International Food Research Journal 19 (3): 1253-1260 (2012)

Antioxidant and antifungal activity of Coriander *(Coriandrum sativum L.)* essential oil in cake

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Abstract: In recent years, essential oils have been qualified as natural antioxidants and proposed as potential substitutes of synthetic antioxidants in food preservation. The safety of the synthetic antioxidants has been doubted. In this study essential oil of coriander were analyzed by GC/MS and its chemical compositions were identified. Camphor (44.99%), cyclohexanol acetate (cis-2-tert.butyl-) (14.45%), limonene (7.17%), α-pinene (6.37%), were the main components of coriander essential oil (CEO). Then, antioxidant and antifungal activities of CEO were evaluated in cake during 60 day storage at room temperature. The results indicated that, CEO at 0.05, 0.10 and 0.15% inhibited the rate of primary and secondary oxidation products formation in cake and their effects were almost equal to BHA at 0.02 % ($p<0.01$). Antioxidant effects of this essential oil may be due to its trepene and terpenoid components. CEO at 0.15 % could inhibit the growth of fungal in the cake. Organoleptic evaluation of cakes containing CEO at 0.05 % was not different from the control ($p<0.01$). Results showed that this essential oil could be used as natural antioxidant and antifungal in foodstuffs especially those lipid containing.

Key words: Coriander *(Coriandrum sativum L.)*, GC/MS, cake, antioxidant activity, preservative.

Introduction

Essential oil from aromatic and medicinal plants has been known to possess potential as natural agents for food preservation, including antibacterial, antifungal and antioxidant; in fact, many essential oils have been qualified as natural antioxidants and offered as potential substitutes of synthetic antioxidants in specific sections of food preservation where their use is not in contrast with their aroma (Ruberto and Baratta 2000; Politeo *et al*., 2007).

Lipid peroxidation causes oxidative stress, resulting in the development of rancidity, unpleasant tastes and odors as well as changes in color and losses of nutritional value (Iqbal-Bahanger *et al.*, 2008). Antioxidants are used in the food industry to increase the shelf life of the foods. Antioxidants can also prevent the reaction of free radicals with biomolecules in the human body and reduce cell injury and death, chronic and cardiovascular diseases and etc. (Ayoughi *et al*., 2011). Since the beginning of this century, synthetic antioxidants such as butylated hydroxyl anisole (BHA) and butylated hydroxy toluene (BHT) have been used as antioxidants in foods (Reddy *et al.,* 2005). However, the safety of these synthetic antioxidants has been doubted due to toxicity, liver damage and carcinogenicity (Nanditha *et al*., 2008). Therefore a trend about the use of natural additives in foods has been revealed for quite some time as a result of consumer demand and recently the search for natural antioxidants has been widely encouraged (Suja *et al*., 2005).

Natural aromatic plants and spices have been widely used in many food products such as meat and meat products, dairy and bakery products for preserving and for their medicinal value (Reddy *et al*., 2005; Shahsavari *et al*., 2008). Most antioxidant activity investigations have been managed on refined oils, with limited reports on the antioxidant activity of natural herbs/spices in bakery products (Lean and Mohamed, 1999).

Lipid peroxidation in crude soybean oil was related can be controlled by the required amount of Essential Oil of *Bunium persicum* (Shahsavari *et al*., 2008). *Artemisia dracunculus L.* essential oil and *Matricaria chamomilla L.* essential oil had demonstrated effective to reduce the oxidation rate of soybean oil under accelerated conditions at 60 °C (oven test) (Ayoughi *et al.*, 2011). Iqbal Bhanger *et al.* (2008) explored that rice bran extract has a potential source of antioxidants with good antioxidant activity and thermally stable compounds to establish cookies against lipid oxidation. The antioxidant properties of these plant extracts have been mainly attributed

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to their polyphenolic compounds, which are plant secondary metabolites, have many positive effects on human health, including their anti-inflammatory activity and anti-carcinogenic properties. Moreover, the activity of these components as food lipid antioxidants is well known (Iqbal Bhanger *et al*., 2008; Fazel *et al.,* 2008).

Coriander (*Coriandrum sativum L.*) also called as ‘‘cilantro’’ is an annual herbaceous plant originally from the Mediterranean and Middle Eastern regions, cultivated for its culinary, aromatic and medicinal use (Mildner-Szkudlarz *et al*., 2009). This plant is of economic importance since it has been used as a flavoring agent in food products, perfumes, cosmetics and drugs. This culinary and medicinal plant widely distributed and mainly cultivated for the seeds which contain an essential oil (ranges between 0.3% and 1.1%) (Neffati *et al.,* 2011). The essential oil and various extracts from coriander have been shown to possess antibacterial, antidiabetic, anticancerous, antimutagenic, antioxidant and free radical scavenging activities (Sreelatha *et al.,* 2009; Zoubiri and Baaliouamer, 2010). In addition to its culinary value, coriander is known for its wide range of healing properties. It is generally used in gastrointestinal complaints such as anorexia, dyspepsia, flatulence, diarrhea, griping pain and vomiting. Coriander fruit is also reputed as refrigerant, tonic, diuretic and aphrodisiac, while, the oil is considered useful in flatulent colic, rheumatism, neuralgia, etc. Coriander is also used as antiedemic, anti-inflammatory, antiseptic, emmenagogue, antidiabetic, antihypertensive, lipolytic and myorelaxant, and possess nerve-soothing property (Jabeen et al., 2009). The aims of this study are as follows: (i) to determine CEO composition by GC/MS (ii) to study antioxidant activity of CEO in butter cake compared with BHA (iii) to evaluate its antifungal activity in butter cake.

Materials and Methods

Cake ingredients

White flour without any preservatives, salt and baking powder were purchased from Golha industry (Tehran, Iran). Refined, bleached and deodorized soybean oil without any antioxidants was purchased from Parsghoo oil industry (Tehran, Iran). Eggs and sugar were purchased from local supermarket.

Extraction of essential oil

Dried fully ripe fruits (seeds) of *Coriandrum sativum L.* which cultivated in Iran were purchased from Iranian Medical Plant Research Center (Karaj, Iran) and then the essential oil was extracted by steam distillation, using a Clevenger-type apparatus. The obtained essential oil was dried over anhydrous sodium sulphate and kept at 4°C for other experiments (Anonymous, 1988).

GC/MS analysis of essential oil

GC analysis was carried out on an Agilent Technologies 6890 gas chromatograph equipped with flame ionization detector (FID) and a HP-5 capillary column (30 m× 0.25 mm; 0.25 μm film thickness). The oven temperature was held at 50°C for 5 min, and then programmed at 3°C min−1 to 240°C and after that programmed at 15°C min−1 to 300°C (held for 3 min) and finally reached 340°C (at 3 °C min−1). Other operating conditions were: carrier gas, He with a flow rate of 0.8 mL/min; injector and detector temperatures were 290°C and 209°C, respectively; split ratio, 1:10. GC/MS analysis was performed on a GC mentioned above coupled with a Agilent Technologies 5973 Mass system. The other operating conditions were the same conditions as described above, mass spectra were taken at 70 eV. Mass range was from m/z 35–375 amu. Quantitative data were obtained from the electronic integration of the FID peak areas. The components of the essential oil were identified by comparison of their mass spectra and retention indices with those published in the literature and presented in the MS computer library (Davies, 1990; Adams, 1995).

Preparation of cake

Cakes were prepared according to the previously reported method (Lean and Mohamed, 1999). soybean oil (500 g), sugar (450g), salt (2.5 g) were creamed for 12 min at medium speed until light and airy (the CEO and BHA was blended with the oil and the emulsion was mixed with sugar). Whole eggs (450 g) were then slowly added at low speed to avoid curdling, the mixing bowl was scraped, and this was followed by continuous mixing for a few minute. Water (300 g) was added to the mixture and mixed for 1min. Sifted flour (500 g) and baking powder (9 g) were then mildly folded in at low speed for 1min, before the batter was mixed for another minute at medium speed. The batters were baked at 180°C for 20min.

For further studies, cakes with CEO as natural antioxidant at the chosen concentration (0.05, 0.10 and 0.15%), cakes with synthetic antioxidant BHA at concentration of 0.01 and 0.02% (w/w of oil) and cakes without the addition of any antioxidant and preservatives as control sample were prepared. The cakes were cooled, packed in polypropylene film and stored at room temperature (25°C) for 60 days. All experiments were carried out triplicate and results were averaged.

Determination of antioxidant activity

Extraction of lipids- Cakes were ground roughly and extracted in n-hexane by subjected to steam for 30 min. The extracted oil was used for various analyses described in the following paragraphs.

Peroxide value (PV)

Oxidation was periodically assessed by the measurement of peroxide value (PV) at 1st, 5th, 8th, 15th, 30th, 45th and 60th days of storage according to the AOCS method (AOCS, 1989).

Thiobarbituric acid (TBA)

Thiobarbituric acid (TBA) values of the samples were measured during the same days of storage according to the AOCS method (AOCS, 2006).

Determination of antifungal activity

Mold counting

Number of molds in cakes were counting in Dicholoran Glycerol Agar (DG18%) medium during 1st, 10th, 15th, 30th, 45th and 60th days of storage according to the method of ISO (ISO, 2008).

Free fatty acid (FFA)

Free fatty acid contents were determined during 1st, 5th, 8th, 15th, 30th, 45th and 60th days of storage following IUPAC standard method (Iqbal-Bhanger *et al*., 2008).

Moisture

Moisture values of the samples were assessed during the same days of storage by oven at 133°C (Pearson, 1976).

Sensory analysis of cakes

Sensory evaluation of cakes was conducted to determine the acceptability of the product prepared by adding essential oil. Various sensory parameters such as color, texture, taste and total acceptability were analyzed and the mean of the values was taken. Twenty trained panelists were selected for the sensory analysis.

Statistical analysis

Data were analyzed statistically using analysis of variance (ANOVA) and differences among the means were determined for significance at $P \leq 0.01$ using least significant differences (LSD) test (by SAS software). The data are presented as mean ± standard deviation of the three determinations.

Table 1. Chemical composition of CEO (%) determined by GC/MS

	Compound	KI	Amount (%)
1	α-pinene	1940	6.37
2	β-pinene	978	0.90
3	β-myrcene	992	0.32
4	Limonene	1030	7.17
5	1,8-cineole	1032	0.75
6	γ-terpinene	1064	3.53
7	trans-linalool oxide	1075	0.50
8	trans-carvone oxide	1079	1.01
9	Cyclohexanol acetate (cis-2-tert.butyl-)	1114	14.45
10	Camphor	1140	44.99
11	Methyl geranate	1198	1.27
12	Geranial	1264	2.24
13	β-elemene	1388	3.66

Results and Discussion

Chemical compositions of CEO

The results of the CEO analysis by GC/MS are presented in Table 1. Thirty six compounds were identified, representing 95.15% of the total essential oil (note: components which were lower than 1% not shown in the Table 1). Camphor (44.99%), cyclohexanol acetate (cis-2-tert.butyl-) (14.45%), limonene (7.17%), α-pinene (6.37%), β-elemene (3.66%), γ-terpinene (3.56%) and geranial (2.24%) were the main components of CEO.

In analysis of CEO, which has been done in Algeria, 17 constituent were identified through GC, and GC–MS. Linalool (73.11%), p-mentha-1, 4-dien-7-ol (6.51%), a-pinene (3.41%) and neryl acetate (3.22%) were its major components (Zoubiri and Baaliouamer, 2010). The main components of Coriander fruit extracts of Tunisia were Linalool and camphor (Neffati *et al*., 2011). Cantore *et al.* (2004), reported that linalool (64.5%), camphor (6.4%), p-cymene (6.3%) and a-pinene (5.1%) were the main components of essential oil of the Coriander fruits. Samojlik *et al.* (2010) noted that the content of essential oil of coriander fruits was 0.8 (v/w in dry matter). They identified 14 chemical constituents in CEO and its main components were linalool

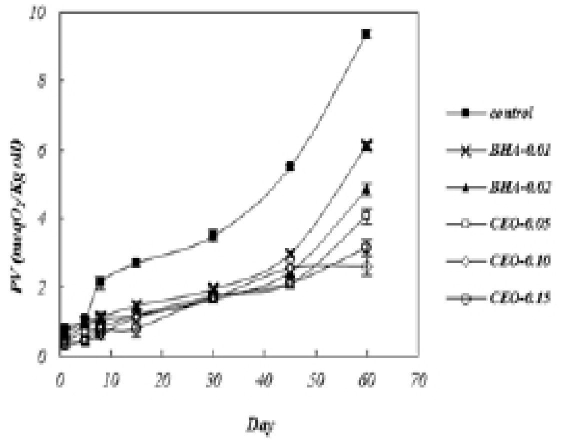

Figure 1. Effect of CEO on PV of samples during 60 days storage at room temperature.

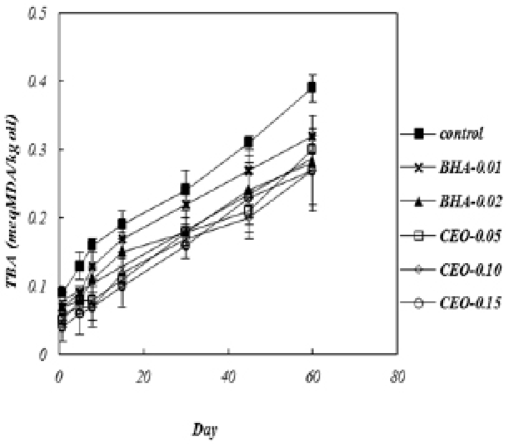

Figure 2. Effect of CEO on TBA of samples during 60 days storage at room temperature.

(74.6%), camphor (5.9%), geranyl acetate (4.6%), and p-cymene (4%). It would also be pointed out that the composition of any essential oil is dependent on the several factors such as, local, climatic, seasonal, different stages of seeds maturity and experimental condition (Shahsavari *et al.,* 2008).

Antioxidant activity of CEO in cake

The addition of natural and synthetic antioxidants to the cake affected the peroxide (PV) and TBA values during 60 days storage (Figures 1 and 2). During the 60 days of storage, PV for control cakes increased with time, indicating that the oil in the cakes had oxidized to lipid hydroperoxides. PV measures primary products of lipid oxidation. These unstable, primary oxidation products were consequently broken down by a free radical mechanism in which the O-O bond was cleaved on either side of the carbon atom bearing the oxygen atom to give the hydroxyl free radical and many types of secondary products such as alcohols, aldehydes, ketones and malonaldehydes which cause off-flavors (Lean and Mohamed, 1999; Rossel, 2005).

As shown in Figure 1, after 60 days of storage the peroxide value was increased in all samples but in the case of control sample there was a sudden increase in PV. Addition of antioxidant reduced the oxidation of oil, so that the highest peroxide value was concerned to control sample and CEO at concentration of 0.05% had lower PV value than control and BHA at both 0.01 and 0.02% concentrations. CEO at 0.10% had the least PV, its PV was lower than BHA at 0.02% which indicated the higher antioxidant activity of CEO than the AOA of BHA at 0.02%. PV increased by increasing the concentration of CEO from 0.10% to 0.15%, appeared to be practically pro-oxidative, which could be due to increasing of phenolic components or contaminants. PV of CEO at concentration of 0.15% was also lower than PV of BHA at 0.02% ($P<0.01$).

TBA index measures the formation of secondary oxidation products, mainly malonaldehyde, which may contribute off- flavors to oxidize oil (Rossel, 2005). Also, TBA of control sample increased with storage time owing to the simultaneous increase in PV. After 60 days of storage, TBA of CEO at 0.05% was no much different from the BHA 0.01 and 0.02%. CEO at 0.10 and 0.15% also had TBA value almost equal to BHA at 0.02%. It indicates that CEO at 0.05, 0.10 and 0.15% inhibited the formation rate of primary and secondary oxidation products in cake and their effects were almost equal to BHA at 0.02% ($p<0.01$).

Our results confirmed earlier reports about bakery products. Addition of purified ether extracts of marjoram, spearmint, peppermint and basil in soda cracker biscuit at 0.5% was reported to have an excellent antioxidant effect compared with BHA (Bassiouny and Hassanien, 1989). The effect of six Malaysian plant extracts, namely turmeric, lemon-grass, clove, betel leaves, garcinia atriviridis and black pepper leaves, as antioxidants was evaluated on the cake by measuring PV and TBA of cakes. After four weeks, turmeric, betel leaves, clove and lemongrass at 1 g kg^{-1} fat concentration were often more effective in preventing oxidative loss of shelf-life in processed cakes than 0.1 g kg-1 fat of BHA and BHT. Black pepper leaves appeared to be pro-oxidative (Lean and Mohamed, 1999). Extracts of garcinia and turmeric powder also were found to be suitable for use in biscuits as natural antioxidants and results were comparable with BHA.

The extracts of the amla, drumstick leaves and raisins were used as sources of natural antioxidants in biscuit. All extracts exhibited an excellent antioxidative effect on the biscuits in comparison with

Table 2. Mould ratio of sample cakes to control.

Sample	Day			
	10	30	45	60
Control	1.0±0.0[a]	1.0±0.0[a]	1.0±0.0[a]	1.0±0.0[a]
BHA-0.01	0.91±0.08[b]	0.85±0.1[ab]	0.93±0.3[b]	0.92±0.02[a]
BHA-0.02	0.85±0.07[b]	0.51±0.26[c]	1.01±0.03[a]	0.82±0.11[a]
CEO-0.05	0.0±0.0[d]	0.76±0.05[b]	0.22±0.04[c]	0.16±0.04[bc]
CEO-0.10	0.29±0.09[c]	0.49±0.1[c]	0.19±0.03[c]	0.29±00.02[bc]
CEO-0.15	0.0±0.0[d]	0.16±0.03[d]	0.06±0.00[d]	0.37±0.05[b]

* In the first day, the number of moulds in all samples was zero.
** Means within each column with the same superscript letters are not significantly different ($p<0.01$).

BHA. The higher efficiency of the plant extracts was contributed to the stability of this natural antioxidant during baking (Reddy *et al*., 2005).

Antioxidant effects of CEO in cake may be due to the presence of terpenoid components (Camphor, limonene, α-pinene and geraniol). Several researchers have reported the antioxidant activities of monoterpenes and diterpenes (Yanishlieva *et al.*, 1999; Grassman, 2006). Phenolic antioxidants are proton donors which act as inhibitors for radical chain reactions on autoxidation of organic substrates. Radical scavenging activity (RSA) of coriander seed oil and oil fractions were investigated and stated Coriander seed oil and its fractions exhibited the strong RSA and can use as a natural antioxidant in lipid-containing foods (Ramadan *et al.*, 2003). In another study, by three different bioassays, indicated that extract and oil of leaves and seeds of coriander has strong antioxidant activity and thus, probably prevent oxidative deterioration of food (Wangensteen *et al.*, 2004). The extract of coriander leaves was added to refined sunflower and groundnut oils heated to frying temperature and were kept for four weeks. This plant had good antioxidant activity and it is stable at high temperatures and can be used as substitutes for synthetic antioxidant.

Antifungal activity of CEO

Moisture content and pH of the foodstuffs have been reported as the main biotic factors affecting the fungal deterioration. In the present investigation, the moisture content and pH of cakes ranged between 13.0 to 15.5% and 6.64 to 7.81, respectively. So, the fungal distribution on the samples was not influenced by their moisture content and pH. Mould ratio of sample cakes per control is showed in Table 2. After 30 days, the percentage of moulds in cake containing CEO at 0.05% didn't differ from cake with 0.01% BHA and CEO at 0.10% didn't differ from cake with 0.02% BHA, since cake with BHA had no preservatives, so it could be as control sample, hence we could result that CEO at 0.05 and 0.10% concentration could not prevent growth of moulds in cake, but by increasing the concentration of the essential oil, antifungal activity of the CEO increased, since CEO at 0.15% had significantly lower percentage of moulds than control and BHA samples. So, CEO at 0.15% had antifungal activity in cake.

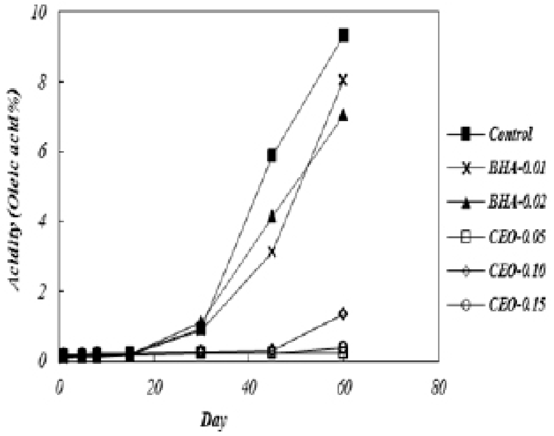

Figure 3. Effect of CEO on FFA of samples during 60 days storage at room temperature.

Formation of free fatty acid is also a measure of the rancidity of foods. FFA is formed because of hydrolysis of triglyceride and may get promote by enzymatic activity due to microbial growth (Politeo *et al*., 2007). After 30 days of storage, acidity of control and samples containing BHA at 0.01 and 0.02% was higher than cakes containing different concentrations of CEO (Figure 3). According to the mould ratio, this behavior may be due to mould growths and production of hydrolytic enzymes. However, CEO may be inhibiting the mould growth or decrease the activity of hydrolytic enzymes, thus acidity was low in samples containing CEO.

Common moulds found in cakes and bakery products are *Penicillium expansum, Penicillium stoloniferum, Rhizopus stolonifer, Aspergillus niger, Monilia sitophila* and species of Mucor and Geotrichum. Among these, *Penicillium expansum, Penicillium stoloniferum* and Mucor are mycotoxin producers (Lean and Mohamed, 1999). The contamination of the butter cakes was from various moulds, which in this study, the types of moulds were not identified. CEO at 0.15% had antifungal activity in the cake. Other studies reported antimicrobial effect of essential oils of *Coriandrum sativum*. CEO showed antimicrobial activity against five species of Candida albicans (Begnami *et al*., 2010). Also, it is reported that coriander essential oil has pronounced antibacterial activity against both Gram positive

Table 3. Sensory scores of the cakes after 10 days storage at 25 °C

Sample	Color	Taste	Texture	Acceptability
Control	4.60±0.59[ab]	3.95±0.99[a]	4.30±0.65[ab]	4.30±0.65[ab]
BHA-0.01	4.4±0.68[ab]	3.75±0.71[a]	3.95±0.68[abc]	3.85±0.49[abc]
BHA-0.02	4.45±0.75[ab]	3.90±0.71[a]	4.05±0.75[abc]	4.05±0.6[abc]
CEO-0.05	4.55±0.6[ab]	2.90±1.07[b]	4.29±0.78[ab]	3.40±0.99[cd]
CEO-0.10	4.25±1.06[b]	2.85±1/18[b]	4.10±1[abc]	2.95±1.09[d]
CEO-0.15	4.20±0.76[b]	2.80±1[b]	3.70±1.1[c]	2.85±1.03[d]

* Means within each column with the same superscript letters are not significantly different ($p<0.01$).

(*Staphylococcus aureus* and *Bacillus* spp.) and Gram negative (*Echerichia coli, Salmonella typhi, Klebsiella pneumonia* and *Proteus mirabilis*) bacteria (Cantore *et al*., 2004; Matasyoh *et al*., 2009). In addition, this extract showed anthelmintic activities against *Haemonchus contortus*.

Several researchers have been described the antifungal activity of limonene, a terpenoid hydrocarbon isolated from different plant species including coriander, against *Aspergillus niger* (Razzaghi-Abyaneh *et al.*, 2009). Other plants have also reported that have antimycotic activity in bakery products. Six different plants, turmeric, lemon-grass, *Garcinia atriviridis* and clove have been reported were efficient in retarding mould growth in butter cakes and these effect attributed to oxygenated sesquiterpenes and monoterpene hydrocarbons in turmaric and eugenol and phenolic compounds in clove. Betel leaves and black pepper leaves showed no antimycotic activity and may even promote mould growth in butter cakes (Lean and Mohamed, 1999). The volatile substances from mustard, cinnamon, garlic and clove essential oils were effective in the control of common bread spoilage fungi (Nielsen and Rios, 2000). Fungicidal activity was also found in lemongrass essential oil followed by cinnamon bark and thyme oils (Inouye *et al*., 2000). Goynot *et al*. (2003), investigate the effect of volatile fractions of 16 essential oils on the more common fungi causing spoilage of bakery products including, *Eurotium amstelodami, E. herbariorum, E. repens, E. rubrum, Aspergillus flavus, A. niger* and *Penicillium corylophilum*. They reported that volatile substances from cinnamon leaf, clove, bay, lemongrass and thyme essential oils had good antifungal activity against common fungi causing spoilage in bakery products but these essential oils show poor activity when they were used in a sponge cake. However, comparison of the data obtained by different studies is difficult, because of differences in plants extract compositions, in methodologies followed to assess antimicrobial activity and in microorganisms chosen to be tested (Guynot *et al*., 2003).

Sensory analysis of cakes

The cakes were subjected to sensory analysis by 25 trained panelists. Table 3 summarizes the results of sensory analysis of the produced cakes and gives the mean scores for four variations after 10 days storage. It was observed that cakes containing CEO at 0.05, 0.10 and 0.15% were acceptable in the terms of color and texture in comparison with control and cakes containing 0.01 and 0.02% BHA. Cakes with CEO have shown lower acceptability in terms of taste. The overall acceptability of cakes containing 0.05% CEO was close to that of BHA. Increasing the concentration of CEO to 0.15% had small negative effect on overall acceptability. Nanditha *et al*. (2008), explored microstructure and electrophoretic characterization of biscuit dough to understand the influence of natural antioxidants from plant sources such as garcinia, peltophorum ferrugineum flower extract, turmeric powder and curcumin, in the processing of biscuits. The study indicated that protein subunits of biscuit dough were not affected by these antioxidants. Extracts of garcinia and turmeric powder were found to be suitable for use in biscuits as natural antioxidants and results were comparable with BHA.

Conclusion

CEO showed good antioxidant activity in butter cake and its effect was compared with the synthetic antioxidant (BHA 0.02%). CEO also showed better antifungal activity in sample cakes at 0.15%. These effects could be due to the presence of terpenes and terpenoids compounds in the CEO. The overall acceptability of cakes containing 0.05% CEO was almost equal to that of BHA, however, cakes which containing 0.10 and 0.15% of CEO were less acceptable than control and BHA containing sample. It seems, if the active substances of CEO could be added to the cake then we could have samples with desirable organoleptic properties. Results showed that this essential oil could be used as natural antioxidant and antifungal in foodstuffs especially those lipid containing.

Acknowledgments

The authors thank the financial supports of Tarbiat Modares University Research Council and Center of Excellency for Recycling and Losses of Strategic Agricultural Products.

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