Effects of adding green tea extract on the oxidative stability and shelf life of sunflower oil during storage

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ABSTRACT

This study aimed to compare different concentrations effect of green tea extract (GTE) (200, 400, and 800 ppm) with TBHQ (75 ppm) in extend the shelf-life of sunflower oil (SO) and to evaluate the protective effect of GTE on the oxidation of refined SO. The sample's peroxide value (PV), acidity value (AV), anisidine value (*p*AV), Totox value (TV), oxidative stability, and total phenol content (TPC) were analyzed at specific intervals during 12-month at 25 °C and 60-day at 60 °C. The optimum kinetic model corresponding to the first order for PV, TV, and *p*AV was obtained at 25, 35, and 45 °C. SO containing GTE (800 ppm) had a similar performance to TBHQ at 25 °C and 60 °C and possessed a longer shelf life than samples treated with TBHQ. Due to synthetic antioxidant's health risk and toxicity, GTE can be a good substitute for TBHQ in the edible oil industry.

1. Introduction

One of the most important oil seeds in the world is sunflower (*Helianthus annuus*) (Adeleke and Babalola, 2020). SO is a rich source of monounsaturated (MUFAs) and polyunsaturated fatty acids (PUFAs), including oleic acid (14–40 %) and linoleic acid (48–74 %), with positive health effects, including brain function development, preventing heart disease, control of the obesity and tumor progression, and inducement ROS-mediated cell death of pro-tumor macrophage (Liu et al., 2020; Liu et al., 2017; Emami et al., 2016). The structures of MUFA and PUFA are dependent on the position of double bond counting from the carbon chain methyl end (Liu et al., 2020). However, SO is more prone to oxidation and/or degradation due to a relatively higher proportion of unsaturated fatty acids. The auto-oxidation of SO during processing, storage, and consumption has raised serious concerns regarding the safety and quality of SO (Sharma et al., 2019). Furthermore, exposure to light, heat, high temperatures, trace metals, and oxygen can increase the degradation of unsaturated fatty acids (Rashidi et al., 2016). To address this issue, Farhoosh and Nyström, (2018) stated that the application of synthetic and/or natural antioxidants can be an ideal way to limit liquid oxidation and degradation. However, recent studies have shown that the prolonged consumption of synthetic antioxidants (particularly BHA, BHT, and TBHQ) can lead to serious health

concerns such as carcinogenesis and cancer. As a result, they are excluded from the Generally Recognized as Safe List of Compounds (GRAS) (Rashidi et al., 2016; Sheybani et al., 2023 a & b). Therefore, due to safety concerns, there is an increasing trend to replace synthetic antioxidants with natural ones. The effectiveness of various natural antioxidants in terms of their scavenging activity, chelating ability, and reducing power has been previously reported to successfully replace synthetic antioxidants (Altemimi et al., 2017). However, due to limited scientific and technological data, seasonal availability of resources, and higher input costs, natural antioxidants have not been used on a large commercial scale. Researchers are currently focused on finding a cost-effective and commonly available origin of phytochemicals (Jain et al., 2021). Natural antioxidants or bioactive agents with antioxidant properties like tannins, phenolics, terpenoids, flavonoids, curcumanoids, lignans, coumarins, anthocyanins, xanthons, vitamins, carotenoids, and phospholipids are existed in different parts of plants as fruits, seeds, oils, and leaves which they are known to maintenance the oxidizable components of the edible oil or foods from oxidation damage (Shahidi & Camargo., 2016; Keivanfar et al., 2023; Ghreishi Rad et al., 2023). Alpha-tocopherol as a major tocopherol acts as the pro-oxidant and delays oxidation. The optimal effective amount of α-tocopherol in several edible oils like sunflower oil (50 ppm), olive oil (100 ppm) corn oil (250–500 ppm) was reported (Nain et al., 2021). There are several

studies on the effect of natural antioxidants extracted from different parts of plants compared to synthetic antioxidants for reducing edible oil oxidation which some of natural antioxidants showed better or similar antioxidant properties compared to synthetic antioxidants (Sharme et al., 2019; Nain et al., 2021; Silva et al., 2021; Blasi et al., 2020; Keivanfar et al., 2023). Given their health benefits and with their antioxidant properties, it is interesting to explore the usage of natural extracts like green tea in food applications. Considering the necessity of phytochemicals that are safe, nutritious, and appealing to the senses, green tea could be a suitable option. Green tea, which is particularly popular in Asian countries, is among the most widely consumed beverages worldwide (Shrivastava et al., 2018). Green tea leaves contain bioactive compounds, including polyphenols, flavonoids such as flavanols (catechins and procyanidins), flavonols (rutin, quercetin, and kaempferol), and phenolic acids (gallic acid and caffeic acid) that offer a range of health benefits (Gramza-Michałowska et al., 2016; Lorenzo and Munekata, 2016). Catechins have been found to have moderate antioxidant properties and antimicrobial activity against both Gram-positive and Gram-negative bacteria (Parvez et al., 2019). Epigallocatechin (EGC), epigallocatechin gallate (EGCG), epicatechin (EC) and epicatechin gallate (ECG) are the major catechins of GTE. GTE antioxidant properties were studied in marine oils (Wanasundara & Shahidi, 1997; Nain et al., 2021), sponge-fat cakes (Kozlowska et al., 2019), biscuits (Mildner-Szkudlarz et al., 2009), pastry products (Żbikowska et al., 2017), meat (Tian and Hung, 2019), butter (Thakaeng et al., 2020), bulk oil (Shen et al., 2021) and table spreads (Dwyer et al., 2012).

To the best of our knowledge, the application of GTE as a natural antioxidant in sunflower oil and monitoring of its quality for one year has not yet been reported. Therefore, the objective of the present study was to evaluate the potential of GTE as a natural antioxidant source to extend the shelf life of sunflower oil. For this purpose, different concentrations of GTE were added to sunflower oil samples and then quality tests were performed for one year at 25 °C and for 60-day at 60 °C. Results were compared to those obtained for sunflower oil samples containing 75 ppm of TBHQ. The kinetic parameters for three thermal stages of sunflower oil samples containing 800 ppm of GTE, blank, and 75 ppm of TBHQ were determined.

2. Materials and methods

2.1. Materials

The lipid soluble green tea extract (GTE) containing 81 % palymitoylated catechins (expressed as epigallocatechin gallate (EGCG)) in sunflower oil (US FDA GRAS Notice GRN 000772; product code: 356150), which belongs to the Theaceae family, was purchased from Kemin (Belguim). TBHQ, ethanol, 2–2-diphenyl-1-pycrylhydrazyl (DPPH), and Folin-Ciocalteu reagent were purchased from Merck (Darmstadt, Germany). The sunflower oil was obtained from Behshahr Company (Tehran, Iran). All other reagents and chemicals used in this study were purchased from Sigma-Aldrich Company (St. Louis, Missouri, United States) with an analytical grade.

2.2. Preparation of the oil samples

In this study, refined SOs without any antioxidants were prepared, and then treated with three different concentrations of GTE (200, 400, and 800 ppm) (as named SO-GTE$_{200ppm}$, SO-GTE$_{400ppm}$, SO-GTE$_{800ppm}$) as well as TBHQ at a concentration of 75 ppm (SO-TBHQ$_{75ppm}$). The control sample, which did not contain any antioxidants, was processed under similar storage conditions. The oil samples were evaluated during 12 months of storage (every 30-day) at room temperature in a dark place. In addition, a Shaal oven test was performed on the samples maintained in an oven at 60 °C for 60 days. The kinetic model for PV, TV, and pAV was obtained at 25, 35, and 45 °C shelf life of the samples (blank, SO-GTE$_{800ppm}$, and SO-TBHQ $_{75ppm}$).

All samples were prepared in triplicate, and all measurements were repeated three times for each test.

2.3. Peroxide value (PV)

ISO 3960 (2017) method was used to assess the PVs of the oil samples. The determination was conducted by titration of KI (0.1 N) saturated solution of the oil with 0.01 N Na$_2$S$_2$O$_3$ and starch as an indicator. The PV was calculated using the Eq. (1):

$$PV = \frac{(V_2 - V_1)}{M} \times T \times 1000 \tag{1}$$

where PV is the peroxide value (meqO$_2$/kg oil), M is the quantity of oil (g), V$_2$ is the volume of Na$_2$S$_2$O$_3$ (mL), V$_1$ is the volume of blank (mL), and T is the normality of Na$_2$S$_2$O$_3$ (0.01 N).

2.4. p-Anisidine value (pAV)

pAV of the oil samples was assessed based on the ISO 6885 (2016) method as follows: 1 g of the oil sample was dissolved in isooctane in 25-mL conical flasks. Then, 5 mL of this solution was mixed with 1 mL of 0.25 % p-anisidine solution in glacial acetic acid. After 10 min, the solution adsorption was read at 350 nm using a spectrophotometer (Perkin Elmer 2500, USA). This method provides a measure of the secondary oxidation products in the oil samples, particularly aldehydes, which are responsible for the rancid smell and taste. pAV was calculated using the Eq. (2):

$$pAV = 25 \times \frac{(1.2A_s - A_b)}{m} \tag{2}$$

A$_s$, A$_b$ was illustrative of solution adsorption before and after reaction with the p-anisidine solution, and m is the mass of the sample (g).

2.5. Total oxidation (TOTOX) value (TV)

TOTOX value (TV), which indicates both primary and secondary oxidation products, is calculated by adding twice the PV to the pAV (3), as described by Abbas Ali et al. (2017).

$$TV = 2 \times PV + (pAV) \tag{3}$$

2.6. Oxidative stability

The oxidative stability of the oil samples was analyzed according to the ISO 6886 (2016) method using a rancimat apparatus (Metrohm, model 743, Herisau, Switzerland). The determination was performed at 110 °C with a 20 L/h air flow and 60 mL of distilled water in the reaction vessel, using 2.5 ± 0.5 g of SO samples. The control sample was prepared using SO without antioxidants. The conductivity cell was used to detect volatile decomposition products (Liu et al., 2018). The induction time for each model system was determined by triplicate analyses.

2.7. Acidity value (AV)

The AV of the oil samples was determined by potentiometric titration using an automatic titrator model G20 (Mettler Toledo, Urdrof, Switzerland) following the method of ISO 660 (2020). Determination was conducted by titration using 0.1 N NaOH and phenolphthalein as an indicator.

The AV is calculated using Eq. (4):

$$AV = \frac{V \times C \times M}{10 \times m} \tag{4}$$

where M is the molar mass (g), V is the volume of NaOH solution (mL), C is the exact concentration of NaOH standard solution (mole/L), and m is

the mass of sample (g).

2.8. Antioxidant activity

To determine the antioxidant activity of the samples, the DPPH (2, 2-diphenyl- 1-picrylhydrazyl) reagent was used. This method involves discoloration of a methanolic solution of DPPH reagent, which was prepared by mixing 2.5 mg DPPH and 100 mL methanol. 3.9 mL of the DPPH methanolic solution was added to a 100 µL methanolic extract of the samples. The obtained mixture was left in a dark place for 60 min, and the absorbance was recorded at 517 nm using a UV/VIS spectrophotometer. The percentage of DPPH radical inhibition by the extract was calculated using the Eq. (5):

$$Antioxidant\ activity\ (\%) = \frac{(A_C - A_S)}{A_S} \times 100 \qquad (5)$$

where A_C and A_S are the absorbance of the control and sample, respectively (Yousefi et al., 2023; Sheybani et al., 2023 a & b).

2.9. Total polyphenol content (TPC)

The total polyphenol content of the samples was assessed using a modified Folin-Ciocalteu method as described by Sheybani et al., (2023 a & b). Briefly, 1 mL of the extract (5 mg/mL) was mixed with 1 mL of Folin-Ciocalteu reagent. Then, 1 mL of 7.5 % saturated sodium carbonate solution was added to the mixture, and the total volume was adjusted to 10 mL using distilled water. The mixture was kept in a dark place for 90 min, and the absorbance was measured at 720 nm using the spectrophotometer. TPC was expressed as mg of gallic acid equivalents (GAE)g^{-1} of SO oil which the gallic acid (GA) calibration curve was obtained at different concentrations of GA (0–200 µg/mL) (y = 0.0159x-0.0361, R^2 = 0.988).

2.10. Schaal oven test

The oxidative stability of SO oil samples at high temperatures was assessed as follows: 250 g of each sample was transferred into a 300 mL dark glass bottle and then incubated at 60 °C in an oven. The samples were then evaluated at 0, 15, 30, and 60 days (Walallawita et al., 2016).

2.11. Kinetic study of oxidation in sunflower oil samples

Three storage temperatures (25 °C, 35 °C, and 45 °C) were chosen and sampling was performed at 0, 30, and 60 days. Then, PV, pAV, and TV were determined for all the samples. The modification in the quality of lipids can be determined using the following Eq. (6):

$$r_A = -\frac{dA}{dt} = KA^n \qquad (6)$$

where A is the indices (PV, p-AV, and TV); n is the apparent order of reaction which can be 0 (7), 1 (8), and 2 (9); and K is the rate constant.

$$Zero\ order : A = A_0 - Kt \qquad (7)$$

$$First order : A = A_0 e^{-Kt} \qquad (8)$$

$$Second order : \frac{1}{A} = \frac{1}{A0} - Kt \qquad (9)$$

Good linearity and fit indicated the best reaction order. The best kinetic model was obtained by plotting the indices (PV, pAV, and TV) vs. time and calculating the regression.

2.12. Effect of temperature on the shelf-life and oxidation rate of sunflower oil

In this study, the kinetic parameters for three thermal stages of SO samples were determined using the developed kinetic parameter determination program.

The effect of temperature on the rate of PV, pAV and TV was illustrated through Arrhenius Eq. (10) (Labuza and Riboh, 1982):

$$\ln K = \ln A - \frac{E_A}{RT} \qquad (10)$$

where K is the reaction rate constant, A is the pre-exponential parameter, T is the absolute temperature (K), E_A is the activation energy based on kJ/mol (E_A = slope × R) and R is the molar gas constant (8314 KJ/mol°K) where ln A and E_A are obtained from the intercepts and slopes respectively of the lines obtained by regressing ln K vs. 1/T using a least squares linear regression.

The Q_{10} (which is known as the temperature acceleration factor, is applied to predict the increase in oxidation rate by raising the value of temperature at the rate of 10 °C can be calculated using different mathematical methods and applied to predict the shelf life of samples stored at lower temperatures. For zero-order and first-order conditions, Q_{10} is calculated based on Eqs. (11) and (12), respectively (Labuza and Schmidl, 1985):

$$Q_{10} = \frac{kat(T + 10^\circ C)}{Kat T^\circ C} \qquad (11)$$

$$\log Q_{10} = \frac{0.523 E_A}{T(T + 10)} \qquad (12)$$

2.13. Statistics

The experiments were performed three times to ensure accuracy and reliability, and statistical analysis was conducted using one-way ANOVA with SPSS software v.26 (IBM Analytics, IBM, Armonk, NY, USA). The results are presented as the mean and standard deviation (SD). Differences were considered statistically significant when a p-value was less or equal to 0.05.

3. Results and discussion

3.1. Peroxide value (PV)

The changes in PVs of SOs containing different concentrations of GTE and TBHQ are illustrated in Fig. 1A. According to the obtained results, the PVs in SO samples containing TBHQ (75 ppm) showed a similar increase as samples containing 200, 400, and 800 ppm of GTE. The results also indicated the effectiveness of GTE in controlling the increase in PVs. Additionally, this increase was low during the first 5 months of the storage period. In the CODEX-STAN 210 (1999) and the Iranian national standard, INSO 1300 (2018), the specified limits for PV of refined sunflower oil are up to 10 and 5 meq of active oxygen/kg oil, respectively. Based on CODEX-STAN 210 (1999), PV in the blank sample was above codex limit after 8 months, whereas in the SOs containing TBHQ or GTE, the PV amounts were lower than 6 meqO$_2$/kg oil after 12 months. Determination of PV in docosahexaenoic acid (DHAO) samples containing different concentrations of GTE (1000, 400, and 160 ppm) and α-tocopherol (500, 200, and 80 ppm) showed that PV increased during the storage period (9 weeks at 30 °C), but SO containing 1000 ppm of GTE showed lower PV compared with other samples (Nain et al., 2021). In another study, GTE with two different content including GTE1: EGC (58 %), EGCG (30.1 %), EC (7.9 %), and ECG (3.9 %), and GTE2: EGC (17.7 %), EGCG (60.6 %), EC (9.8 %), and ECG (11.8 %) was added to DHA-rich oil at a concentration of 250 ppm and then samples were stored at 30 °C for 21 days (Nain et al., 2022). PV determination of the
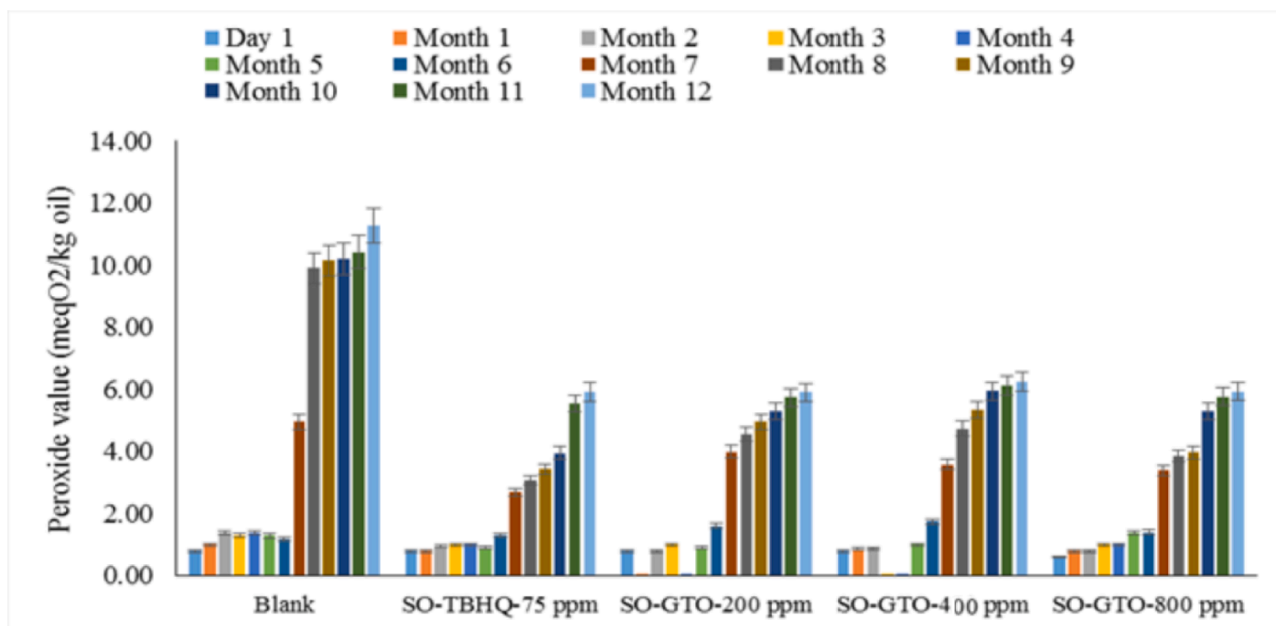
(A)
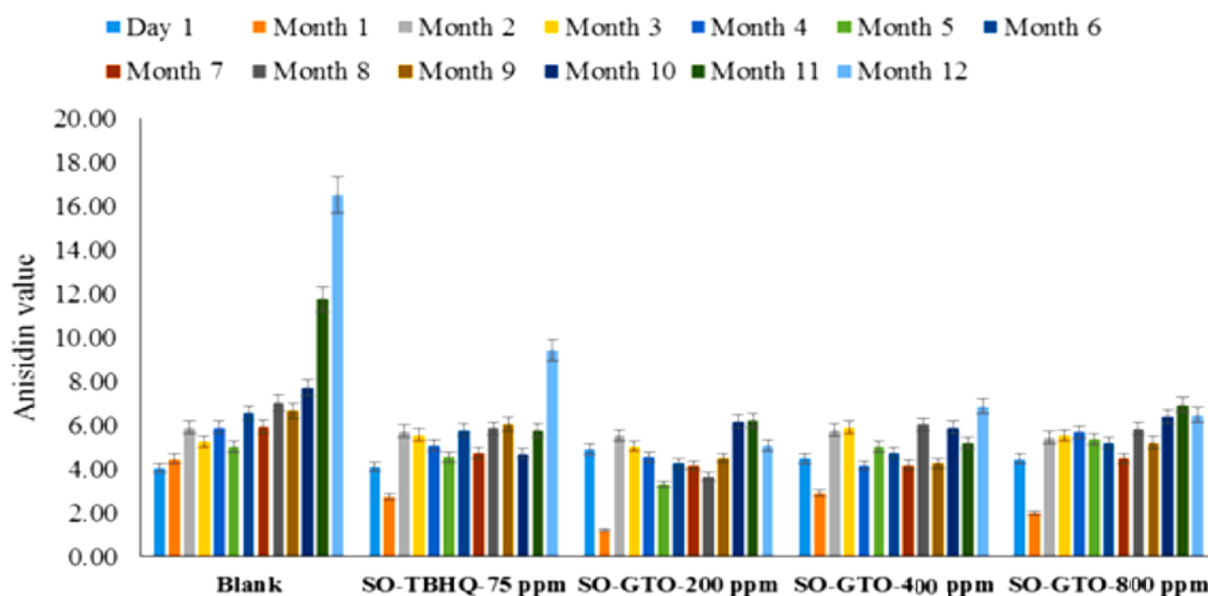
(B)

**Fig. 1.** PV (A), *p*-AV (B), TV (C), oxidative stability (D), and AV (E) of the extract.

samples (in each week) showed no significant difference between samples – treated by GTE1 or GTE2. Increase of PV during 21 days of storage was observed in all samples which DHA-rich oils containing GTE1 or GTE2 had lower increase rate of PV compared to control samples (without antioxidant) (Nain et al., 2022).

The results of the present study are in agreement with Shen et al. (2021) who compared PVs measurements in untreated canola oil and canola oil samples containing a mixture of tocopherols (alpha, beta, delta, and gamma) (300 ppm), GTE (2500 ppm and 5000 ppm), TBHQ (200 ppm), and rosemary extract (500 ppm), under open container conditions (at 24 °C and 60 °C). At room temperature, GTE at two concentrations had a similar performance to canola oil containing 200 ppm TBHQ.
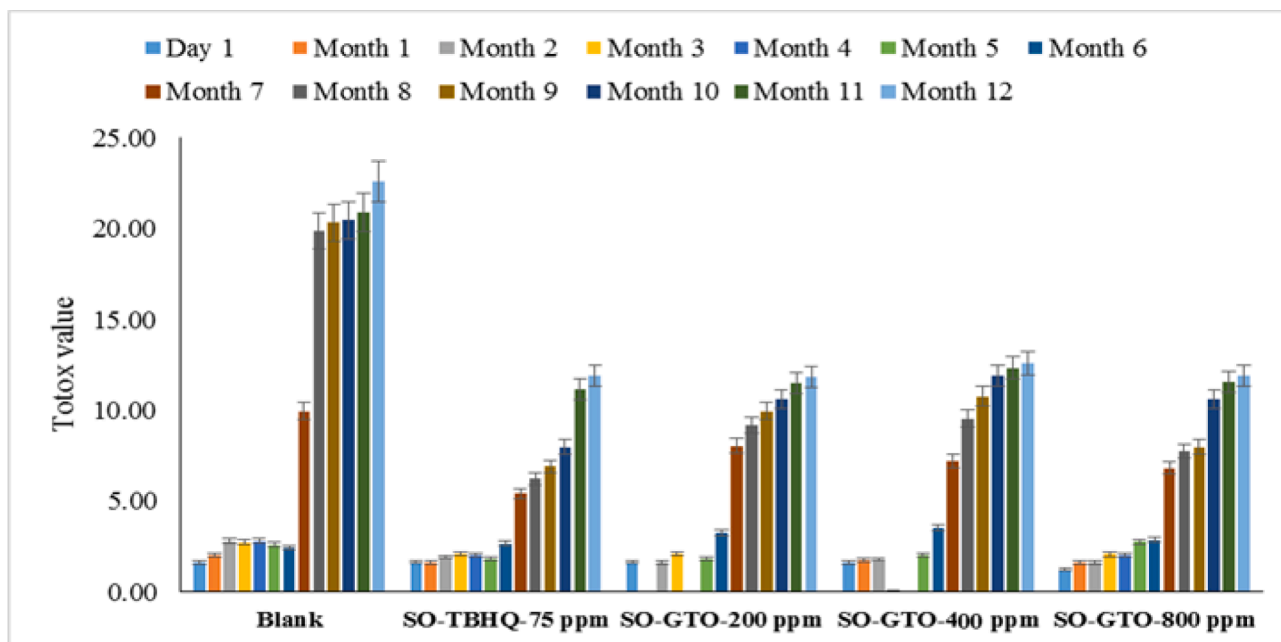
(C)
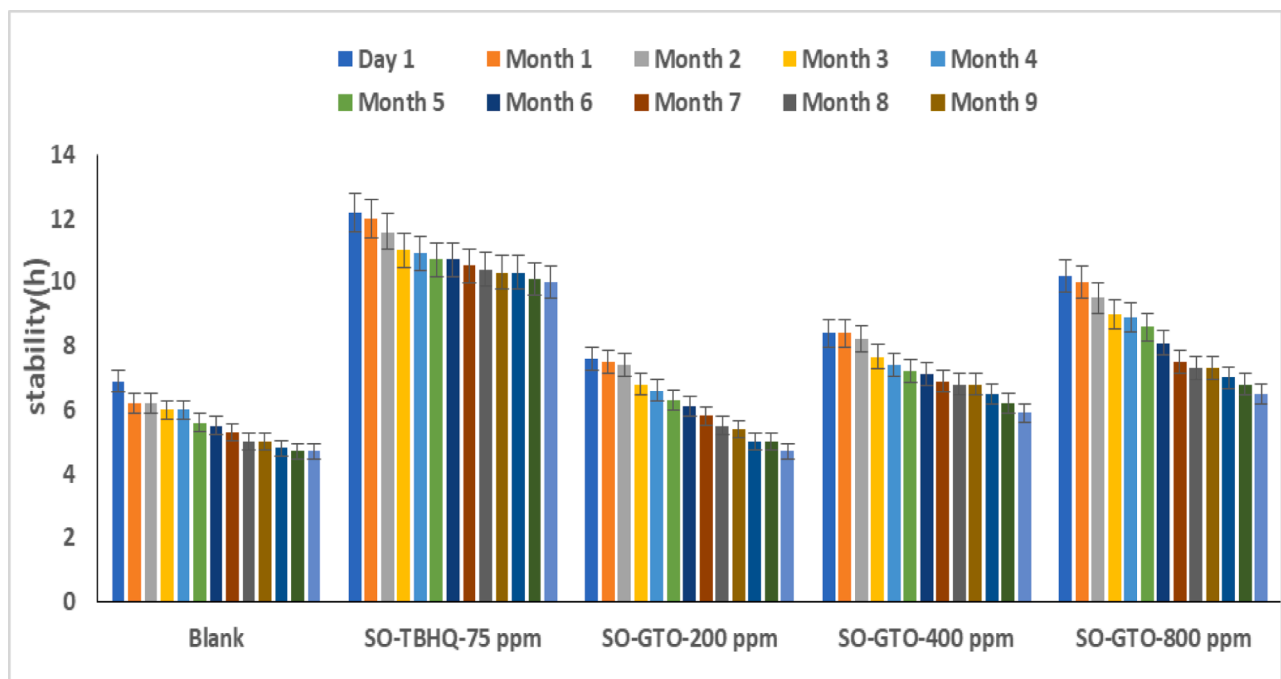
(D)

Fig. 1. (*continued*).

3.2. p-Anisidine value (pAV)

The second stage of oxidation occurs when hydroperoxides decompose and are converted to carbonyls (particularly aldehydes), which results in a rancid odor (Sheybani et al., 2023 a & b). The quantity of these compounds was determined by measuring the *p*AV. Changes in sunflower oils' *p*AV throughout the storage period are shown in Fig. 1B.

The specified limit of the Iranian national standard for *p*AV is 6.5 (INSO 1300, 2018). The control sample exhibited the highest increase in *p*AV, followed by the sample containing TBHQ (75 ppm). As shown in
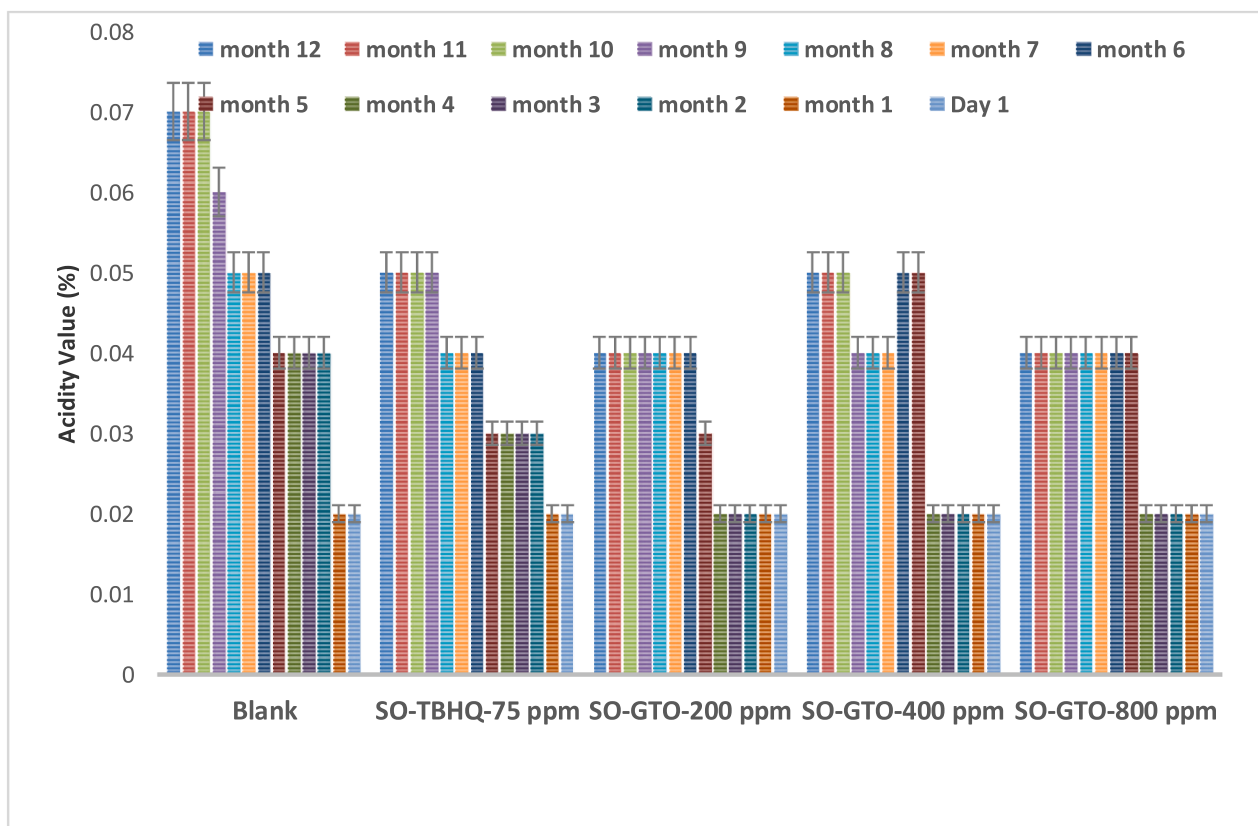
(E)

Fig. 1. (*continued*).

Fig. 1B, *p*AV increased slowly in samples containing different concentrations of GTE during one year of storage. All SOs containing different concentrations of GTE had a *p*AV of less than 6.5. The samples containing 800 ppm GTE showed lower *p*AVs than samples containing 200 ppm and 400 ppm GTE. The differences between the *p*AVs in SOs containing different concentrations of GTE were significant during the storage period ($p < 0.05$). In one study, the *p*AV of DHAO samples containing different concentrations of GTE and α-tocopherol increased during the 9-week storage at 30 °C. In the first 3 weeks of storage, *p*AV of DHAO containing GTE remained stable and lower than that in samples containing α-tocopherol (Nain et al., 2021). The addition of GTE into DHAO improved the oxidative stability whereas the formation of secondary oxidation products increased in DHAO containing α-tocopherol (Nain et al., 2021).

In another study, added GTE at concentrations of 200 ppm, 500 ppm and 1000 ppm to the refined menhaden oil (10 % of DHA) and seal blubber oil (7.5 % of DHA) samples was more effective than the sample containing α-tocopherol at concentration of 500 ppm which resulted the pro-oxidative effect of GTE (Wanasundara and Shahidi, 1997). Also, the increase rate of *p*AV in samples containing GTE was inversely dependent on the GTE concentrations in the oils. Our results are in agreement with pervious results reported by Nain et al., 2021 & 2022.

3.3. TOTOX value (TV)

TV, an index of fat oxidation, indicates the primary and secondary oxidation of oil. The TV indicates the lipid oxidation products and shows the oxidation state of the edible oils (Sheybani et al., 2023 a & b). A TV of 26 has been recommended for fish oil as a maximum limit whereas there is no limit of TV for edible oils in international standards (CODEX STAN, 2017).

The results of TVs are illustrated in Fig. 1C. According to the results, all SOs containing different concentrations of GTE and 75 ppm of TBHQ, showed higher TV than the appropriate quality criterion of oil extracted from oil seeds from the seventh month onwards. The results indicated an increase in the TV of SOs during 12-month storage (Fig. 1C). During the entire storage period, TVs for oils containing 800 ppm GTE were lower than those of the other samples. The results also indicate that the increase of TV in SOs containing 800 ppm GTE was lower than that in SOs containing 75 ppm TBHQ. The control samples exhibited a rapid increase in TV over 12 months, whereas the samples containing different concentrations of GTE showed a slower change. This increase in TVs was more quickly from the 11th and 12th months. Increase of TVs in the samples containing different concentrations of GTE is in agreement with those who investigated the effect of free-α-tocopherol and its encapsulated on oxidative stability (TV) of camelia oil during 90 days of storage (Sheybani et al., 2023b). Also, an increase rate of TV was observed in canola oil samples containing aqueous extracts of the *Bifurcaria bifurcate,* seaweeds *Ascophyllum nodosum* and *Fucus vesiculosus* during the storage (Agregán et al., 2017).

3.4. Oxidative stability

The oxidative stability of SO samples containing different concentrations of GTE (200, 400, and 800 ppm), TBHQ (75 ppm), and the control samples are presented in Fig. 1D. According to the obtained results, control samples significantly showed a lower resistance and decreased rapidly throughout keeping time. Samples treated with TBHQ also showed the highest oxidative stability, which decreased from 12.2 h to 10 h after 1 year. The oxidative stability of samples containing 800

ppm of GTE also decreased from 10.2 h to 6.5 h. The stability of every other sample that received GTE treatment significantly decreased as well. Yin et al. (2012) used the Rancimat test to examine the impact of GTE on the oxidative stability of sunflower oil; the study's findings indicated that the induction time of SO with varying GTE concentrations was enhanced by the increase in GTE concentration in the samples. In one study, induction period (IP) for a shortening used for making pastry cakes was 13.36 h but after baking, the longest IP was observed in the sample containing BHA (48.01 h) which decreased to 24.10 h during 28 days of storage. The IP in the samples containing different concentrations of GTE, 0.02, 0.2 and 1 % was 25.67 h, 26.93 h and 36.9 h which reduced to 18.39 h, 19.1 h, and 21.92 h after storage, respectively (Żbikowska et al., 2017).

3.5. Acidity value (AV)

A key indicator of food rancidity is the measurement of free fatty acids (FFAs). Triglycerides hydrolyze as a result of an enhanced interaction between moisture and oil, which enhances the creation of FFAs (Kariminejad et al., 2023). The AV modifications are shown in Fig. 1E. After a year of storage, the data showed a non-significant increase in AV. Furthermore, the findings demonstrated that, throughout the storage, the AVs in every sample were below the maximum allowable quantity for refined sunflower oil (1.0 % w/w oleic acid) (INSO 1300, 2018). The control samples showed the largest increase in AV. The samples with 800 ppm of GTE had lower AVs than the other samples, as seen in Fig. 1E.

Increase of AV was observed in fat extracted from the sponge -fat cakes containing GTE (0.02 %, 0.2 % and 1 %) and BHA (0.02 %) through the sampling in 7, 14, 21 and 28 days compared to control sample but results showed that GTE kept the hydrolysis level in fat content of cakes during storage (Kozłowska et al., 2019).

3.6. Antioxidant activity

The antioxidant activity of oil samples throughout 12 months is shown in Fig. 2A. The results obtained indicate that the antioxidant activity of sunflower oil samples was higher when synthetic or natural antioxidants were added, as compared to the control oil. The antioxidant activity of the samples was reduced when the samples were stored at room temperature for a period of six to twelve months. Furthermore, the refined SO sample's antioxidant activity was also 73 %, suggesting that the basic sample contained antioxidant-producing substances such as tocopherols. Butter samples containing GTE (2–10 % (w/w)) exhibited higher antioxidant activity (7.27 % to 13.94 %) than control butter (nd) (no additive) (Thakaeng et al., 2020). Also, a significant increase of antioxidant activity percentage was observed with increase of GTE concentration in the samples. In addition, a high antioxidant activity percentage was reported for GTE by Al-Ghafari et al., (2016). In one study, the addition of green tea powder (GTP) enhanced the antioxidant activity percentage in pan bread sample (Ning et al., 2017). It was found that antioxidant activity of GTE depends on the amount of catechins content. It was reported that the antioxidant activity percentage was 81.83 ± 0.24 which was higher than ascorbic acid (76.62 ± 0.35) or BHA (59.91 ± 0.42) (Mildner-Szkudlarz et al., 2009).

3.7. Total phenol content (TPC)

The total phenol content results are illustrated in Fig. 2B. Higher TPC was the outcome of increasing the GTE concentration. The control sample had the least quantity of TPC, as seen in Fig. 2B. Samples of SO with 800 ppm of GTE had the largest quantity of TPC, whereas samples with 75 ppm of TBHQ and oil devoid of antioxidants had the lowest amount. TPC dropped as storage time increased in all samples of SO. According to some research, GTE polyphenolic chemicals assist prevent lipid oxidation in edible oils by having unique color, taste, and aroma as well as antioxidant qualities (Chen & Chan, 1996). Additionally, GT polyphenols' antioxidant activity is connected to the OH number linked to the ring molecule of polyphenols (Ghreishi et al., 2023). In one study, TPC of GTE was estimated 136.54 ± 1.91 mg GAE/g dry starting material (Mildner-Szkudlarz et al., 2009). The butter samples containing GTE (2–10 % (w/w)) showed a very low TPC (0.01 to 0.1 GAE % w/w db) which might be related to the degradation or oxidation of polyphenol content of GTE (Thakaeng et al., 2020).

3.8. Schaal oven test

3.8.1. PV

When compared to samples that included GTE or TBHQ, the control samples showed the highest PV during the storage period. Samples having 800 ppm GTE and 75 ppm TBHQ had the lowest PV (Table 1). The much stronger antioxidant effects of GTE were evidenced by the reduced PV in SO containing natural antioxidants when compared to the control. The presence of phenolic chemicals, which can neutralize free radicals by giving hydrogen atoms or electrons, may be connected to the antioxidant properties of GTE. This result is consistent with the research conducted by Womeni et al. (2016), which demonstrated a comparable activity in palm olein enhanced with green tea leaf extract during high-temperature storage. The authors showed that the oil supplemented with black tea leaf extract exhibited lower PVs compared to the control during storage at 55 °C and 140 °C.

Similar activities were also observed with methanolic extracts from other plants in soybean and sunflower oils (Pietta, 2000). The results of the present study are paralleled with Shen et al. (2021) who compared PVs quantities in a blank canola oil and canola oil samples containing 300 ppm of mixed tocopherols, GTE (2500 ppm and 5000 ppm), TBHQ (200 ppm), and rosemary extract (500 ppm), under open container conditions at 60 °C. The authors indicated that the PVs in untreated canola oil and canola oil containing mixed tocopherols had similar rates of shelf life increase at 60 °C for up to 21 days. However, at 60 °C, neither of the two concentrations of GTE had a behavior like canola oil containing TBHQ (Shen et al., 2021).

The effect of GTE on the oxidation of menhaden oil (MHO) and seal blubber oil (SBO) in the forms of refined, bleached, and deodorized was investigated by a Schaal oven test at 65 °C. The authors found that GTE indicated a pro-oxidant effect in SBO and MHO which was related to the chlorophyll content of GTE (Wanasundara, and Shahidi, 1997). Therefore, they applied the dechlorophyllized GTE (DGTE) in both oils which PV determination of the samples containing more than 200 ppm of DGTE showed a significant decrease under the accelerated oxidation condition.

3.8.2. pAV

According to Table 1, the control sample had the highest *p*AV, which was significantly higher than the *p*AVs of the oil samples that contained TBHQ and GTE. Between days 0 and 15, there were no significant differences observed among samples *p*AVs. Between days 0 and 15, no discernible variations were found in the *p*AV samples. The SO sample containing 75 ppm TBHQ showed a significant increase in *p*AV on the 60th day of storage, while the other stabilized samples' anisidine values stayed unchanged. These results suggest that the natural extracts are more effective in delaying the formation of secondary oxidation products in SO. This activity might be related to the presence of various phenolic antioxidants identified in the extract, as previous studies have demonstrated the ability of these compounds to delay the formation of secondary oxidation products in edible oils (Sheybani et al., 2023 a,b).

*P*AV measurement in lipid fractions extracted from sponge-fat cakes containing GTE (0.02 %, 0.2 % and 1 %) and BHA (0.02 %), after baking and 28 days of storage, showed a gradual increase of *p*AV in the samples compared to the samples without any additives. Samples containing 1 % of GTE and 0.02 % of BHA showed the lowest *p*AV (Kozłowska et al., 2019).
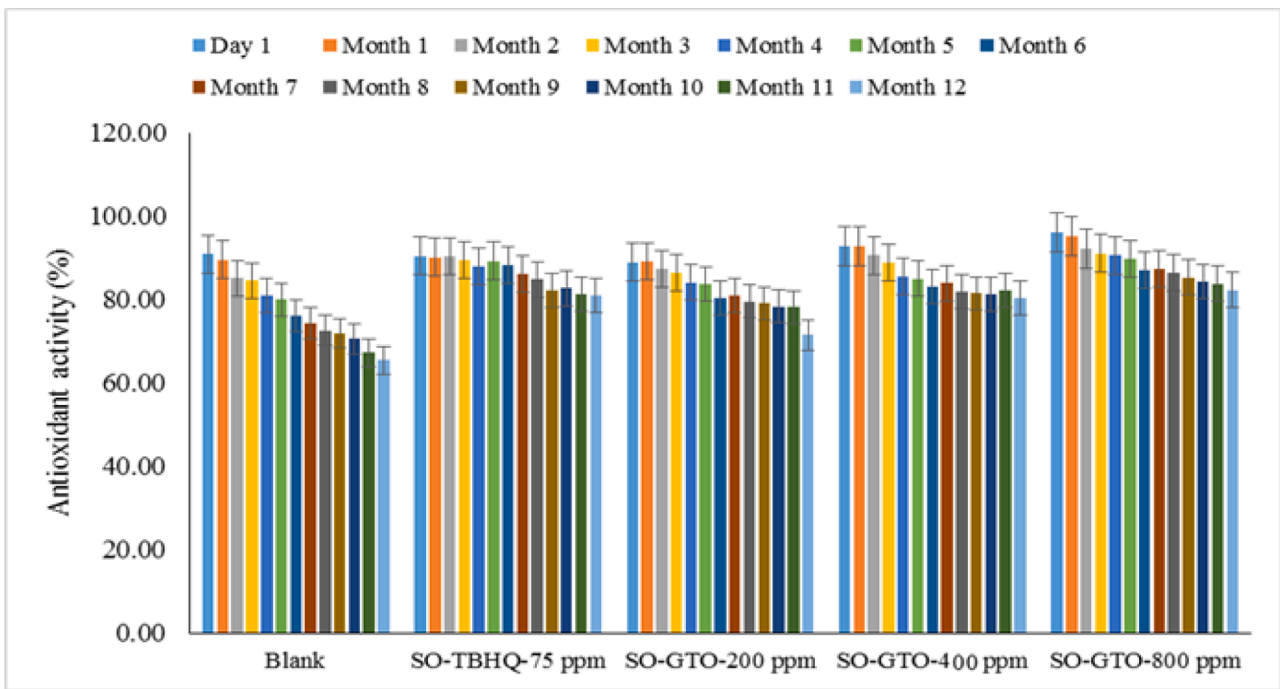
(A)
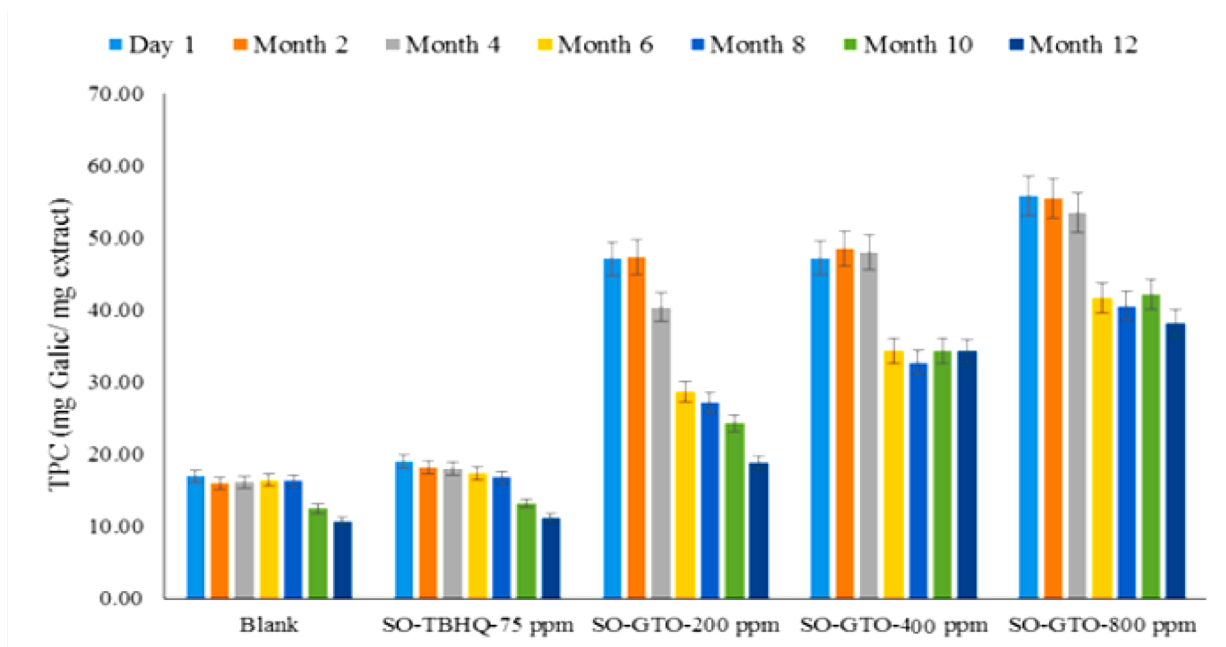
(B)

Fig. 2. DPPH radical scavenging activity (A) and TPC (B) of the extract.

Table 1
Changes in peroxide value, *p*-anisidine value, TOTOX value, oxidative stability, acidity value, antioxidant activity, and total phenol content during 60 days of storage at 60 °C.

Characteristic	Samples	Storage time (days)			
		0	15	30	60
PV (meqO2/kg)	Control	0.79 ± 0.10 ^{Da}	3.51 ± 0.12 ^{Ca}	3.96 ± 0.13 ^{Ba}	6.37 ± 0.21 ^{Aa}
	SO + TBHQ$_{75}$ ppm	0.79 ± 0.07 ^{Da}	2.58 ± 0.15 ^{Cc}	2.92 ± 0.11 ^{Bb}	5.39 ± 0.22 ^{Ac}
	SO + GTE$_{200}$ ppm	0.79 ± 0.07 ^{Da}	2.79 ± 0.16 ^{Cb}	2.99 ± 0.13 ^{Bb}	5.98 ± 0.20 ^{Ab}
	SO + GTE$_{400}$ ppm	0.79 ± 0.08 ^{Ca}	2.59 ± 0.10 ^{Bc}	2.70 ± 0.11 ^{Bc}	5.96 ± 0.15 ^{Ab}
	SO + GTE$_{800}$ ppm	0.79 ± 0.05 ^{Da}	2.18 ± 0.12 ^{Cd}	2.66 ± 0.10 ^{Bc}	4.99 ± 0.10 ^{Ad}
p-AV	Control	4.06 ± 0.10 ^{Dc}	4.49 ± 0.15 ^{Cd}	6.89 ± 0.24 ^{Ba}	11.49 ± 0.35 ^{Aa}
	SO + TBHQ$_{75}$ ppm	4.09 ± 0.19 ^{Dc}	5.03 ± 0.11 ^{Cb}	5.77 ± 0.15 ^{Bb}	7.46 ± 0.20 ^{Ab}
	SO + GTEGTE$_{200}$ ppm	4.91 ± 0.07 ^{Ca}	5.58 ± 0.14 ^{Ba}	5.63 ± 0.20 ^{Bc}	5.78 ± 0.24 ^{Ac}
	SO + GTE$_{400}$ ppm	4.49 ± 0.08 ^{Cb}	4.81 ± 0.10 ^{Bc}	5.31 ± 0.15 ^{Bd}	5.40 ± 0.20 ^{Ad}
	SO + GTE$_{800}$ ppm	4.46 ± 0.05 ^{Cb}	4.89 ± 0.12 ^{Bb}	4.97 ± 0.13 ^{Ae}	5.03 ± 0.12 ^{Ae}
TV	Control	8.12 ± 0.20 ^{Da}	11.51 ± 0.27 ^{Ca}	16.81 ± 0.35 ^{Ba}	24.23 ± 0.56 ^{Aa}
	SO + TBHQ$_{75}$ ppm	5.63 ± 0.26 ^{Dd}	10.87 ± 0.26 ^{Cb}	13.89 ± 0.26 ^{Bc}	17.00 ± 0.42 ^{Ad}
	SO + GTE$_{200}$ ppm	6.40 ± 0.14 ^{Db}	10.56 ± 0.28 ^{Cc}	15.21 ± 0.26 ^{Bb}	19.42 ± 0.44 ^{Ab}
	SO + GTE$_{400}$ ppm	6.07 ± 0.16 ^{Dc}	9.99 ± 0.20 ^{Cd}	15.24 ± 0.26 ^{Bb}	19.06 ± 0.22 ^{Ac}
	SO + GTE$_{800}$ ppm	5.64 ± 0.10 ^{Dd}	9.33 ± 0.24 ^{Ce}	13.63 ± 0.23 ^{Bc}	16.77 ± 0.20 ^{Ae}
Oxidative stability (h)	Control	6.90 ± 0.10 ^{Ae}	5.60 ± 0.13 ^{Be}	5.40 ± 0.10 ^{Ce}	5.30 ± 0.18 ^{Ce}
	SO + TBHQ$_{75}$ ppm	12.00 ± 0.15 ^{Ab}	10.20 ± 0.16 ^{Ba}	8.40 ± 0.10 ^{Cb}	6.40 ± 0.20 ^{Dd}
	SO + GTE$_{200}$ ppm	8.60 ± 0.18 ^{Ad}	8.40 ± 0.15 ^{Bd}	8.10 ± 0.10 ^{Cc}	6.50 ± 0.20 ^{Dc}
	SO + GTE$_{400}$ ppm	10.20 ± 0.25 ^{Ac}	8.50 ± 0.15 ^{Bc}	8.00 ± 0.20 ^{Cd}	7.64 ± 0.15 ^{Db}
	SO + GTE$_{800}$ ppm	12.20 ± 0.18 ^{Aa}	9.50 ± 0.10 ^{Bb}	10.00 ± 0.12 ^{Ca}	8.90 ± 0.12 ^{Da}
AV (%)	Control	0.04 ± 0.01 ^{Da}	0.08 ± 0.01 ^{Ca}	0.12 ± 0.00 ^{Ba}	0.18 ± 0.01 ^{Aa}
	SO + TBHQ$_{75}$ ppm	0.03 ± 0.01 ^{Db}	0.04 ± 0.01 ^{Cc}	0.05 ± 0.00 ^{Bc}	0.10 ± 0.01 ^{Ad}
	SO + GTE$_{200}$ ppm	0.02 ± 0.01 ^{Dc}	0.05 ± 0.01 ^{Cb}	0.08 ± 0.00 ^{Bb}	0.14 ± 0.01 ^{Ab}
	SO + GTE$_{400}$ ppm	0.02 ± 0.01 ^{Dc}	0.03 ± 0.01 ^{Cd}	0.05 ± 0.00 ^{Bc}	0.12 ± 0.01 ^{Ac}
	SO + GTE$_{800}$ ppm	0.02 ± 0.01 ^{Cc}	0.02 ± 0.01 ^{Ce}	0.04 ± 0.01 ^{Bd}	0.09 ± 0.01 ^{Ae}
Antioxidant activity (%)	Control	73.59 ± 0.00 ^{Ae}	69.46 ± 0.01 ^{Be}	62.04 ± 0.01 ^{Ce}	53.93 ± 0.00 ^{De}
	SO + TBHQ$_{75}$ ppm	91.80 ± 0.02 ^{Ac}	88.32 ± 0.01 ^{Bc}	81.15 ± 0.01 ^{Cc}	77.98 ± 0.00 ^{Dc}
	SO + GTE$_{200}$ ppm	86.41 ± 0.03 ^{Ad}	77.35 ± 0.00 ^{Bd}	72.17 ± 0.01 ^{Cd}	56.61 ± 0.00 ^{Dd}
	SO + GTE$_{400}$ ppm	96.79 ± 0.00 ^{Ab}	90.23 ± 0.01 ^{Bb}	86.83 ± 0.01 ^{Cb}	81.81 ± 0.00 ^{Db}

Table 1 (*continued*)

Characteristic	Samples	Storage time (days)			
		0	15	30	60
	SO + GTE$_{800}$ ppm	98.54 ± 0.00 ^{Aa}	92.52 ± 0.00 ^{Ba}	88.12 ± 0.01 ^{Ca}	83.87 ± 0.00 ^{Da}
TPC (mgGA/Kg Oil)	Control	16.94 ± 0.02 ^{Ae}	13.27 ± 0.02 ^{Be}	11.93 ± 0.02 ^{Ce}	11.61 ± 0.02 ^{De}
	SO + TBHQ$_{75}$ ppm	19.05 ± 0.02 ^{Ad}	17.65 ± 0.02 ^{Bd}	15.75 ± 0.02 ^{Cd}	14.05 ± 0.02 ^{Dd}
	SO + GTE$_{200}$ ppm	47.11 ± 0.02 ^{Ac}	46.02 ± 0.02 ^{Bc}	41.01 ± 0.02 ^{Cc}	31.37 ± 0.02 ^{Dc}
	SO + GTE$_{400}$ ppm	49.18 ± 0.02 ^{Ab}	47.40 ± 0.02 ^{Bb}	44.81 ± 0.02 ^{Cb}	36.94 ± 0.02 ^{Db}
	SO + GTE$_{800}$ ppm	56.79 ± 0.02 ^{Aa}	54.05 ± 0.02 ^{Ba}	50.03 ± 0.02 ^{Ca}	45.22 ± 0.02 ^{Da}

*The various letters in each column show a statistically significant difference ($P < 0.05$).

3.8.3. TV

Every sample's TV showed a significant increase in storage duration (Table 1). The control samples showed the highest total oxidation like PVs and *p*AVs. In contrast, oxidation levels were significantly lower in sunflower oil samples that contained TBHQ and GTE. As shown in Table 1, demonstrates that there was no significant difference in TVs between the TBHQ and SO samples enhanced with GTE, indicating that the GTE was successful in preventing SO from oxidizing while in storage. The extract's reported protective effect may have been caused by GTE's antioxidant components. These findings are in agreement with the results reported by Nyam et al. (2013), who demonstrated that kenaf and roselle extracts, at 1500 ppm, efficiently decreased the total oxidation of SO during an accelerated storage period of 24 days at 65 °C. In one study, the lipid extracted from the sponge -fat cakes containing GTE (0.02, 0.2, and 1 %) and BHA (0.02 %) showed the lowest amount of TV compared to control sample (without antioxidant) during 28 days of storage at 63 °C (Kozlowska et al. 2019). According to the results, GTE indicated protection effect on hydropreoxides formation which is in agreement with those of Mildner-Szkudlarz et al., (2009). They determined TV for biscuits containing BHA (0.02 %) and GTE at different levels of 0.02 %, 0.1 % and 1 % during 20 days of storage at 60 °C. Their results showed that biscuits containing GTE (1 %) were more efficient in delaying lipid oxidation than samples having BHA (0.02 %).

3.8.4. Oxidative stability

The results of the Rancimat analysis indicated that GTE had a significant impact on oil oxidative stability. The results of the rancimat test showed that the lowest and highest induction times were for the control and GTE samples (800 ppm), respectively (Table 1). Treated samples with 800 ppm GTE demonstrated greater oxidative stability than those containing TBHQ after 60 days of storage. Yin et al. (2012) used a Rancimat test to evaluate the effects of GTE on SO oxidative stability. The authors indicated that by increasing the antioxidant levels, the induction period of oil samples was increased. They showed that the induction periods for control and sample oils containing GTE at 0.25, 0.5, and 1 mol/g oil concentrations were approximately 1.9, 4.8, 7.5, and 11.8 h, respectively. Gramza et al. (2006) evaluated the antioxidant properties of aqueous and ethanol GTEs at concentrations of 200, 500, and 1000 mg/L on the oxidative stability of SOs in different investigations. According to their findings, the oil samples with the longest induction times were those that contained 1000 mg/L of ethanolic GTEs.

3.8.5. Acidity value AV

The results indicated that AV of all samples increased during the

storage period (Table 1). The control sample significantly had the highest AV (0.18 %) on day 60 ($p < 0.05$), while samples containing 800 ppm GTE and 75 ppm TBHQ had the lowest acidity values (0.10 and 0.09 %, respectively) on the same day. These findings indicated the high antioxidant attributes of GTE. At the beginning of storage, there was no significant increase in acidity value (ranging from 0.02 % to 0.06 %). However, the AV in the treated samples increased to a range between 0.04 % and 0.12 %, after 15 to 30 days. According to the results shown in Table 1, the samples containing 800 ppm GTE had lower levels of AV than the other samples, with values of 0.02 %, 0.04 %, and 0.09 % on days 0, 15, 30, and 60 respectively, which were similar to the AV levels

of the reference sample (TBHQ), which were 0.03 %, 0.03 %, 0.05 %, and 0.10 % on days 0, 15, 30, and 60. These findings indicated that GTE can effectively enhance the shelf life of oils and lipids by inhibiting acidity value. Iqbal and Bhanger (2007) found that garlic extracts exhibited antioxidant activity in SO, resulting in lower or equal AV when compared to the synthetic antioxidant BHT. Consequently, the current investigation shows that GTE has antioxidant activity equivalent to that of synthetic antioxidants, making it a useful natural antioxidant source for oils and fats.
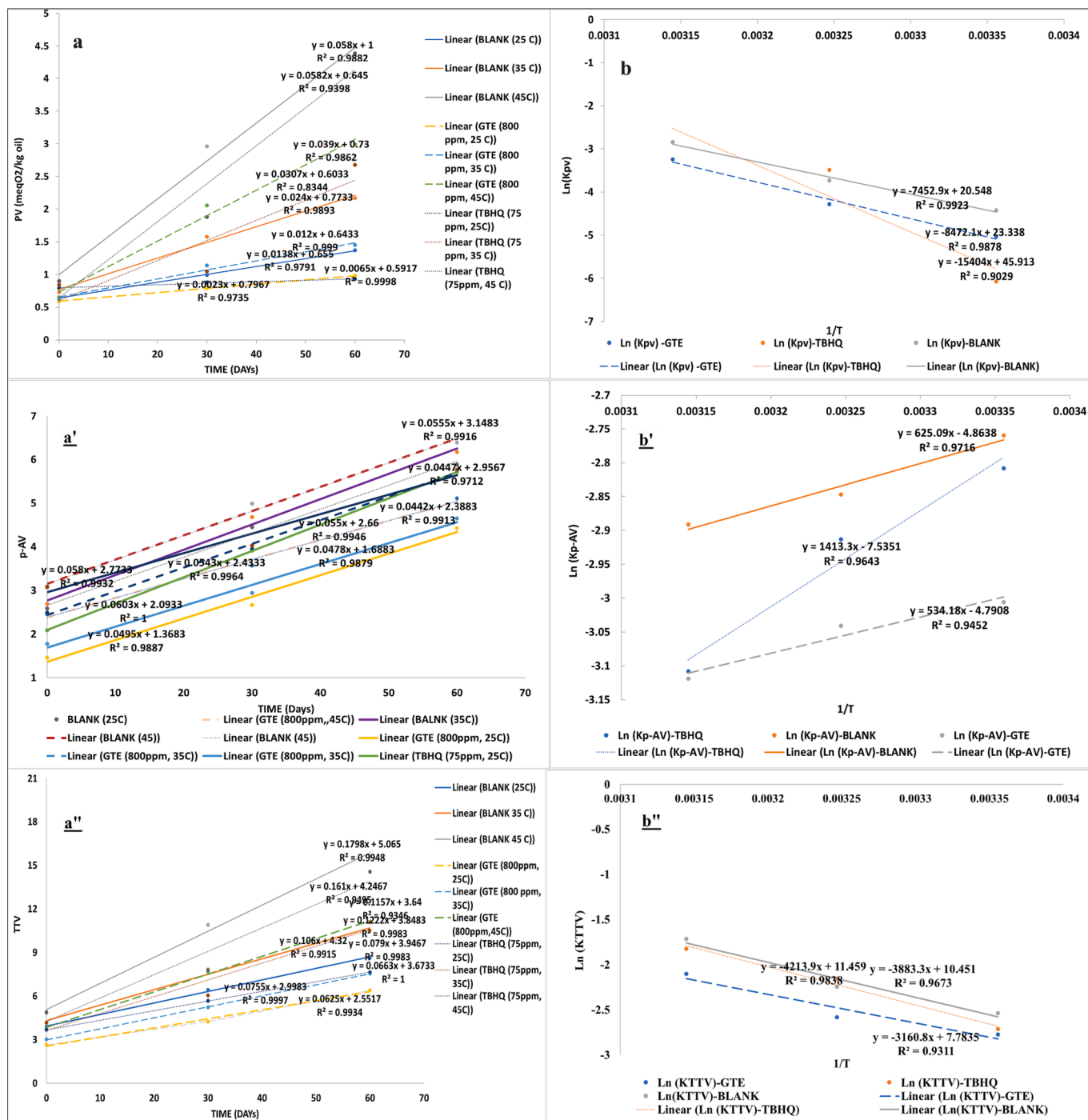

Fig. 3. Formation of hyperoxides, aldehydes, and total oxidation into the control and the sunflower oil samples containing GTE (800 ppm) and TBHQ (75 ppm) at 25, 35, and 45 °C (a, a', a") and relationship between the constant rate and temperature that was obtained from kinetic models of first order in at 25, 35, and 45 °C (b, b', b").

3.8.6. Antioxidant activity

As shown in Table 1, the antioxidant activity of all samples was decreased during storage at 60 °C. The antioxidant activity of treated samples with 800 ppm and 400 ppm GTE decreased similarly. The control and samples with 800 ppm GTE had the lowest and highest levels of antioxidant activity, respectively. Gramza et al., (2006) studied the antioxidant properties of heated SO samples at 110 °C containing ethanol extract or water extract of green and black tea leaves which the highest antioxidant activity was obtained in the sample containing 1000 ppm of ethanolic GTE and the other concentrations (below 1000 ppm) showed clearly less activity. Also, the important impact of the catechin content was observed in the determined antioxidant activity of the heated SO. In one study, the antioxidant activity in the MHO and SBO samples containing DGTE, BHA, BHT and α-tocopherol (stored at 65 °C) was determined which the excellent antioxidant activity was observed in oil samples containing DGTE in concentrations of upper 200 ppm (Wanasundara, and Shahidi, 1997).

3.8.7. Total phenol content (TPC)

The results of TPC in SO samples are shown in Table 1. The sample containing 800 ppm of GTE exhibited the highest TPC, while the control sample had the lowest TPC, followed by the sample containing 75 ppm of TBHQ. It was observed that TPC was significantly decreased during 60 days of storage. During 60 days of storage, TPC was decreased for the samples containing 800 ppm of GTE from 56.79 to 45.22 (mgGA/Kg Oil) for the samples containing TBHQ from 19.05 to 14.05 (mgGA/Kg Oil), and for blank samples from 16.94 to 11.61 (mgGA/Kg Oil). According to the results obtained from determination of antioxidant activity and TPC in the samples for one year and also for the oven test, the positive correlation of TPC and antioxidant activity was obtained which agreed with reports by Zhang et al., (2023).

3.9. Shelf life of the SO

According to the obtained results, 800 ppm of GTE was the optimized concentration and was taken into account for measuring the shelf-life of SO. Also, the shelf life of the control sample and treated samples with TBHQ (75 ppm) was determined. Fig. 3a, 3a', and 3a'' indicate the formation of hyperoxides (primary oxidation), aldehydes (secondary oxidation), and total oxidation, respectively at temperatures 25 °C, 35 °C, and 45 °C. The results showed that the formation of hyperoxides was enhanced with the increase in temperature. In the samples containing antioxidants, lipid peroxidation was low which might be related to the action of antioxidants for quenching most of the produced free radicals. For estimation of the shelf life of SO samples containing GTE (800 ppm), TBHQ (75 ppm), and control, PV, pAV, and TV were determined at different temperatures to reach the kinetic models that fit better with obtained experimental data. Almost all the reactions related to loss of food quality have been categorized as zero, first, and second order which are dependent on matrix and temperature. Based on the coefficient of linear regression (r^2), the kinetic model corresponded to the first order for PV, TV, and pAV for all samples in different temperatures. Therefore, the oxidation rate of SOs was fitted with a first order, Eqs. (13)–(15)) for all treatments (Labuza and Riboh, 1982).

$$Ln(PV) - Ln(PV_0) = K_{PV} \times t \tag{13}$$

$$Ln(pAV) - Ln(pAV_0) = K_{AV} \times t \tag{14}$$

$$Ln(TV) - Ln(TV_0) = K_{TV} \times t \tag{15}$$

where K_{PV}, K_{AV}, and K_{TV} are the rate constants for hyperoxides (meqO$_2$/kg oil-day), aldehydes, and total aldehydes and hyperoxides compounds formation, respectively; t is the reaction time, and PV_0, AV_0, and TV_0 are the peroxide, anisidine and total oxidation values, respectively at first order reaction.

Fig. 3b, 3b', and 3b'' indicate the relationship between constant rate and temperature which were drawn based on Arrhenius plot, Ln (K_{PV}), Ln (K_{pAV}) or Ln (K_{TV}) Vs 1/T, and the values of K_{PV}, K_{pAV}, and K_{TV} were obtained from kinetic models of the first order in each temperature. Table 2 indicates the coefficient of linear regression (r^2), activation energy (E_A), the rate constant (K), shelf life and Q_{10} of samples at 25 °C. Shelf life was estimated based on the specific limitations in the national standard of Iran (No. 1300) which are 5 meqO$_2$/kg of oil for PV and 6.5 for pAV (INSO 1300, 2018). Also, the estimation of the shelf life of sunflower oil was performed based on the specified limitation for fish oils which was 26 in the codex standard for fish oils (2017). There is no limit for TV in the SO national or international standards. It was obtained that shelf-life values calculated based on PV were higher than pAV due to the intended limitation (6.5) in the national standard of Iran and also TV with a limitation of 26. The r_2 value close to 1 indicated that the Arrhenius model is well-fitted to the resulting experimental data. The Q_{10} results showed that the length of shelf life based on PV was higher than those calculated based on TV and pAV. The shelf life of control and sunflower oil containing TBHQ were lower than those obtained for sunflower oils containing 800 ppm of GTE when shelf life was calculated by pAV (6.5) and TV (26).

4. Conclusions

This study investigated the substitution of TBHQ in sunflower oil with the optimum concentration of GTE. We of course realize that using GTE in sunflower oil production may incur extra costs, but the health benefits of natural antioxidants outweigh the ensuring cost increment. The results indicated that 800 ppm of GTE is a suitable substitute for TBHQ (75 ppm). Also, sunflower oil containing 800 ppm of GTE had a longer shelf life than both the control sample and the oil samples containing TBHQ, which was obtained via parameters of pAV and TV. However, the calculated shelf life using PV parameter was longer for TBHQ compared with other samples. The estimated shelf life based on PV was obtained longer than when using TV or pAV. In this study, GTE has demonstrated potential as a natural alternative to synthetic antioxidants.

CRediT authorship contribution statement

Nadia Ahmadi: Writing – original draft, Formal analysis. **Mehrdad Ghavami:** Writing – review & editing. **Ladan Rashidi:** Writing – review & editing, Supervision, Methodology. **Maryam Gharachorloo:** Writing – review & editing. **Leila Nateghi:** Writing – review & editing, Software, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence

Table 2
Coefficient of linear regression (r^2), activation energy (E_A), the rate constant (K), shelf life and Q10 of samples at 25 °C.

Parameter	Treatment	R$_2$	E$_A$ (KJ/mol°K)	K$_{25°C}$	Shelf-Life (days)	Q$_{10}$
PV	BLANK	0.992	61.96	0.012	178.09	1.0008
PV	SO-GTE	0.988	70.44	0.0065	328.78	1.0009
PV	SO-TBHQ	0.903	114.89	0.0023	929	1.0010
p-AV	BLANK	0.972	6.10	0.055	16.72	1.0001
p-AV	SO-GTE	0.945	25.94	0.0495	30.17	1.0003
p-AV	SO-TBHQ	0.964	14.71	0.0603	18.82	1.0001
TV	BLANK	0.967	32.287	0.0790	23.34	1.0004
TV	SO-GTE	0.931	26.279	0.0625	32.73	1.0003
TV	SO-TBHQ	0.984	35.034	0.0663	31.47	1.0004

the work reported in this paper.

Data availability

The data that has been used is confidential.

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Author's contribution

Nadia Ahmadi performed tests and wrote the manuscript.
Ladan Rashdi revised the draft, and she is the corresponding author.
Mehrdad Ghavami reviewed and edited the manuscript.
Maryam Gharachorloo reviewed and edited the manuscript.
Leila Nateghi performed statistical analysis and reviewed the manuscript.

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