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Retraction

Retracted: Improving the Oxidation Stability and Shelf-Life of Peanut Oil by Addition of Rosemary Extract Combined with Vitamin C and Ascorbyl Palmitate

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This article has been retracted by Hindawi following an investigation undertaken by the publisher [1]. This investigation has uncovered evidence of one or more of the following indicators of systematic manipulation of the publication process:

(1) Discrepancies in scope

(2) Discrepancies in the description of the research reported

(3) Discrepancies between the availability of data and the research described

(4) Inappropriate citations

(5) Incoherent, meaningless and/or irrelevant content included in the article

(6) Manipulated or compromised peer review

The presence of these indicators undermines our confidence in the integrity of the article's content and we cannot, therefore, vouch for its reliability. Please note that this notice is intended solely to alert readers that the content of this article is unreliable. We have not investigated whether authors were aware of or involved in the systematic manipulation of the publication process.

Wiley and Hindawi regrets that the usual quality checks did not identify these issues before publication and have since put additional measures in place to safeguard research integrity.

We wish to credit our own Research Integrity and Research Publishing teams and anonymous and named external researchers and research integrity experts for contributing to this investigation.

The corresponding author, as the representative of all authors, has been given the opportunity to register their agreement or disagreement to this retraction. We have kept a record of any response received.

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[1] Y. Wang, H. Deng, D. Huang, and X. Zeng, "Improving the Oxidation Stability and Shelf-Life of Peanut Oil by Addition of Rosemary Extract Combined with Vitamin C and Ascorbyl Palmitate," *Journal of Food Quality*, vol. 2022, Article ID 7229412, 7 pages, 2022.

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Research Article

Improving the Oxidation Stability and Shelf-Life of Peanut Oil by Addition of Rosemary Extract Combined with Vitamin C and Ascorbyl Palmitate

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Rosemary extracts are natural antioxidants, which can be considered an alternative for synthetic antioxidants in the food industry. The aim of the present study was to evaluate the oxidation stability and shelf-life of rosemary extracts combined with vitamin C (VC) and ascorbyl palmitate (AP) in peanut oil stored at 65°C. Peanut oil with tertbutyl hydroquinone (TBHQ) and without additives served as positive and negative controls, respectively. The peroxide value (POV), thiobarbituric acid reactant (TBARs), conjugated diene (CD), and conjugated triene (CT) values of the peanut oil samples were evaluated during accelerated storage every 48 h. Among them, 0.23 g/kg rosemary extracts combined with 0.13 g/kg VC and 0.07 mg/kg AP exhibited the best oxidative stability. Additionally, the oxidation kinetics model predicated that the rosemary extracts combined with VC and AP could effectively prolong the shelf-life of peanut oil. In accelerated storage, the rosemary extracts combined with VC and AP not only inhibited peanut oil oxidation like chemical antioxidants, but also were safer than chemical antioxidants. Therefore, the rosemary extracts combined with VC and AP were an effective alternative to chemical antioxidants, which could improve the oxidation stability and shelf-life of peanut oil.

1. Introduction

Unsaturated fatty acids, especially polyunsaturated fatty acids, are widely present in peanut oil, which is the main nonoxidizing natural edible oil in most parts of China [1]. However, peanut oil is more easily oxidized, resulting in a decrease in the taste and nutritional quality of the edible oil, and even the production of substances harmful to human health [2]. The addition of antioxidants is the most feasible way to control the oxidative degradation and oxidation stability of fats in peanut oil and to extend their shelf-life [3]. Chemically synthesized antioxidants (butylated hydroxytoluene (BHT) butylated, butylated hydroxyanisole (BHA), and tertbutyl hydroquinone (TBHQ) et al.) can inhibit the oxidation of edible oils [4]. However, recent studies have shown that long-term consumption of synthetic antioxidants may cause damage to human health [5]. Today, synthetic antioxidants have been restricted in Canada, Japan, the United States, and European countries. Many countries tend to replace synthetic antioxidants with natural antioxidants [6].

Rosemary (*Rosmarinus officinalis* L.) extracts have become a popular plant-based antioxidant because of their strong antioxidant properties as well as fat solubility and have been officially incorporated into food additive standards in various countries [7]. Li et al. have reported that rosemary extracts are obtained from rosemary whose antioxidative activity is clearly stronger than that of BHA and BBHT [8]. Wang et al. also report that rosemary extracts have strong inhibition in lipid oxidation [6]. Due to the presence of phenol diazole, the antioxidant capacity of rosemary extracts could be improved, which could remove hydroxyl free radicals, monoline oxygen, and lipid propylene oxygen free radicals, thereby inhibiting lipid oxidation [9]. Sage oxalic acid, fennel, and rose phenol are the main phenol diazines in rosemary extracts, of which sage oxalic acid is the

Table 1: Combinations of different types and quantities of antioxidants (g/kg) in six groups.

	I	II	III	IV	V	VI
Rosemary extracts	—	0.7	0.35	—	0.35	0.23
VC	—	—	0.2	—	—	0.13
AP	—	—	—	—	0.1	0.07
TBHQ	—	—	—	0.2	—	—

VC, vitamin C; AP, ascorbyl palmitate; TBHQ, tertbutyl hydroquinone.

main phenol diazole [10]. Additionally, Xie et al. have reported that the most effective antioxidant in rosemary extracts is carnosic acid [11]. The synergy of rosemary extract with other antioxidants has been studied. An interesting finding revealed the synergistic effects of rosemary extracts with ascorbyl palmitate (AP) in providing oxidative stability to sunflower oil [12]. Therefore, more data are needed to fully evaluate the synergistic effects of rosemary extract with other natural antioxidants.

The shelf-life of food refers to the period of time that prepackaged food maintains quality under the storage conditions specified in the label, which is of great significance for us to evaluate the shelf-life of edible oil [13]. Calligaris et al. established the oxidation kinetic model of the peroxide value (POV) of gardenia fruit oil as an evaluation index to predict its shelf-life [14]. Shen et al. established the Arrhenius equation and found that it could predict the shelf-life of DHA seaweed oil [15]. As far as we know, although plant extracts have been used many times to increase the oxidation stability of edible oils [16, 17], shelf-life applications for peanut oil containing rosemary extracts have not been reported. This also includes the Arrhenius model combined with accelerated shelf-life testing (ASLT) which has also been rarely reported. Therefore, it is interesting to develop a shelf-life prediction model for peanut oil mixed with natural antioxidants.

In this study, we evaluated the oxidation stability and shelf-life of rosemary extracts combined with vitamin C (VC) and AP in peanut oil stored at 65°C. Peanut oil with TBHQ and without additives served as positive and negative controls, respectively. Additionally, the oxidation kinetics model of the peanut oil with addition of different antioxidant compounds was also established; so as to provide evidence for the use of rosemary extracts combined with VC and AP instead of chemical antioxidants.

2. Materials and Methods

2.1. Materials. The peanut oil without addition of antioxidants in this study was provided by Shaoyang edible oil factory (Shaoyang, Hunan, China). Rosemary extracts (70% carnosic acid) were obtained from Hunan Warner Pharmaceutical Co. Ltd (Shaoyang, Hunan, China). Sigma–Aldrich (St Louis, MO, USA) provided the vitamin C (VC), ascorbyl palmitate (AP), and tertbutyl hydroquinone (TBHQ). All other chemicals and reagents were of analytical grade (AR).

2.2. Preparation of Peanut Oil with Antioxidants. According to the standards of food additives in China, each antioxidant had a maximum addition amount [15]. As shown in Table 1, combinations of different types and quantities of antioxidants (g/kg) were prepared in Groups I, II, III, IV, V, and VI. The mixtures of peanut oil and different antioxidant compounds were prepared according to Wang et al.'s [18] procedure with slight modifications. Briefly, 20 mg of antioxidant compounds was added separately to cold-pressed peanut oil (100 g). Each mixture was blended thoroughly using a Panda high-pressure homogenizer (PLUS 2000, GEA Niro Soavi, Austria) for 2 min at 15,000 r/min at room temperature and then stored in an oven (65°C, 48 h).

2.3. Detection of Peroxide Value (POV). The POV of peanut oil with addition of different antioxidant compounds was evaluated following a research report [6]. In brief, each peanut oil sample (2 g) was dissolved in $CHCl_3$ (30 mL) and CH_3COOH (20 mL, v/v) mixtures, and then saturated KI solution (1 mL) was added into the mixtures and mixed by hand and left to incubate in darkness for 3 min. After removing the solvent, they added 100 mL of distilled water, ticked to yellow with 0.002 M $Na_2S_2O_3$ standard, and almost disappeared by adding about 1.0 mL of starch indicator (1%) solution to blue. The calculation formula for POV was as follows:

$$\mathrm{POV} = (V - V_0) \times C \times 0.1269 \times 100/m, \quad (1)$$

where V is the volume of $Na_2S_2O_3$ solution added (mL); V_0 is the volume of solution without $Na_2S_2O_3$ (mL); C, the concentration of $Na_2S_2O_3$ solution (g/mL); and m is the mass of peanut oil (g).

2.4. Detection of Conjugated Diene (CD) and Conjugated Triene (CT) Values. The CD and CT values were evaluated according to the previous study [19]. In short, each peanut oil sample (0.1 g) was dissolved in isooctane (25 mL), and the absorbance of CD and CT was detected at $\lambda = 232$ or 268 nm by an ultraviolet spectrophotometer (Metash Instruments Co., Ltd, Shanghai, China). CD and CT were calculated as follows:

$$E = \left(\frac{A}{C}\right) \times L, \quad (2)$$

where A is the absorbance at 232 nm and 268 nm, respectively; C is the concentration of peanut oil sample (g/100 mL); and L is the length of the cuvette (cm).

2.5. Detection of Thiobarbituric Acid Reactive Substances (TBARs). The TBARs of peanut oil with addition of different antioxidant compounds were evaluated following the previous study [7]. In short, each peanut oil sample (5.0 g) was mixed with a trichloroacetic acid-tetraacetic acid (TCA-EDTA) solution (50 mL), including 37.5 g of TCA and 0.5 g of EDTA. The peanut oil samples were shacked and filtered immediately after 30 min. The filtrate (5.0 mL) and thiobarbituric acid (TBA, 5 mL) solution were added into the

tube and heated for 30 min in a thermostat water bath (100°C) to turn pink. The mixture was then cooled to 25°C, and absorbance was obtained using an ultraviolet spectrophotometer (Metash Instruments Co., Ltd, Shanghai, China) at $\lambda = 532$ nm. A blank solution using the 5.0 mL TCA-EDTA solution under similar conditions was prepared. The value of TBARs was expressed in malondialdehyde (MDA)/peanut oil samples (mg/kg).

2.6. Prediction of Shelf-Life Prediction. The prediction of the shelf-life of peanut oil with addition of different antioxidant compounds was evaluated following the experimental procedure described by a previous study [20] with minor modifications. According to the change of POV of each peanut oil sample containing all types antioxidants at different storage temperatures (45, 50 and 55°C) with storage time (0 to 10 days), the kinetic model was used to obtain the oxidation reaction rate constant of the peanut oil samples, and the shelf-life of the peanut oil samples was obtained, which was calculated by the kinetic equation. Additionally, the POV of the peanut oil samples was detected to verify the accuracy of the prediction at 25°C. The shelf-life prediction and oxidation dynamics model analysis were used as the following equations:

Zero-order reaction model:

$$\mathrm{POV} = k_0 t + \mathrm{POV}_0. \tag{3}$$

First-order reaction model:

$$\ln \mathrm{POV} = \ln\ \mathrm{POV}_0 + k_0 t. \tag{4}$$

Arrhenius model:

$$\ln\ k = \frac{-Ea}{RT} + \ln\ k_0. \tag{5}$$

Shelf-life prediction:

$$\text{Shelf} - \text{life prediction} = \frac{\left[\ln\left(\mathrm{POV}_{\mathrm{lim}}\right) - \ln\left(\mathrm{POV}_0\right)\right]}{\left[k_0 \times e^{(-(Ea)/RT)}\right]}, \tag{6}$$

where k_0 and k are the reaction rate constants; T is the absolute temperature (°C); R is the molar gas constant; Ea is the activation energy (J/mol); POV_0 is the POV of peanut oil samples at 25°C; and $\mathrm{POV}_{\mathrm{lim}}$ is the POV of peanut oil samples according to the Chinese standard.

3. Statistical Analysis

All data were analysed using statistical product and service solutions (SPSS) version 21.0 (SPSS Inc. Illinois, USA). Data were exhibited as the mean ± SD and detected by a one-way analysis of variance (ANOVA), and the significance level was $P < 0.05$.

4. Results and Discussion

4.1. Analysis of Peroxide Value (POV). The POV reflects the extent of primary oxidation products, especially in the initial stages of lipid oxidation [6]. Fats and oils form large amounts of peroxides and water peroxides in the oil during oxidation and degradation [21]. Therefore, the POV measurement could be used to describe the degree to which antioxidants inhibit these oxidation products. Figure 1(a) exhibits the effect of different antioxidant compounds on POV in peanut oil under accelerated storage. Firstly, the POV of all the peanut oil samples was very low at day 0, and a slow increase in POV implied higher oxidation stability. On the second day, the POV of peanut oil with addition of group II–VI antioxidant compounds was 0.0743, 0.0694, 0.0658, 0.0721, and 0.0615 g/100 g, respectively, and group I (control) was 0.1090 g/100 g. Additionally, the POV in peanut oil with addition of group II–VI antioxidant compounds was markedly lower than that in group I, suggesting that the oxidation process of the peanut oil was reduced. The maximum POV for antioxidant-free peanut oil was 0.7630 g/100 g after storage for 10 days, while the POV of group VI antioxidant compounds was 0.1352 g/100 g. However, of these antioxidant compounds, group VI was the most effective, with the lowest POV values throughout the period. This is in agreement with the findings of Upadhyay et al. [22] who found that the sunflower oil blended with rosemary extracts and AP outperforms synthetic antioxidants. Yin et al. [23] also reported that AP and vitamin E showed a strong negative effect on the POV of microalgal DHA-rich oil with rosemary extracts. The above results showed that rosemary extracts combined with AP and VC had a strong antioxidant effect and a stable effect on peanut oil.

4.2. Analysis of Conjugated Diene (CD) and Conjugated Triene (CT) Values. The oxidation deterioration of fats and oils can be measured with CD and CT values [24]. The absorbance at $\lambda = 232$ or 268 nm could calculate the CD and CT values, which are used to indicate primary and secondary oxidation products of edible oils [21]. In fact, the higher the CD and CT content, the lower the oxidation stability of the edible oil. Figures 1(b) and 1(c) show the CD and CT values of the peanut oil with addition of different antioxidant compounds under increasing storage, respectively. Figure 1(b) exhibits that the CD value of the control group improved from 2.3722 to 13.7920 after 10 days. The CD value of each type of antioxidant in peanut oil samples was increased to 10 d under increasing storage conditions. As shown in Figure 1(c), the CT value of the peanut oil with addition of different antioxidant compounds continued to improve with the extension of the storage period. After storage of 10 d, the CT value of group I increased to 1.5036, markedly higher than that of the other groups (group II–VI). This finding was consistent with Upadhyay et al. [25], which depicted a beneficial correlation between the values of CD and CT during the assessment of the oxidative stability of olive oils with addition of rosemary extracts and AP. Our results support the findings of Farhoosh et al. [26], who suggested that CD and CT are suitable for monitoring the oxidation quality of peanut oil. The above results showed that group II–VI antioxidant compounds are found to have antioxidant effects compared to group I, based on changes in CD and CT values.

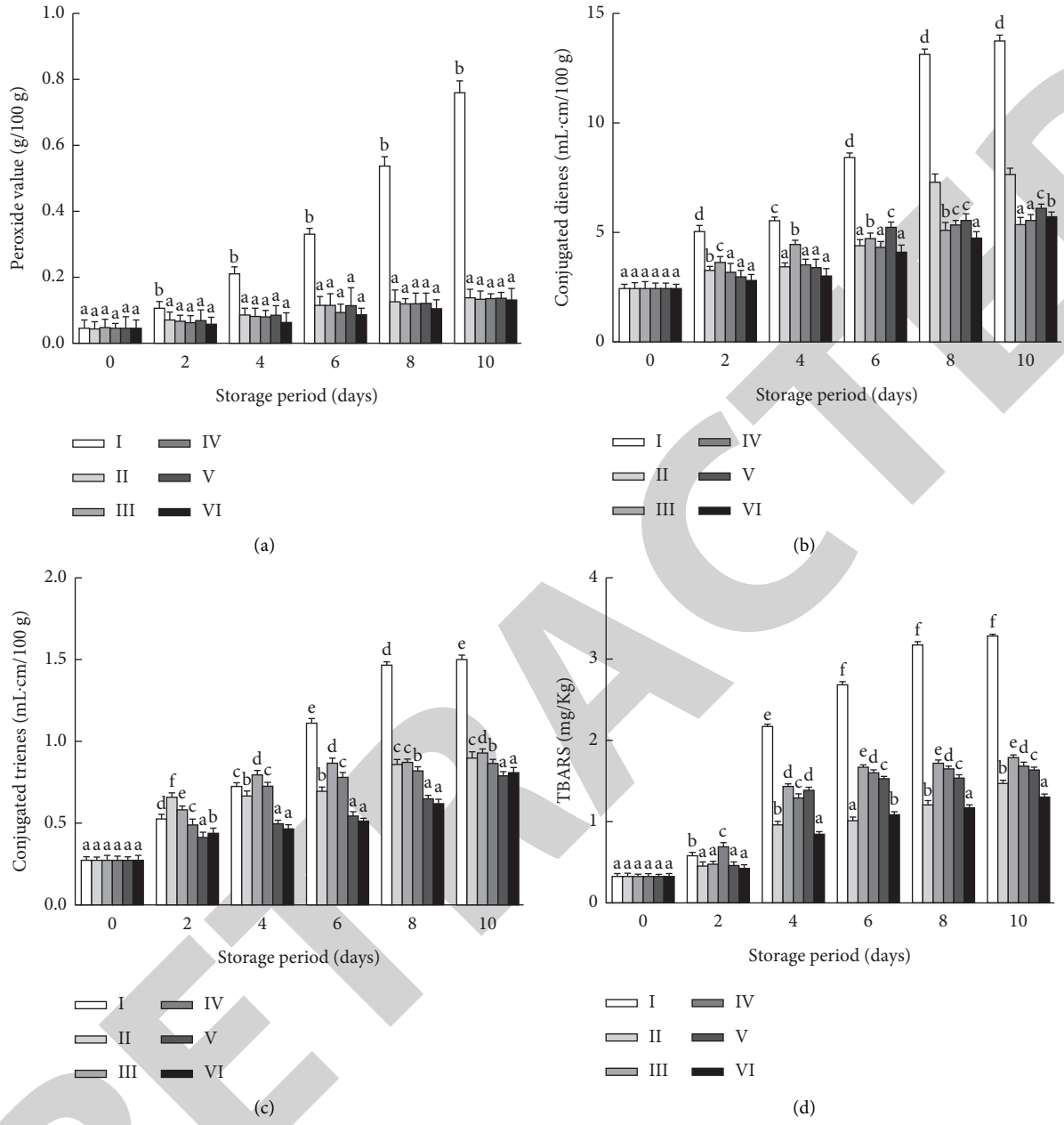

Figure 1: The changes of POV (a), CD (b), CT (c), and TBARs (d) in the peanut oil with addition of Group I II, III, IV, V, and VI antioxidants compounds during the different storage period at 65°C. The value was the average of the three repeated tests ± SD ($n = 3$); values after different letters were considered, $P < 0.05$. POV, peroxidation value; CD, conjugated diene; CT, conjugated triene; and TBARs, thiobarbituric acid reactive substances.

4.3. Analysis of Thiobarbituric Acid Reactive Substances (TBARs). The TBARs are widely used to measure the formation of secondary products of lipid peroxide, which is a signaling of lipid oxidation and measures the content of malignant dialdehyde (MDA) [2, 6]. Therefore, a high TBARs index in an edible oil indicated that the oil was more oxidizing and less stable. The effect of accelerated storage for 10 days on the TBARs value of peanut oil with addition of different antioxidant compounds is shown in Figure 1(d). The initial TBRAs of all peanut oil samples were very low, but gradually increased after 4 days of storage. The peanut oil with addition of group II–VI antioxidant compounds showed a markedly lower TBARs value during accelerated storage than group I. The TBAR value of group I reached the maximum value of 3.2973 mg/kg after storage for 10 days. In controlling for test values, group II–VI antioxidant compounds produced effects in a similar way at all levels. Group VI increased the TBARs value (0.3398 to 1.6448 mg/kg) after storage for 10 d at 65°C. Okhli et al. [4] and Guo et al. [27] also found that rosemary extracts combined with antioxidants are more effective than chemical antioxidants (BBHT and BHA) in

Table 2: The zero-order and first-order equations parameters of peanut oil with addition of Group I or Group VI antioxidant compounds at different temperatures.

Sample	Temperature (oC)	Zero-order reaction regression curve	R^2	First-order reaction regression curve	R^2
Group I	45	$y = 0.0055x + 0.0432$	0.9270	$y = 0.0464e^{0.0769x}$	0.9548
	50	$y = 0.0120x + 0.0444$	0.9782	$y = 0.0527e^{0.1206x}$	0.9951
	55	$y = 0.0256x + 0.0436$	0.9929	$y = 0.0609e^{0.1748x}$	0.9580
	65	$y = 0.0712x - 0.0216$	0.9507	$y = 0.0599e^{0.271x}$	0.9786
Group VI	45	$y = 0.0019x + 0.0476$	0.9658	$y = 0.0482e^{0.0335x}$	0.9794
	50	$y = 0.0034x + 0.0482$	0.9918	$y = 0.0493e^{0.0526x}$	0.9987
	55	$y = 0.0070x + 0.0422$	0.9363	$y = 0.0469e^{0.0896x}$	0.9688
	65	$y = 0.0085x + 0.0425$	0.9562	$y = 0.0483e^{0.1012x}$	0.9860

R^2, regression coefficient.

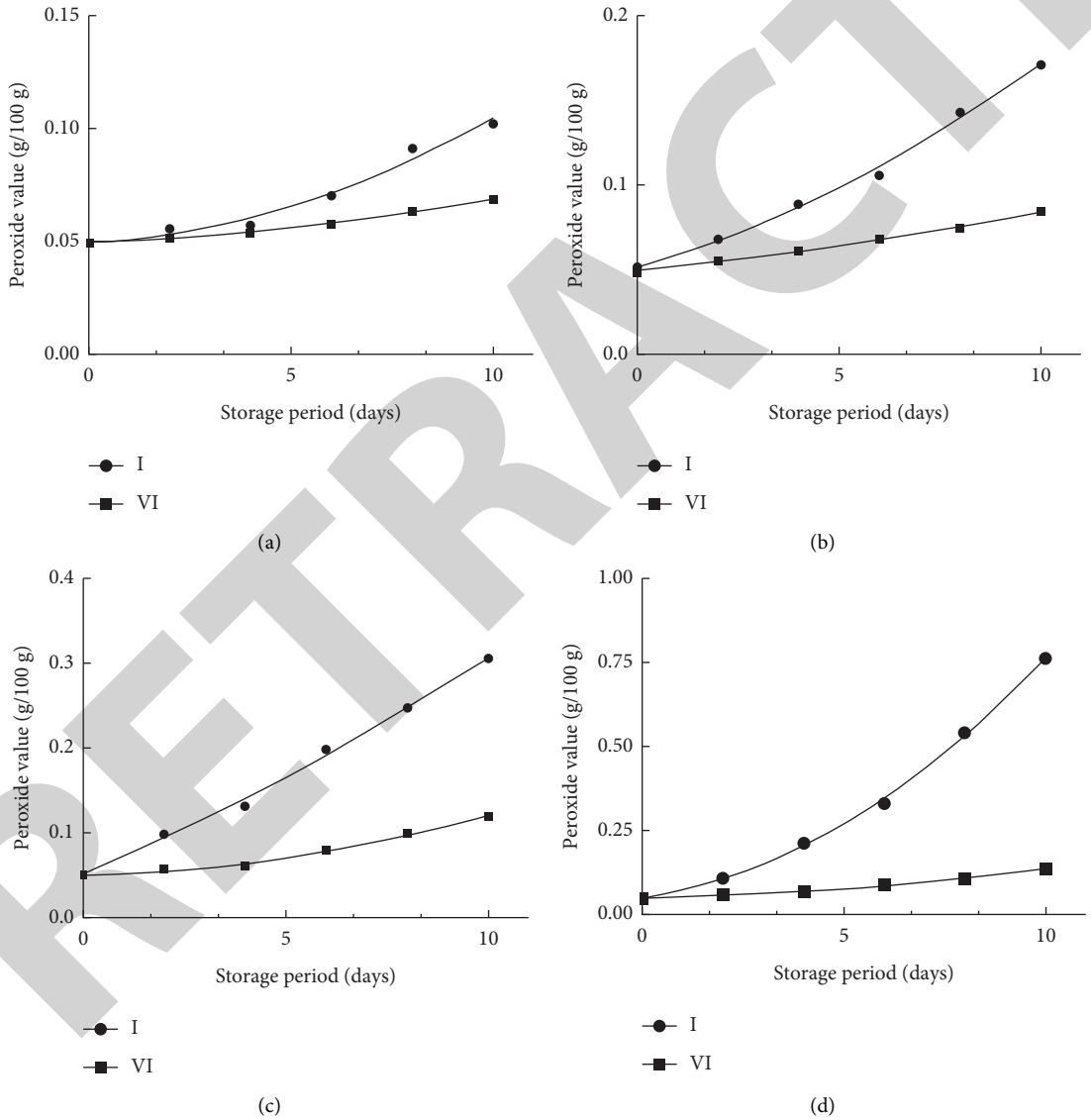

Figure 2: The first-stage dynamic model fitting of POV changes in peanut oil with addition of Group I or Group VI antioxidants compounds during the different storage period at 45°C (a), 50°C (b), 55°C (c), and 60°C (d). POV, Peroxide value.

inhibiting oxidation in edible oil. The above results data showed that antioxidants in rosemary extracts were superior to synthetic antioxidants because they have greater antioxidant activity and stability.

4.4. Analysis of Prediction of Shelf-Life. In the process of oil storage, the kinetic energy of oxidation reactions is mostly zero-order or first-order reaction [28]. The zero-order and first-order equation parameters of peanut oil with addition

Table 3: The Arrhenius model parameters, predicted shelf-life, observed shelf-life, and RE of peanut oil with addition of Group I or Group VI antioxidant compounds.

Samples	Arrhenius model	R^2	Predicted shelf-life (d)	Observed shelf-life (d)	RE (%)
Group I	$\ln k = -6679/T + 18.509$	0.9738	79.28	82.86	4.32
Group VI	$\ln k = -5945.2/T + 15.435$	0.8528	146.63	149.38	1.84

R^2, regression coefficient; RE, relative error.

of group I or group VI antioxidant compounds at different temperatures are shown in Table 2. Table 2 exhibits a higher R^2 value in the fitted first-order kinetic model than that in the zero-order kinetic model at four different storage temperatures. Therefore, the oxidation reaction of peanut oil with addition of group I or group VI antioxidant compounds was a first-order reaction. The first-order reaction kinetic models were respectively fitted with the POV value of peanut oil with addition of group I or group VI antioxidant compounds at different temperatures are shown in Figure 2. The change rate of POV increased with the increase of temperature (Figure 2). Plot the absolute temperature equivalent 1/T to get a straight line with a slope of equation (5) to get Ea and K (Table 3). As shown in Table 3 the predicted shelf-life of peanut oil with addition of group I or group VI antioxidant compounds was 79.28 and 146.63 days at 25°C, respectively. At the same time, the observed shelf-life corresponding to 25°C was 82.86 and 149.38 days, and the relative error (RE) of the shelf-life prediction was 4.32% and 1.84%, respectively. Therefore, this study predicted the shelf-life of peanut oil with addition of group I or group VI antioxidant compounds by measuring POV value. At the same time, to shorten the test time of shelf-life prediction, the shelf-life of peanut oil was predicted based on the Arrhenius model [29]. Table 3 exhibits the shelf-life prediction of the peanut oil with addition of group I or group VI antioxidant compounds was 146.63 days, while the shelf-life of group I was 79.28 days, indicating that the shelf-life of peanut oil could be prolonged by adding group VI antioxidant compounds.

5. Conclusions

In conclusion, the present study showed that 0.23 g/kg rosemary extracts combined with 0.13 g/kg VC and 0.07 mg/kg AP had strong antioxidant effect, which could effectively improve the oxidation stability of peanut oil in an accelerated oven for 10 days. By establishing a model of the oxidation kinetics of peanut oil, it was found that rosemary extracts combined with VC and AP could better extend the shelf-life of peanut oil. It was worth mentioning that oxidation kinetic models had some limitations on the prediction of the shelf-life of peanut oil. We believe that models for rapidly estimating the oxidation stability and shelf-life estimation of peanut oil should be further studied before actual shelf-life experiments are performed under environmental conditions. Therefore, rosemary extracts combined with VC and AP could be regarded as a potential low-cost antioxidant source to increase the oxidation stability of edible oil or as a green alternative to chemical antioxidants. Although the method presented in this study was related to peanut oil, it could be used to develop new edible oils.

Data Availability

All data generated or analysed during this study are included in this published article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Authors' Contributions

WY was responsible for article writing and revision, data analysis, and financial support. DH was involved in study conceptions, procurement of samples, and paper drafting. HDC was involved in procurement of samples and results analysis. ZXH was responsible for data collection, analysis, study conception, and paper drafting. All authors read and approved the final version of the present research manuscript and agreed to be accountable for all aspects of the work.

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