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Comparison of Antioxidative Effect of Tea and Sesame Seed Oils Extracted by Different Methods

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ABSTRACT

Tea and sesame seed oils extracted by different methods (including supercritical fluid extraction (SFE), at a pressure of 350 atm, a temperature of 60°C, a static extraction time of 20 minutes, a dynamic extraction time of 30 minutes and 150 g kg^{-1} of ethanol as a modifier). Solvent (SE) and ultrasound-assisted solvent extraction (UE)), as natural antioxidants, were evaluated during 15 days storage by monitoring their effects on refined sunflower oil at 60°C. The peroxide value (PV) and the thiobarbituric acid (TBA) value were used to assess the antioxidant activity of these extracted oils. The highest extraction yields were from SE and SFE, while UE gave a lower yield. Considering oil extraction yield and antioxidant activity, SE and SFE were the preferred methods. The results showed that tea seed oil had strong antioxidant activity, which was almost equal to sesame oil. Therefore, tea seed oil can be used in the same way as sesame seed oil in fat, oil and other food products as a natural antioxidant to suppress lipid oxidation.

Keywords: Antioxidant activity, Sesame seed oil, SFE, Solvent extraction, Sunflower oil.

INTRODUCTION

Antioxidants are organic substances. By adding them to fats, autoxidation is retarded and a food product's shelf life is extended. Antioxidants can be synthetic or natural in origin. The use of synthetic antioxidants is restricted in several countries, because of possible undesirable effects on human health [1] and so, there is great interest in obtaining and utilizing antioxidants from natural sources, especially of plant origin [2].

Different extraction methods, such as liquid-solid extraction (by stirring), Soxhlet, sonication and supercritical fluid extraction have been used to isolate oils from the plants seeds. However, none of these is optimal. The main disadvantages of Soxhlet extraction are the generally long extraction time and limited solvent choice. Solvent extraction aided by sonication and stirring may also be time-consuming, and requires large volumes of solvents that are sometimes hazardous and require concentration steps.

Supercritical fluid extraction (SFE) with CO_2 is an attractive method for the food industry because the solvent (CO_2) is safe, non-toxic and is easily removed. The technique is fast and the extraction parameters can be changed over a wide range of pressures and temperatures.

Camellia seed has been utilized in China for more than 1,000 years. Tea seed oil is a high quality cooking oil, which is comparable to olive oil. It stores well at room temperature. Tea seed oil is also a good industrial raw material and is used in the manufacture of soap, margarine, hair oil and paint [3].

Roberts and Silva (1972) found that tea seed oil could be stored at room temperature for about three months without any quality loss or free fatty acid (FFA) content. Its degree of unsaturation is similar to that of olive, corn and sesame oils [4]. Ravichandran (1993) determined the storage stability of south Indian tea seed oil. They reported that crude tea seed

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oil had high storage stability, due to a low content of the acylglyceroles of linoleic and linolenic acids and the presence of polyphenols and vitamin E as antioxidants [5]. Sahari *et al.* (2004) studied the characteristics and shelf life of Iranian tea seed, sunflower and olive oils by the oven test method (at 63°C) [6]. They reported that tea seed oil had an antioxidant effect and it enhanced the shelf life of sunflower oil at a 5% level.

Sesame (*Sesamum indicum* L.) is one of the most important global oilseed crops [7] and is consumed as a nutritious food. The oxidative stability of sesame oil is superior to that of other vegetable oils although it contains nearly 85% unsaturated fatty acids [8]. For this reason; it was selected for the purpose of comparison with tea seed oil.

This study compares SE, UE and SFE applied to tea and sesame seeds to determine which process would give the highest yield of antioxidatively active components. The study also evaluated the effect of extracted tea and sesame seed oil (at 50, 100 and 150 g kg^{-1}) on the oxidative stability of sunflower oil stored under Schaal oven conditions (60°C).

MATERIALS AND METHODS

Tea (cv. *Lahijan*) and sesame (cv. *Karaj 1*) seeds were obtained from Lahijan Tea Research Center and the Plant and Seed Research Center of Iran, respectively. The tea and sesame seeds were dried (25°C) in the dark to a moisture content of 70 and 80 g kg^{-1}, respectively. The dried seed was then kept in a cool place (4°C) until testing.

The following chemicals were used: n-hexane, ethanol, petroleum benzene from Merck (Darmstadt, Germany), thiobarbituric acid, potassium iodide and sodium thiosulphate from Aldrich (Milwaukee, WI USA), and CO_2 (99.99%) from Roham Gas Co. (Tehran, Iran).

Five grammes of dried and ground seed (milled through, mesh 40) were placed in a beaker and extracted with 150 ml of petroleum benzene (b.p. 50-70°C) by stirring the mixture for 4 hours with a magnetic stirrer. After extraction, the solvent was removed from the extracted oil in a vacuum rotary evaporator (Heidolph, Germany) at 50°C. All extractions were in triplicate.

Five grammes of dried, ground seed was mixed with 70 ml of petroleum benzene (b. p. 50-70°C) in a flask. The mixture was sonicated in an ultrasonic bath (Elma Transonic model 690/H, 35 kHz, Germany) for 30 minutes. The solvent in the extracted oils was evaporated at 50°C.

Supercritical fluid extraction was carried out in the Suprex MPS/225 system (Pittsburgh, PA, USA). Exactly 2.0 g of powdered seed (mesh 40) was mixed with an appropriate quantity of glass beads and placed in the extraction vessel (10 ml). The samples were covered with filter paper at the top and bottom of the extractor. A Duraflow manual variable restrictor (Suprex) was used in the SFE system to collect the extracted oils. The extracted oils were collected in 3-4 ml of hexane. The restrictor was heated at 40°C to prevent freezing. The supercritical carbon dioxide flow rate through the Duraflow restrictor was approximately 1.0±0.1 ml min^{-1} (compressed).

The tea and sesame seed oils were extracted under optimum extraction conditions (350 atm, 60°C, 30 min and 150 g kg^{-1} ethanol as a modifier) as previously reported by our research group [9].

The oils extracted (from tea and sesame seed) by SFE, solvent and ultrasound-assisted solvent extraction methods were added to refined sunflower oil at 50, 100 and 150g kg^{-1}. The oven test method at 60°C was used to check stability [10]. The PV and TBA indices of oils stored under accelerated oxidation conditions were determined periodically according to AOCS guidelines [11] and IUPAC standard methods [12], respectively. A control sample was prepared, under the same conditions, without adding tea or sesame seed oil. All the experiments were carried out in triplicate and the results were averaged.

Statistical Analysis

Statistical analysis was performed using ANOVA, and significant differences among means from triplicate analyses at ($P<0.05$) were determined by LSD using the Statistical Package of Social Science (SPSS).

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RESULTS AND DISCUSSION

The extraction yields for SE, SFE and UE were 23.3, 31.6 and 21.0%, respectively. The main fatty acids of tea and sesame seed oils were as follows: palmitic (21.5%), stearic (2.9%), oleic (49.5%), linoleic (20.4 %) and palmitic (8.7%), stearic (5.1%), oleic (47.2 %), linoleic (39.0 %), respectively.

The PV and TBA indices were measured during accelerated oxidation at 60°C for 12 and 15 days, respectively. The PV is a chemical indication of how much of the oil is in the early stages of oxidation, and it reflects the degree of oxidation. The effect of tea seed oil addition on oil PV reduction indicates the potential of tea seed oil as an antioxidant (AO) to inhibit lipid peroxidation in its early stages by reducing formation of the primary lipid oxidation product (LOP).

Aldehydes (especially malonaldehyde) –the breakdown products (secondary LOPs) of oxidized fatty acids- produce off-flavors in oxidized oils that can be quantified through their reaction with TBA [13].

Changes in the PV and TBA index of the refined sunflower oil without the addition of extracted oils (the control), and with extracted tea or sesame seed oil (extracted by different extraction methods, at 50, 100 and 150 g kg^{-1}) were incorporated during 12 and 15 days storage. The results are shown in Figures 1 and 2. Figure 1 shows that all the extracted oils, irrespective of extraction method, slowed down the rate of peroxide formation, since the PVs of all the samples which contained extracted oil were lower than in the control sample, during storage.

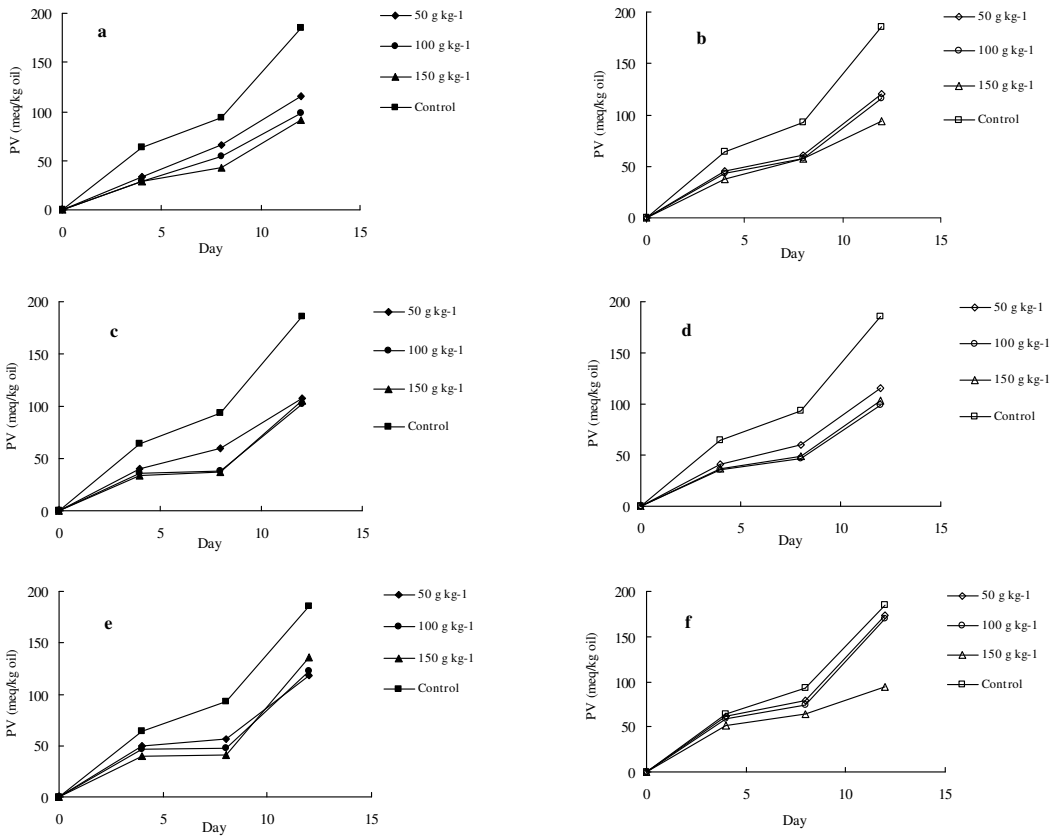

Figure 1. Effect of extracted tea and sesame seed oil on refined sunflower oil oxidation expressed as PV formation at 60°C. (a), (c) and (e): Tea seed oil extracted by solvent, ultrasound assisted solvent and SFE methods, respectively; (b), (d) and (f): Sesame seed oil extracted by solvent, ultrasound assisted solvent and SFE methods, respectively.

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Figures 1a and 1b show that the antioxidant activities (AOA) of tea and sesame seed oil (by the SE method) increased with their concentration in the range 50-150 g kg^{-1}. There was a significant difference ($P<0.05$) between the 150 g kg^{-1} and 50 g kg^{-1} level. As seen in Figures 1c and 1d, the AOA of extracted tea and sesame oil by UE was constant at different concentrations. The AOA of tea seed oil was not significantly different at 50, 100 and 150 g kg^{-1} ($P<0.05$). The AOA of 100 and 150 g kg^{-1} extracted sesame oil were not significantly different ($P<0.05$), but the difference at 50 g kg^{-1} was significant. As seen in Figure 1e, the AOA of extracted tea seed oil (by the SFE) was constant with increased extracted oil concentration and the AOA was not significantly different ($P<0.05$) at any level. There was an inverse trend with sesame oil (Figure 1f). The AOAs of 50 and 100 g kg^{-1} of sesame oil were significantly different from 150 g kg^{-1}. In addition, Figure 1 shows that sunflower oil samples treated with different concentrations of tea seed oil, extracted by the SE method, generally had better stability compared with the other methods.

The TBA values of refined sunflower oil samples treated with tea and sesame seed oil (extracted by SE, UE and SFE methods) are given in Figure 2. Figure 2a shows that the AOA of extracted tea seed oil (by the SE method) was not significantly different at 50, 100 and 150 g kg^{-1} ($P<0.05$). However, Figure 2b shows that the AOA of sesame oil increased when the amount added was increased to 150 g kg^{-1}. On the other hand, as Figures 2a and 2b show, tea seed oil at 50-150 g kg^{-1} exhibited stronger AOAs than the same level of sesame oil. Figure 2c shows that tea seed oil (extracted by the UE method) has a better AOA at 100 g kg^{-1} compared with sesame oil (Figure 2d). The AOAs of oils extracted by

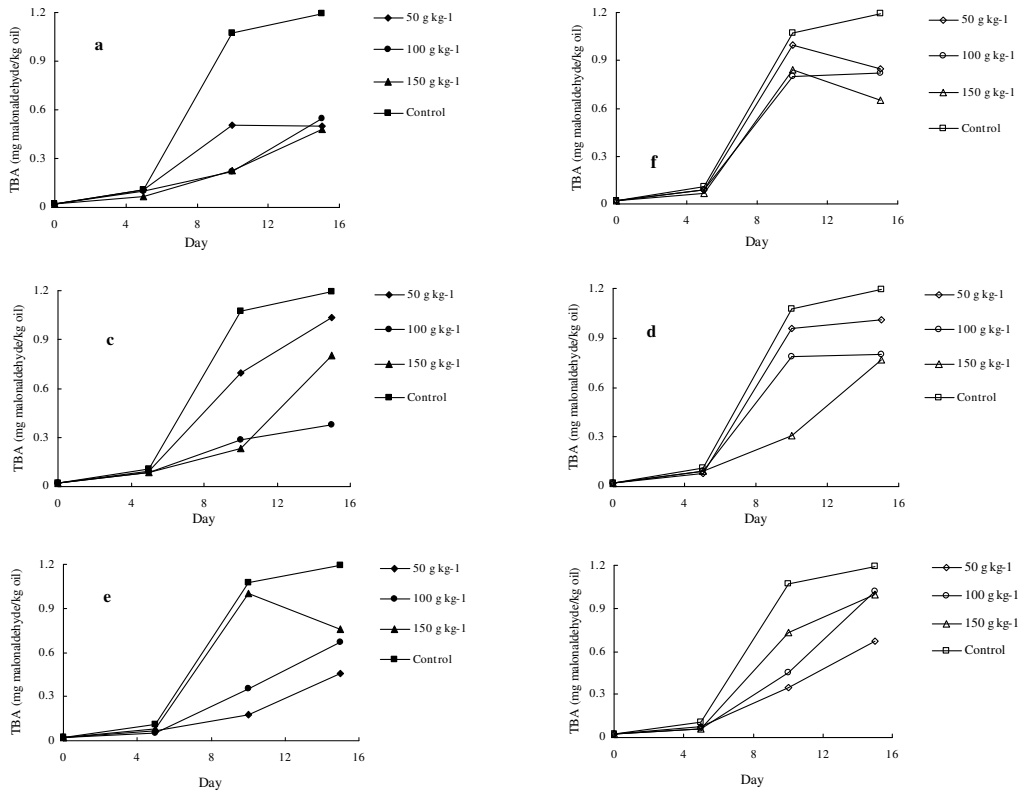

Figure 2. Changes in the TBA values of refined sunflower oil treated with tea and sesame seed oil (extracted by different methods) during storage at 60°C. (a), (c) and (e): Tea seed oil extracted by solvent, ultrasound assisted solvent and SFE methods, respectively; (b), (d) and (f): Sesame oil extracted by solvent, ultrasound assisted solvent and SFE methods, respectively.

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Table 1. Effect of oil type and extraction method on the PV and TBA indices of sunflower oil. [a]

Method	PV (meq kg^{-1} oil)		TBA value (mg malonaldehyde kg^{-1} oil)	
	Tea seed oil	Sesame seed oil	Tea seed oil	Sesame seed oil
SE	101.4 [b]	110.1 [b]	0.51 [a]	0.67 [ab]
UE	105.1 [b]	105.9 [b]	0.74 [ab]	0.77 [b]
SFE	125.5 [a]	145.8 [a]	0.63 [ab]	0.73 [ab]

[a] Values with different letters are significantly different at $P < 0.05$.

SFE are shown in Figures 2e and 2f. The antioxidant effect decreased when the amount added was increased gradually to 15%. The AOA level of 100 and 150 g kg^{-1} of tea seed oil was not significantly different ($P<0.05$). Figure 2f shows the same trend.

The results of this study show that tea seed oil exhibited a strong antioxidant activity, almost equal to that of sesame oil. The high stability of tea seed oil is probably due to the low content of glycerides of linolenic and linoleic acids and to the presence of polyphenols and vitamin E and other antioxidants [5, 6]. In addition, a comparison between the antioxidant effect of tea and sesame seed oils extracted by different methods showed that samples treated with tea and sesame seed oil extracted by the SFE method, and in some cases by the SE, had higher levels of AOAs than the other methods. The AOA and amount of added tea seed oil (extracted by the SFE) were inversely related. The observed differences, compared with other extraction methods, reflect the ability of SFE to extract antioxidative components from tea seed (Figures 1 and 2). Thus, SC-CO_2 is a good solvent for the extraction of non-polar and moderately polar compounds [14, 15]. It is therefore an excellent choice for extracting fats and oils without co-extracting other interfering polar and non-fat components. In this way, it is possible to extract triglycerides without the co-extraction of phospholipids which are generally present in oil-seeds. However, to extract polar compounds such as phospholipids, some sort of modifier or co-solvent must be added to the SF CO_2. Although there are many potential modifiers, the most commonly used modifier is ethanol. It increases the polarity of the SF CO_2 and allows the extraction of more polar compounds not removed by SF CO_2 alone. It is a good choice because of it's generally regarded as safe (GRAS) status, availability and cost. So, in this study ethanol was used as modifier. Modified SF CO_2 can extract polar antioxidative compounds from tea seed. Sometimes, the AOA of phenolic compounds are decreased at high concentration and they became prooxidants. Huang *et al*. (1995) showed that increased addition of α-tocopherol to oil might increase the PV while reducing hexanal formation [16]. Finally, the results suggest that the tea seed and sesame seed oils have antioxidant properties and could be used as alternative natural antioxidants (Table 1). No single compound can be considered responsible for the observed stability. This study will provide a base for future studies in this area.

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مقایسه خاصیت آنتی‌اکسیدانی روغنهای بذر چای و کنجد استخراج شده به روشهای مختلف

ا. رجایی، م. برزگر و م. ع. سحری

چکیده

اثر آنتی‌اکسیدانی روغن‌های بذر چای و کنجد استخراج‌شده به روشهای مختلف (شامل استخراج با سیال فوق بحرانی، SFE، در فشار ۳۵۰ اتمسفر، دمای ۶۰ درجه سانتی‌گراد، زمان استخراج استاتیک ۲۰ دقیقه، زمان استخراج دینامیک ۳۰ دقیقه و ۱۵۰ گرم/ کیلوگرم اصلاحگر اتانول) با حلال (SE) و استخراج با حلال به کمک امواج ماورائ صوت (UE))، در روغن تصفیه‌شدهٔ آفتابگردان نگهداری شده در دمای ۶۰ درجهٔ سانتیگراد به مدت ۱۵ روز ارزیابی شد. از مقادیر پراکسید (PV) و تیوباربیتوریک اسید (TBA) به‌عنوان شاخص فعالیت آنتی‌اکسیدانی روغنهای استخراج شده استفاده شد. روش استخراج با حلال (SE) و SFE بیشترین و روش UE کمترین راندمان استخراج را داشتند. براساس راندمان استخراج روغن و فعالیت آنتی-اکسیدانی، روشهای SE و SFE به عنوان روشهای بهتر انتخاب شدند. همچنین نتایج مشخص کرد که روغن بذر چای فعالیت آنتی‌اکسیدانی بالایی دارد که تقریباً معادل فعالیت آنتی اکسیدانی روغن کنجد است. بنابراین به‌طور مؤثری می‌توان روغن بذر چای را، مشابه روغن کنجد، به عنوان یک آنتی‌اکسیدان طبیعی به منظور متوقف ساختن اکسیداسیون چربی، در روغن به‌کار برد.

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