 ppm to 150 ppm) considerably decreased oxidative rancidity. Color is a limiting factor in incorporating higher level of lycopene (more than 150 ppm) in cow ghee products as induced deep red color which is not acceptable (63).

2.3. Whey butter

Previous studies disclosed that dairy products with high level of unsaturated fatty acids are considerably susceptible to rancidity and the oxidation of unsaturated fatty acids reduced the quality and shelf life of whey butter. Therefore, the application of natural antioxidants could convert this industrial waste by-product to functional dairy products. The impact of almond peel extract was studied on oxidative stability of whey butter at supermarket storage temperature. Whey butter was enriched with four various concentrations of almond peel extract (100, 200, 300, 400 ppm) and all samples stored at 6+1°C. Observations demonstrated that the peroxide value of whey butter enriched with 400 ppm almond peel extract after 90 days and control sample were 0.62 and 1.59 (meqO_2/kg), respectively. The 400 ppm of almond peel extract could enhanced the shelf life of whey butter better than other concentrations. The organoleptic properties and refractive index of treated samples did not significantly changed compared to control samples (64).

3. Other strategies for rancidity prevention

3.1. Butter

Sert and Mercan in 2020 estimated the oxidation controlling of butter samples produced with thermosonicated cream. The treated samples produced with three groups of cream which exposed to thermosonication for 5,10 and 15 min. Thiobarbituric acid value markedly reduced in treated samples and the lowest value found in the butter samples which made by cream thermosonicated for 15 min. The free fatty acid content and peroxide values of treated butters were lower than control (raw and pasteurized cream) samples. Thermosonication could repress the oxidative activity of natural and microbial enzymes and ultimately contribute to higher oxidative consistency of treated butters (65). One of the studies emphasized that the application of non-transparent and oxygen permeable packaging could protect butter from light exposure and reduced the rate of oxidation reaction in butter samples (66). The optimization of storage condition such as light, temperature, atmosphere, agitation, shape of storage tanks, metals and material used in storage tanks and packaging could dramatically influence on oxidation consistency of butter (13).

The Pectin/Nanoclay/*Carum copticum* essential oils/β-Carotene (Pec/Clay/CCE/βC) film was investigated on oxidative stability of local butter. The films containing higher level of essential oil and β-Carotene depicted maximum antioxidant potential. The color of this packaging altered from orange to light yellow and this color alteration was considered as a smart color indicator for oxidation monitoring of local butter and showed the expiration time of this product (67).

3.2. Clarified butter fat

The shelf life of clarified butterfat called "smen" produced from pasteurized and non-pasteurized cow's milk was appraised. Observations depicted that peroxide value and Thiobarbituric acid of pasteurized smen were lower than non-pasteurized one. So the oxidation consistency and subsequently shelf life of pasteurized samples were higher than non-pasteurized samples. Heating process in pasteurized smen could eradicate microorganisms and enzymes caused lipid oxidation. So heat treatment could prevent the rancidity originated from oxidation reaction in pasteurized samples (68).

4. Conclusion

Butter and butter products are fat rich food, which are susceptible to chemical deterioration and bring about off-flavor and rancidity in them. Moreover, the consumption of rancid butter brings about many diseases like cancer, heart disease and early aging in consumers. So, optimization of storage condition (Temperature, light, atmosphere, agitation, packaging, shape of storage tank, material and metals applied for storage tanks) and antioxidant treatment could retard oxidation reaction in these dairy products. Synthetic antioxidant involving BHT, BHA, TBHQ and PG indicated detrimental effect in human body. So the utilization of harmless and safe antioxidant is one of the serious challenges in food industry. Addition of natural antioxidants is emerging methods, which could enhance radical scavenging activity and remove them from oxidation reaction. Natural antioxidants originated from plants, herbs and spices, which showed suitable antioxidants activity and have many health benefits for consumers. The application of two or more natural antioxidants was more effective than one and could dramatically inhibit from rancidity. In some cases, natural antioxidant was less effective than artificial antioxidants but they considered acceptable for shelf life extension and delaying lipid oxidation. The use of antioxidants and other strategies for suppressing oxidation reaction and prolong their shelf life should be safe, available and have the least effect on sensory characteristics and do not show any side effect on treated products.

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Fatemeh Barzegar contributed to Project administration; resources; writing–original draft. Mahboubeh Soleimani Farsani contributed to Project administration; resources; writing–original draft. Mohammad Fallahasgari contributed to Conceptualization; data curation; resources validation; writing–original draft; writing–review and editing. Abdorreza Mohammadi contributed to Supervision; investigation; visualization; writing–review and editing.

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