

# Investigating the impact of palm oil refining on the oxidative stability of its blend with linseed oil: a kinetic approach

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**SUMMARY:** This study investigates whether blending palm oil with linseed oil can enhance the oxidative stability of linseed oil, which is prone to oxidation due to its high polyunsaturated fatty acid content. Four blends were prepared: 66 % linseed oil with 33 % crude palm oil (33C), 33 % linseed oil with 66 % crude palm oil (66C), 66 % linseed oil with 33 % refined palm oil (33R), and 33 % linseed oil with 66 % refined palm oil (66R). Oxidation kinetics were analyzed using peroxide value, *p*-anisidine value, TOTOX value, induction period, activation energy, and Gibbs free energy. The results showed that palm oil blends, particularly refined palm oil, significantly improved oxidative stability. The 66R sample exhibited the highest stability, with lower oxidation values, longer induction periods, and higher activation energy. Refining palm oil further enhanced its stabilizing effect. These findings offer practical solutions for the food industry to extend the shelf-life of linseed oil-based products, such as margarines and dressings, while maintaining their nutritional quality.

**KEYWORDS:** Linseed oil; Oil blending; Oxidation kinetic; Oxidative stability; Palm oil; Refining.

**RESUMEN:** *Investigación sobre el impacto del aceite de palma refinado en la estabilidad oxidativa de su mezcla con aceite de linaza: un enfoque cinético.* Este estudio investiga si la mezcla de aceite de palma con aceite de linaza puede mejorar la estabilidad oxidativa del aceite de linaza, que es propenso a la oxidación debido a su alto contenido de ácidos grasos poliinsaturados. Se prepararon cuatro mezclas: 66 % de aceite de linaza con 33 % de aceite de palma crudo (33C), 33 % de aceite de linaza con 66 % de aceite de palma crudo (66C), 66 % de aceite de linaza con 33 % de aceite de palma refinado (33R) y 33 % de aceite de linaza con 66 % de aceite de palma refinado (66R). Se analizó la cinética de oxidación utilizando el índice de peróxido, el índice de *p*-anisidina, el índice de TOTOX, el período de inducción, la energía de activación y la energía libre de Gibbs. Los resultados mostraron que las mezclas de aceite de palma, en particular el aceite de palma refinado, mejoraron significativamente la estabilidad oxidativa. La muestra 66R exhibió la mayor estabilidad, con valores de oxidación más bajos, períodos de inducción más largos y mayor energía de activación. El refinado del aceite de palma mejoró aún más su efecto estabilizador. Estos hallazgos ofrecen soluciones prácticas para que la industria alimentaria pueda extender la vida útil de los productos a base de aceite de linaza, como margarinas y aderezos, manteniendo al mismo tiempo la calidad nutricional.

**PALABRAS CLAVE:** Aceite de linaza; Aceite de palma; Cinética de oxidación; Estabilidad oxidativa; Mezcla de aceites; Refinación.

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## 1. INTRODUCTION

Linseed oil is renowned for its nutritional value, and is an excellent source of alpha-linolenic acid, an essential omega-3 ( $\omega$ 3) fatty acid which is crucial for heart health, brain function, and reducing inflammation. It also contains antioxidants like lignans, which may have cancer-fighting properties, as well as fiber, protein, vitamins, and minerals such as vitamin E, potassium, and magnesium (Golmakani *et al.*, 2020a).

However, its use in the food industry is limited by its susceptibility to oxidation due to its high  $\omega$ 3 content, leading to rancidity and off-flavors. Effective storage and processing methods are required to combat this. Additionally, linseed oil's distinct flavor may not suit all foods, and its high  $\omega$ 3 content makes it unsuitable for high-temperature cooking, as these fatty acids degrade with heat, reducing their nutritional benefits. Despite these challenges, linseed oil remains a valuable nutritional component when properly handled

(Golmakani *et al.*, 2020a). Linseed protein has also been found to support heart health and boost the immune system. Linseed oil has been incorporated into a variety of food products, including baked goods, juices, dairy products, pasta, and meat products, as functional food ingredients (Sahraeian *et al.*, 2024b; Tang *et al.*, 2021).

There are several approaches that can be employed to enhance the oxidative stability of oils. These approaches aim to minimize the oxidation of oils, which can lead to the development of off-flavors, loss of nutritional value, and potential health risks. These approaches include antioxidant addition, encapsulation, hydrogenation, packaging and storage, and the blending of oils (Keramat *et al.*, 2023b; Sahraeian *et al.*, 2023a). To enhance the oxidative stability of oils, antioxidants are commonly used. Natural antioxidants like tocopherols, phenolics, carotenoids, and flavonoids, while present in plant sources, can be less effective and degrade during processing. Synthetic antioxidants such as BHT and BHA are more potent but raise concerns about health, the environment, and allergens, limiting their practical use due to regulations and consumer preferences (Decker *et al.*, 2005). The hydrogenation of oils is a common method for enhancing stability, but it can lead to issues like trans-fat formation, partial hydrogenation drawbacks, nutritional changes, flavor, and texture alterations, cost, equipment demands, and environmental impact (Sahraeian *et al.*, 2023b; Hamm *et al.*, 2013).

The blending approach is a widely recognized method to enhance the oxidative stability of oils by combining different types of oils with varying stability properties (Golmakani *et al.*, 2020a). This method leverages the synergistic effects between components, where oils with high oxidative stability, such as palm, coconut, palm kernel, and avocado oils, protect those more prone to oxidation, like sunflower, soybean, and linseed oils (Keramat *et al.*, 2023a; Asadi-Yousefabad *et al.*, 2021). These high-stability oils act as antioxidants, neutralizing free radicals and slowing oxidation in the vulnerable oils. Besides improving stability, blending can also enhance sensory attributes by selecting oils with desirable flavors and aromas, creating a unique and appealing taste profile (Sahraeian *et al.*, 2024a; Lu *et al.*, 2020). Ensuring success in this approach involves considering the compatibility and proportion of oils,

fine-tuning the blend's stability by adjusting the oil ratios to achieve the desired stability level. Palm oil is a versatile and widely used vegetable oil that is extracted from the fruit of the oil palm tree, scientifically known as *Elaeis guineensis*. It is known for its unique properties, making it a popular ingredient in various industries, including food, cosmetics, and biofuel. One of the key characteristics of palm oil is its highly-saturated fatty acids. This gives palm oil a solid texture at room temperature, making it suitable for use in food products such as margarine and baked products (Basiron 2007). Its stability and resistance to oxidation make it an ideal choice for frying and cooking purposes. Palm oil contains important minor components such as tocopherols, tocotrienols, and carotenoids, which contribute to its unique nutritional properties. Palm oil is said to possess the highest tocotrienol content among vegetable oils (Golmakani *et al.*, 2025; Damanik and Murkovic, 2018).

Hashempour-Baltork *et al.* (2018) reported that blending linseed oil with olive and sesame oils could successfully balance the  $\omega 6:\omega 3$  ratio in oil blends, increasing the oxidative stability of linseed oil due to the presence of bioactive compounds with antioxidant activities. In another study, Golmakani *et al.* (2020b) investigated the oxidative stability of linseed oil by blending it with various oils, including corn, canola, sesame, and bitter almond oils, at different ratios. Their findings revealed that the blend containing 33% linseed oil and 66% corn oil exhibited the highest oxidative stability. This was evidenced by a reduction in the Q10 value from 2.53 for pure linseed oil to 1.80 for the linseed-corn oil blend, indicating improved stability. De Leonardis and Macciola (2012) demonstrated that blending extra virgin olive oil with palm oil significantly influenced its oxidative stability. They reported that a binary blend containing 20% extra virgin olive oil and 80% palm oil exhibited the highest oxidative stability compared to the pure extra virgin olive oil. Fadda *et al.* (2022) reviewed sustainable strategies to enhance the oxidative stability of various vegetable oils. They identified blending plant oils with more oxidatively stable oils as an effective and sustainable approach to improving their stability. According to the literature, blending linseed oil with more oxidatively stable plant-based oils, such as palm oil, offers a practical and sustainable solution to enhance its oxidative stability. This approach eliminates the

need for additional antioxidants, reduces costs, and aligns with sustainable practices.

In this study, linseed oil was chosen as the primary research object due to its high susceptibility to oxidation caused by its highly polyunsaturated fatty acid content. To enhance its oxidative stability, linseed oil was blended with crude and refined palm oils, which are known for their higher oxidative stability owing to their saturated fatty acid content and natural antioxidants. The objective of this research was to investigate the benefits of incorporating different portions of crude palm oil and refined palm oil into linseed oil through various blends. The study primarily focused on assessing oxidative stability in comparison with pure linseed oil and examining its potential to extend linseed oil's shelf-life by analyzing oxidation kinetics. To examine a comprehensive range of linseed oil: palm oil ratios the blending ratios of 33 and 66% for palm oils were selected. These ratios also allow for a clear comparison of the effects of lower and higher concentrations of palm oil on linseed oil stability. Finally, this study underscores the critical role of refining in the final oxidative stability and highlights the effect of impurities in crude palm oil on oxidative stability.

## 2. MATERIALS AND METHODS

### 2.1. Materials

Cold-pressed linseed as well as refined and crude palm oils without any added antioxidants were supplied by Nourhan-Moohaya Company (Shiraz, Fars province, Iran). Acetic acid, chloroform, potassium iodide, sodium thiosulfate ( $\text{Na}_2\text{S}_2\text{O}_3$ , CAS No. 7772-98-7), iso-octane (2,2,4-trimethylpentane) (CAS No. 51685-57-5), and *p*-anisidine reagent (99%, CAS No. 104-94-9) were purchased from Sigma-Aldrich (St. Louis, MO). In all experiments, double-distilled water was used to avoid any unwanted reaction of impurities with oil samples. All solvents used in Gas chromatography (GC) were GC-grade solvents.

### 2.2. Fatty acids profile and oil quality

The fatty acid profile of linseed oil as well as refined and crude palm oil was accurately determined using gas chromatography (SP-3420A, Beijing Beifen-Ruili Analytical Instrument, Beijing,

China) as previously described (Golmakani *et al.*, 2020a). The PV of linseed oil as well as refined and crude palm oil was determined according to Karamat *et al.* (2023b) with some modifications. The oil sample was accurately weighed (5 g) and added to acetic acid and chloroform (9.8 mL, 3:2 v/v) and mixed well to prepare a uniform solution. Saturated potassium iodide (0.5 mL) was then added to the mixture dropwise for 10 min followed by the addition of double-distilled water (30 mL). The resulting solution was titrated using sodium thiosulfate ( $\text{Na}_2\text{S}_2\text{O}_3$ ) (0.1 N) until the disappearance of the yellow color. A starch solution (0.5 mL, 1 %) was added to the solution as an indicator. Again, the solution was titrated while stirring using a thiosulfate solution (0.1 N) until the disappearance of the blue color. The blank solution contained all the reagents except oil. PV was calculated using Equation 1 and the results were reported as milliequivalent oxygen per Kg of oil (meq  $\text{O}_2$ /Kg oil):

$$\text{PV} = [(\text{mL of Na}_2\text{S}_2\text{O}_3 \text{ of sample}) - (\text{mL of Na}_2\text{S}_2\text{O}_3 \text{ of blank})] \times \text{Equation 1} \\ \times \text{N} \times 1000 / \text{W}$$

where N is the normality of the thiosulfate solution (0.1 N) and W is the weight of the sample.

The AV of the oil samples was determined using the procedure described by Golmakani *et al.* (2018) with some modifications. The oil sample (4 g) was accurately weighed and diluted to a volume of 25 mL using iso-octane. The absorbance was then recorded at 350 nm for the solution against iso-octane as the blank. After that, 5 mL of the solution were transferred to a glass tube, and 1 mL of anisidine reagent was added to the tube. The second tube containing 10 mL of iso-octane and 1 mL of anisidine reagent was immediately prepared. After 10 min, the absorbance was recorded at 350 nm against the second tube as the blank. The AV of the oil samples was calculated according to Equation 2 and the results were reported as milligrams of *p*-anisidine per kilogram of oil (mg/kg).

$$\text{AV (mg/kg)} = 25[0.5(\text{A}_2 - \text{A}_1)] / \text{m} \quad \text{Equation 2}$$

where A1 and A2 are the absorbance of solutions before and after the reaction with anisidine reagent,

respectively, and  $m$  is the mass of the sample in grams (4 g).

The total oxidation value (TOTOX) was also evaluated using Equation (3) (Amini *et al.*, 2023).

$$\text{TOTOX value} = (2 \times \text{peroxide value}) + \text{AV} \quad \text{Equation 3}$$

### 2.3. Preparation of oil blends

In this study, refined and crude palm oils were added to the linseed oil to evaluate the effects of additional oil on the oxidation and quality of linseed oil. The chosen proportions of oils were 33 and 66% for both crude and refined palm oils. Each oil was added to the linseed oil according to the predetermined ratios followed by thermally stirring (40 °C, 30 min, 150 rpm) to achieve a uniform mixture of oils. Finally, the samples containing 33% and 66% crude palm oil were named 33C and 66C blends, respectively, and the samples containing 33 and 66% refined palm oils were named 33R and 66R, respectively. The oxidation stability of the oil mixtures was then determined using Rancimat.

### 2.4. Oxidative stability

Using a pre-calibrated Rancimat (Metrohm, model 743, Switzerland), oxidative stability was determined according to Jokar *et al.* (2022). Briefly, the oil blend was accurately weighed (3 g) and transferred to the reaction vessel. The water bath of the Rancimat apparatus was filled with distilled water and the temperature set to 80, 100, and 120 °C, while the airflow rate was set to 20 L/h. The Rancimat will heat the sample to the desired temperature and pass a stream of air through it. After reaching the desired temperature, samples were placed in the heater channel. As the oil blend oxidizes, it releases volatile organic acids which are carried by the air stream into the reaction vessel, where they interact with the deionized water to form conductivity changes. Finally, the induction period (IP) was recorded after the predetermined time.

### 2.5. Oxidation kinetic

The data required for each parameter were obtained using a pre-calibrated Rancimat. These

parameters include the temperature coefficient ( $T_c$ ), acceleration factor ( $Q_{10}$ ), Arrhenius equation parameters (frequency factor “ $A$ ” and activation energy  $E_a$ ), and activated complex theory parameters (entropy  $\Delta S$  and enthalpy  $\Delta H$ ). To determine the temperature coefficient ( $T_c$ ,  $K^{-1}$ ), acceleration factor ( $Q_{10}$ ), and shelf-life, the natural logarithm of IP ( $\ln(IP)$ ) was plotted against the temperature (K). To find the shelf-life of oil samples at 25 °C. Corresponding plots were extrapolated according to Equation 4 (Golmakani *et al.*, 2020b).

$$\log(IP) = a(T) + b \quad \text{Equation 4}$$

where  $a$  is the slope of the plot indicating  $T_c$ .

The  $Q_{10}$  factor can also be determined as Equation 5 (Golmakani *et al.*, 2020b).

$$Q_{10} = 10^{-10/T_c} \quad \text{Equation 5}$$

The rate constant of the oxidation reaction ( $k$ ,  $h^{-1}$ ) was then calculated according to Equation 6 (Arrhenius equation). The intercept of the equation refers to the activation energy ( $E_a$ ,  $KJ \cdot mol^{-1}$ ), and the slope indicates the frequency factor ( $A$ ,  $h^{-1}$ ) (Golmakani *et al.*, 2020b).

$$\log(k) = \log(A) - (E_a/2.303RT) \quad \text{Equation 6}$$

where  $R$  is the molar gas constant ( $8.314 \text{ J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$ ).

According to the activated complex theory, the enthalpy ( $\Delta H$ ) and entropy ( $\Delta S$ ) of the oxidation reaction, which refer to slope and intercept, respectively, were calculated (Equation 7) (Toorani *et al.*, 2019).

$$\log(k/T) = \log(K_b/h) + (\Delta S/R) - (\Delta H/RT) \quad \text{Equation 7}$$

where  $R$  is the molar gas constant ( $8.314 \text{ J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$ ),  $K_b$  is Boltzmann constant ( $1.38 \times 10^{-23} \text{ J} \cdot \text{K}^{-1}$ ), and Planck's constant ( $6.63 \times 10^{-34} \text{ Js}$ ).

Finally, Gibbs free energy ( $\Delta G$ ) was then determined according to Equation 8 (Toorani *et al.*, 2019).

$$\Delta G = \Delta H - T\Delta S \quad \text{Equation 8}$$

It should be noted that  $T$  is temperature in Kelvin.

## 2.6. Statistical analysis

The experiments were conducted in triplicate and the results were reported as mean values. The difference between means was evaluated using one-way ANOVA and Duncan's multiple-range test using SAS statistical software (SAS 9.4, SAS Institute, Cary, NC) with a significance level of 0.05. Plots were generated using Microsoft Excel Spreadsheet Software (Microsoft Office, 2016, Microsoft Corporation, Redmond, Washington).

## 3. RESULTS AND DISCUSSION

### 3.1. Oil composition and quality

The fatty acid profiles of linseed oil, crude palm oil, and refined palm oil are summarized in Table 1. Linseed oil is characterized by a high content of polyunsaturated fatty acids (PUFAs), with  $\alpha$ -linolenic acid (an  $\omega$ 3 fatty acid) being the predominant fatty acid at 55.25%. Oleic acid (21.08%) and linoleic acid (15.15%) were the second- and third-most abundant fatty acids, respectively. Overall, PUFAs accounted for approximately 91.5% of the total fatty acids in linseed oil, making it highly susceptible to oxidation. Saturated fatty acids, such as palmitic acid and stearic acid, constituted only about 12% of the total fatty acids in linseed oil. In contrast, palm oil (both crude and refined) exhibited a significantly different fatty acid profile. Palmitic acid, a saturated fatty acid, was the dominant component, accounting

for 44 and 46.5% of the total fatty acids in refined and crude palm oil, respectively. Oleic acid, a mono-unsaturated fatty acid, was the second-most abundant fatty acid in palm oil, representing 37% and 38.88% in refined and crude palm oil, respectively. Notably, palm oil contained no stearic acid or  $\alpha$ -linolenic acid, which are present in linseed oil. Similarly, Edo *et al.* (2022) found that palmitic acid and oleic acid are the major fatty acids in palm oil while linolenic acid is a micro-constituent.

The blending of linseed oil with palm oil resulted in intermediate fatty acid profiles. For instance, the 33% palm oil blends (33R and 33C) contained 36.83 and 36.8%  $\alpha$ -linolenic acid, respectively, which is significantly lower than the 55.25% found in pure linseed oil. These blends also showed higher levels of palmitic acid (19.48% and 20.31%) and oleic acid (26.39% and 25.83%) compared to pure linseed oil. On the other hand, the 66% palm oil blends (66R and 66C) exhibited even lower levels of  $\alpha$ -linolenic acid (18.42% and 17.89%) and higher levels of palmitic acid (31.74% and 33.41%) and oleic acid (31.69% and 33.19%).

The comparison of these results highlights the significant impact of blending on the fatty acid composition of the oil mixtures. By blending linseed oil with palm oil, the proportion of PUFAs (particularly  $\alpha$ -linolenic acid) reduced, while the proportion of saturated and monounsaturated fatty acids (palmitic acid and oleic acid) increased. This shift in fatty acid composition is expected to enhance the oxidative stability of the blends, as saturated and mono-unsaturated fatty acids are less prone to oxidation than PUFAs. These findings align with the observed improvements in oxidative stability, as discussed in subsequent sections, and underscore the potential of palm oil blends to extend the shelf-life of linseed oil.

PV is a measure of oil freshness and quality, which indicates the hydroperoxide content in oil, and reveals the primary products of oxidation at the early stages of the reaction. Oxidized oils can also have reduced nutritional value and may have negative health effects. It is noteworthy that PV is not the only parameter used to assess the quality of oils. Other parameters such as fatty acid profile can also provide valuable information about the quality and composition of oils (Chen and Liu, 2020). The maximum standard level of PV is reported to be 15 milliequivalent of active oxygen/kg oil for virgin and cold-

TABLE 1. Fatty acid profile of linseed oil and crude and refined palm oil.

| Oil sample       | Fatty acids (%) |              |            |               |                          |
|------------------|-----------------|--------------|------------|---------------|--------------------------|
|                  | Palmitic acid   | Stearic acid | Oleic acid | Linoleic acid | $\alpha$ -Linolenic acid |
| Linseed oil      | 7.22*           | 4.38         | 21.00      | 15.15         | 52.25                    |
| Refined palm oil | 44.00           | ND**         | 37.00      | 9.00          | ND                       |
| Crude palm oil   | 46.50           | ND           | 38.88      | 10.00         | ND                       |
| 33R blend        | 19.48           | 3.22         | 26.39      | 13.10         | 36.83                    |
| 66R blend        | 31.74           | 1.61         | 31.69      | 11.05         | 18.42                    |
| 33C blend        | 20.31           | 3.31         | 25.83      | 13.43         | 36.80                    |
| 66C blend        | 33.41           | 1.55         | 33.19      | 11.72         | 17.89                    |

\* The data were accurately determined using gas chromatography.

\*\* Not detected.

pressed oils and 10 milliequivalent of active oxygen/kg oil for other types of oils and fats (Codex Alimentarius Commission 1999). As shown in Table 2, all samples exhibited PVs lower than the critical level. However, refined palm oil exhibited the lowest PV, followed by crude palm oil, and linseed ( $p < 0.05$ ). The AV is also a measure of the oxidative stability of edible oils and determines the presence and extent of oxidative deterioration in oils and fats. The AV is based on the reaction of *p*-anisidine with the secondary oxidation products of oils and fats which are formed during the oxidation process (Golmakani *et al.*, 2018). According to the literature, there is no specific maximum allowed AV for edible oils. Khakbaz Heshmati *et al.* (2022) reported that *p*-anisidine values below 10 ( $\text{mg}\cdot\text{kg}^{-1}$ ) are regarded as good-quality grades. The maximum AV was recorded for linseed oil and the minimum level was detected for refined palm oil. However, it is crucial to consider TOTOX as a determining factor for the quality of edible oils due to the fact that the TOTOX value is based on both PV and literature (Özdemir *et al.*, 2021). It is reported that edible oils with TOTOX values higher than 10 are not good-quality grade oils (Özdemir *et al.*, 2021). According to Table 2, the only unacceptable sample was linseed oil with a TOTOX value of 18.68. The reason why linseed oil is so susceptible to oxidation is its high content of PUFAs, specifically  $\alpha$ -linolenic acid, as the gas chromatography results revealed. PUFAs contain multiple double bonds in their chemical structure, which makes them more vulnerable to oxidation than saturated or monounsaturated fatty acids.

In summary, the choice of PV, AV, and TOTOX as study parameters is justified by their ability to provide a comprehensive evaluation of oil quality and oxida-

tive stability. The results of this study are consistent with previous findings, highlighting the role of fatty acid composition and refining processes in determining oxidative stability. These findings underscore the potential of palm oil blends, particularly those containing refined palm oil, to improve the shelf-life and quality of linseed oil-based products.

### 3.2. Stability against oxidation

The induction period (IP), or oxidative stability index (OSI), is a term used in chemistry to describe the initial stage of a chemical reaction, during which the reactants are being transformed into the products. This period is characterized by a slow rate of reaction, which gradually increases until it reaches the maximum rate with a steady state. The induction period is a crucial stage of a chemical reaction, as it determines the rate and extent of the reaction. Therefore, the higher IP of a specific oil indicates the higher stability of the oil (Souza *et al.*, 2021).

As shown in Table 3, at 80 °C, refined palm oil with an IP of 54.17 showed the highest oxidative stability among other oil samples. In contrast, linseed oil with an IP of 17.34 showed the lowest oxidative stability. Comparatively, the oxidative stability of oil samples at 80 °C was as follows: Refined palm oil > 66R blend > crude palm oil > 33R blend > 66C blend > linseed oil > 33C blend. Since the highest stability was reported for refined palm oil, the blends containing a higher content of refined palm oil, which were the 66R and the 33R blends, showed higher stability

TABLE 2. Peroxide value (PV) and *p*-anisidine value of crude and refined palm oils and linseed oil.

| Oil sample       | PV<br>( $\text{meq O}_2\cdot\text{kg}^{-1}$ ) | <i>p</i> -anisidine<br>( $\text{mg}\cdot\text{kg}^{-1}$ ) | TOTOX<br>value              |
|------------------|-----------------------------------------------|-----------------------------------------------------------|-----------------------------|
| Linseed oil      | $5.64 \pm 0.15^{\text{a}}$                    | $7.40 \pm 0.66^{\text{a}}$                                | $18.68 \pm 0.99^{\text{a}}$ |
| Crude palm oil   | $0.85 \pm 0.08^{\text{b}}$                    | $1.30 \pm 0.09^{\text{b}}$                                | $3.00 \pm 0.81^{\text{b}}$  |
| Refined palm oil | $0.60 \pm 0.04^{\text{c}}$                    | $0.78 \pm 0.07^{\text{c}}$                                | $1.98 \pm 0.69^{\text{c}}$  |

\* Data are expressed as mean  $\pm$  standard deviation. Duncan's multiple range test procedure was used to identify significant differences ( $p < 0.05$ ). In each column, mean values with different superscript letters are significantly different. All treatments were performed in three replicates.

TABLE 3. Induction period (IP) of linseed oil with different ratios of crude and refined palm oils at different temperatures.

| Sample           | IP                          |                             |                             |
|------------------|-----------------------------|-----------------------------|-----------------------------|
|                  | 80 °C                       | 100 °C                      | 120 °C                      |
| Linseed oil      | $17.34 \pm 0.33^{\text{g}}$ | $3.88 \pm 0.09^{\text{f}}$  | $0.40 \pm 0.06^{\text{f}}$  |
| Crude palm oil   | $37.36 \pm 1.87^{\text{c}}$ | $14.87 \pm 0.24^{\text{c}}$ | $8.23 \pm 0.11^{\text{c}}$  |
| Refined palm oil | $54.17 \pm 2.91^{\text{a}}$ | $30.5 \pm 1.25^{\text{a}}$  | $11.93 \pm 0.16^{\text{a}}$ |
| 33C blend        | $16.39 \pm 0.29^{\text{f}}$ | $3.80 \pm 0.10^{\text{f}}$  | $1.07 \pm 0.29^{\text{e}}$  |
| 66C blend        | $26.13 \pm 0.97^{\text{e}}$ | $5.76 \pm 0.09^{\text{e}}$  | $1.55 \pm 0.33^{\text{e}}$  |
| 33R blend        | $33.34 \pm 1.89^{\text{d}}$ | $12.45 \pm 0.16^{\text{d}}$ | $4.59 \pm 0.99^{\text{d}}$  |
| 66R blend        | $41.84 \pm 2.23^{\text{b}}$ | $16.29 \pm 0.36^{\text{b}}$ | $9.27 \pm 0.13^{\text{b}}$  |

\* Data are expressed as mean  $\pm$  standard deviation. Duncan's multiple range test procedure was used to identify significant differences ( $p < 0.05$ ). In each column, mean values with different superscript letters are significantly different ( $p < 0.05$ ). All treatments were performed in three replicates.

among the oil blends. The higher stability of the 33R blend compared to the 33C blend, and the 66C blend compared to the 33C blend, can be attributed to the differences in their fatty acid profiles. All blends containing 66% palm oil had substantially lower  $\alpha$ -linolenic acid and higher palmitic acid in comparison with their 33% counterparts. At 100 °C and 120 °C, the order of oxidative stability of oil samples was similar to that of 80 °C, except for the case of the 33C blend. The oxidative stability of the 33C blend was not significantly different from that of the linseed oil. This implies that at higher temperatures (above 100 °C), there is no difference between the oxidative stability of linseed oil and the 33C blend, and the blending approach with this proportion cannot be beneficial in terms of oxidative stability.

It is acknowledged that the presence of PUFAs in linseed oil is the main reason for its high susceptibility to oxidation. The addition of other resistant oils and making oil blends is a viable approach to exploit such beneficial oil in the human diet with acceptable stability against oxidation. Golmakani *et al.* (2020b) reported that blending linseed oils with corn, canola, sesame, and bitter almond oils positively improves the oxidative stability of linseed oil. The addition of palm oil to the linseed oil endows higher saturated and monounsaturated fatty acids as well as higher antioxidant compounds including  $\beta$ -carotene and vitamin E, which make the blend resistant to oxidation (Damanik and Murkovic, 2018). However, the results revealed that there was a significant difference between the oxidative stability of refined palm oil and crude palm oil as well as their blends. Palm oil is processed in a refinery where it undergoes various treatments to improve its quality (de Almeida *et al.*, 2019). Degumming, alkali refining, bleaching, and deodorization (removing free fatty acids and impurities) are included in the process of refining, even though palm oil requires a lower extent of refining in comparison with other vegetable oils. In addition, palm oil is known as the richest vegetable oil in tocotrienols. On the other hand, it has been reported that the initial PV of crude palm oil is considerably removed and the contents of tocopherol and tocotrienol are not significantly decreased after the refining process (Damanik and Murkovic, 2018). Therefore, it is not unexpected that the absence of impurities and free fatty acids can be substantially effective on the oxidative stability of refined palm oil and the resultant linseed oil blends.

A key novelty of this study is the comparison between crude and refined palm oil in blends with linseed oil, which has not been extensively explored in previous research. The refining process, which includes degumming, alkali refining, bleaching, and deodorization, removes impurities and free fatty acids, while preserving beneficial compounds like tocopherols and tocotrienols. This study demonstrates that refined palm oil significantly outperforms crude palm oil in enhancing the oxidative stability of linseed oil blends, underscoring the critical role of refining in achieving superior stability.

The practical significance of this research lies in its potential applications in the food industry, where oxidative stability is crucial for extending the shelf-life of edible oils and their derivatives, such as margarines, spreads, and salad dressings. The findings provide a cost-effective and sustainable solution for utilizing linseed oil in food products without compromising stability or nutritional quality. By optimizing blending ratios and emphasizing the benefits of refined palm oil, this study offers a novel approach to improving the oxidative stability of PUFA-rich oils, addressing a key challenge in the food industry.

### 3.3. Oxidation Kinetic

#### 3.3.1. Temperature coefficient and acceleration factor

The temperature coefficient ( $T_c$ ) in oil oxidation indicates how the oxidation rate changes with temperature. As temperature rises, the oxidation rate increases, and  $T_c$  measures this relationship. The acceleration factor ( $Q_{10}$ ) shows how much the oxidation rate rises with a temperature increase, often expressed as a multiple of the original rate. The activation energy ( $E_a$ ) is crucial for calculating  $Q_{10}$ , as it determines the rate increase due to temperature.  $T_c$  and  $Q_{10}$  are essential for understanding oil behavior under different conditions and predicting its lifespan in various applications (Dini *et al.*, 2023). Higher  $T_c$  and  $Q_{10}$  indicate that a lower amount of energy or change in temperature is needed to change the rate of oxidation, which means a higher oxidation rate (Shooli *et al.*, 2024; Golmakani *et al.*, 2020b).

The findings of the oxidation experiments using the Rancimat method are summarized in Table 4. According to the results, linseed oil showed the highest  $T_c$  followed by crude palm oil blends (33C

TABLE 4. Kinetic parameters of crude and refined palm oils, linseed oil, and oil blends using the Rancimat method.

| Oil sample       | Arrhenius equation                     |      |                       |                             | Activated complex theory                              |                                      | Gibbs free energy ( $\Delta G$ ; at 353, 373, and 393 K) |                               |                               |
|------------------|----------------------------------------|------|-----------------------|-----------------------------|-------------------------------------------------------|--------------------------------------|----------------------------------------------------------|-------------------------------|-------------------------------|
|                  | Tc $\times 10^{2*}$ (K <sup>-1</sup> ) | Q10* | A* (h <sup>-1</sup> ) | Ea* (kJ·mol <sup>-1</sup> ) | $\Delta S^*$ (kJ·mol <sup>-1</sup> ·K <sup>-1</sup> ) | $\Delta H^*$ (kJ·mol <sup>-1</sup> ) | 353 K (kJ·mol <sup>-1</sup> )                            | 373 K (kJ·mol <sup>-1</sup> ) | 393 K (kJ·mol <sup>-1</sup> ) |
| Crude palm oil   | -0.02                                  | 1.69 | $1.95 \times 10^7$    | 172.33                      | -0.12                                                 | 57.08                                | 97.86                                                    | 100.18                        | 102.49                        |
| 33C blend**      | -0.03                                  | 2.03 | $4.40 \times 10^{10}$ | 156.41                      | -0.05                                                 | 78.40                                | 95.15                                                    | 96.26                         | 97.37                         |
| 66C blend**      | -0.03                                  | 1.98 | $2.74 \times 10^{10}$ | 161.93                      | -0.06                                                 | 75.64                                | 96.52                                                    | 97.55                         | 98.58                         |
| Refined palm oil | -0.02                                  | 1.46 | $3.30 \times 10^4$    | 232.58                      | -0.17                                                 | 37.51                                | 98.89                                                    | 102.21                        | 105.53                        |
| 33R blend**      | -0.02                                  | 1.46 | $7.11 \times 10^4$    | 169.31                      | -0.16                                                 | 40.52                                | 97.03                                                    | 100.40                        | 103.78                        |
| 66R blend**      | -0.02                                  | 1.42 | $4.55 \times 10^4$    | 199.05                      | -0.17                                                 | 40.32                                | 97.79                                                    | 101.03                        | 104.28                        |
| Linseed oil      | -0.04                                  | 2.53 | $2.93 \times 10^{14}$ | 106.47                      | 0.02                                                  | 103.37                               | 95.64                                                    | 95.21                         | 94.77                         |

\* The data required for each parameter are obtained from a pre-calibrated Rancimat. Temperature coefficient (Tc), acceleration factor (Q10), Arrhenius equation parameters including "A" and activation energy (Ea), Activated complex theory parameters including entropy ( $\Delta S$ ) and enthalpy ( $\Delta H$ ).

and 66C). The obtained Tc of refined palm oil and its blends (33R and 33R) as well as unrefined palm oil was 0.02, which means a lower oxidation rate. The higher oxidation rate of 33R in comparison with other oil samples may stem from the presence of impurities in crude palm along with the higher PUFAs of linseed oil in the blends. The absence of impurities in refined palm oil can also be a positive factor leading 33R and 33R blends and refined palm oil to lower oxidations rates in comparison with other oil samples. In addition, the absence of PUFAs in crude palm oil caused a lower oxidation rate than linseed oil and 33C and 66C blends. Similarly, Heidarpour and Farhoosh (2018) reported that the addition of palm olein oil to different types of olive oil can reduce the Tc and lower the rate of oxidation of the blend oils. They concluded that the lower amounts of PUFAs and a higher level of inherent antioxidants could significantly reduce the oxidation rate of palm olein oil and its blends.

On the other hand, a lower Q10 value indicates that raising the temperature by 10 °C in the reaction chamber caused a lower increase in the rate of oxidation reaction than a reaction with a higher Q10. The Q10 values of oils and blend oils were between 1.42 and 2.53. Similar to that observed for the Tc values of linseed oil, palm oil, and oil blends, the highest Q10 value was observed for linseed oil followed by crude palm oil blends (33C and 66C). Crude palm oil had a Q10 of 1.69, which was higher than refined palm oil and its blends (66R and 33R). Therefore, it

can be stated that the oxidation rate of 33R and 33R oil blends is significantly lower than 33C and 66C as well as linseed oil. Similarly, blending linseed oil with corn, canola, sesame, and bitter almond oils has been reported to be beneficial in lowering the Q10 of the oxidation due to a positive change in the anti-oxidant content in linseed oil blends and their fatty acid profiles (Golmakani *et al.*, 2020b). The acquired Q10 values for the blends, especially 66R and 33R blends, were lower than those found by Ghosh *et al.* (2019) for sunflower and sesame oil and their blends, Heidarpour and Farhoosh (2018) for blends of olive and palm olein oils, and Golmakani *et al.* (2020b) for blends of linseed oil and corn, canola, sesame, and bitter almond oils.

### 3.3.2. Arrhenius equation

In the Arrhenius equation, the frequency factor (A) quantifies the frequency of successful collisions between reactant molecules per unit time. It represents the likelihood of reactant molecules meeting with the correct orientation and sufficient energy to initiate a reaction. Factors influencing A include reactant concentrations, temperature, and molecular nature. A higher A indicates a higher probability of successful collisions and a faster reaction rate, while a lower A implies fewer effective collisions and a slower reaction rate (Golmakani *et al.*, 2020b).

As shown in Table 4, the frequency factor of linseed oil oxidation was  $2.93 \times 10^{14}$ , which was a sig-

nificantly higher frequency factor than other samples. Crude palm oil blends, (33C and 66C) with A values of  $2.74\text{--}4.40 \times 10^{14}$  showed lower oxidation rates than linseed oil. However, it was found that the A value for crude palm oil was substantially lower than its blends (33C and 66C), indicating a lower oxidation rate. The higher reaction frequency of molecules in 33C and 66C may arise from the presence of both the PUFAs in linseed oil and the impurities in crude palm oil, which led to a higher collision and oxidation rate in comparison with crude palm oil (Golmakani *et al.*, 2020b). Similarly, as PUFAs increased in refined palm oil (33R blend), the rate of oxidation increased from  $3.30 \times 10^4$  to  $7.11 \times 10^4$ . Conversely, the oxidation rate decreased from  $7.11 \times 10^4$  to  $4.55 \times 10^4$  due to the lower concentration of PUFAs and higher amount of antioxidants in the 33R blend. However, the oxidation rate of refined palm oil was significantly lower than the 33R blend oil. Therefore, it was revealed that refined palm oil blends (66R blend and 33R blend) were the best blends. The frequency factor values of the oil blends in this study were between  $4.40 \times 10^{10}$ – $4.55 \times 10^4$ , which were lower than the frequency factor of palm olein and olive oil blends reported by Heidarpour and Farhoosh (2018) and sunflower and sesame oil blends reported by Ghosh *et al.* (2019).

Activation energy, a key concept in the Arrhenius equation, is the minimum energy required for a chemical reaction to occur. It is the energy barrier that reactant molecules must overcome to initiate a successful collision and proceed with the reaction. Factors such as the nature of the reactants and the reaction pathway influence the activation energy (Toorani *et al.*, 2019). A higher activation energy means a greater energy hurdle, resulting in slower reaction rates. Conversely, a lower activation energy indicates a lower energy barrier, leading to faster reaction rates. The Arrhenius equation provides insights into how temperature affects reaction rates and the role of energy in chemical reactions.

As shown in Table 4, the energy activation significantly varied [ $106.47\text{--}232.58$  ( $\text{kJ}\cdot\text{mol}^{-1}$ )]. The  $E_a$  of linseed oil was roughly  $106$  ( $\text{kJ}\cdot\text{mol}^{-1}$ ), which was significantly lower than the  $E_a$  of other oil samples. The 66C blend with  $E_a$  of  $156.41$  ( $\text{kJ}\cdot\text{mol}^{-1}$ ) showed that the addition of crude palm oil to linseed oil could significantly improve the oxidation rate. On the other hand, blends of refined palm oil (66R and 33R) showed significantly higher  $E_a$  than crude palm

oil blends, which means that the addition of refined palm oil to linseed oil was a better choice to retard the oxidation rate of linseed oil. The highest energy of activation was found to be  $232.58$  ( $\text{kJ}\cdot\text{mol}^{-1}$ ) for refined palm oil. These findings were consistent with the findings of the induction period analysis. The activation energy values of blends were higher than the  $E_a$  found by Hashemi *et al.* (2016) for soybean and sunflower oil containing tert-Butylhydroquinone (TBHQ) and German chamomile extract.

### 3.3.3. Activated complex theory

Activated complex theory, or transition state theory, is fundamental in chemical kinetics, explaining the mechanism of chemical reactions through a high-energy state called the activated complex or transition state (Toorani and Golmakani, 2022). This theory provides key thermodynamic parameters: enthalpy ( $\Delta H$ ), entropy ( $\Delta S$ ), and Gibbs free energy ( $\Delta G$ ).  $\Delta H$  indicates heat exchange, with positive values for endothermic (heat absorbed) and negative for exothermic (heat released) reactions.  $\Delta S$  measures disorder, with positive values indicating increased disorder and negative values indicating decreased disorder.  $\Delta G$ , combining  $\Delta H$  and  $\Delta S$ , determines reaction spontaneity: negative for spontaneous reactions, positive for non-spontaneous reactions, and zero for equilibrium (Marcus, 1956). These parameters offer insights into the energy and spontaneity of chemical reactions.

The order of  $\Delta H$  for oil samples was as follows: linseed oil > 33C > 66C > crude palm oil > 33R > 66R > refined palm oil. The highest  $\Delta S$  [ $103.37$  ( $\text{kJ}\cdot\text{mol}^{-1}$ )] was found for linseed oil and refined palm oil showed the lowest  $\Delta S$ . At all temperatures, the  $\Delta G$  of blends was higher than the  $\Delta G$  of linseed oil and the order was as follows: linseed oil < 33C < 66C < crude palm oil < 33R < 66R < refined palm oil. Therefore, it can be deduced that the oxidation reaction could occur more easily in linseed oil than in the crude palm oil blends (33C and 66C) and the oxidation reaction was less favorable in refined palm oil and its blends (66R and 33R) in comparison with other oil samples. Referring to the data extracted from the Arrhenius equation and activated complex theory, it can be deduced that blending linseed with palm oil reduced the oxidation of linseed and the role of refined palm oil was bolder than crude palm oil in the reduction of linseed oil oxidation.

The findings of this study demonstrate the novelty of using refined palm oil to enhance the oxidative stability of linseed oil blends, offering a practical and sustainable solution for the food industry. Unlike previous studies which focused on blending linseed oil with oils like corn, canola, or sesame (Golmakani *et al.*, 2020b), this research uniquely highlights the superior performance of refined palm oil due to its absence of impurities, balanced fatty acid profile, and high levels of natural antioxidants such as tocopherols and tocotrienols. The optimized blending ratios (33% and 66%) provide a cost-effective approach to extend the shelf-life of linseed oil-based products, such as margarines, spreads, and salad dressings, without compromising nutritional quality. Compared to similar studies, this work offers key advantages: (1) refined palm oil blends (33R and 66R) exhibit significantly higher oxidative stability than crude palm oil blends and other oil blends reported in the literature, (2) the study identifies the optimal blending ratios for maximum stability, providing practical guidance for industrial applications, and (3) it underscores the critical role of refining in enhancing oxidative stability, offering new insights into the importance of processing methods. These results position refined palm oil blends as a novel and effective solution for improving the stability of PUFA-rich oils such as linseed oil, thus addressing a key challenge in the food industry.

#### 4. CONCLUSIONS

This study investigated the oxidative stability of linseed oil blended with crude and refined palm oil to enhance its resistance to oxidation. The research aimed to assess the impact of blending on fatty acid composition, oxidative stability parameters (PV, AV, and TOTOX), and oxidation kinetics (Tc, Q10, and  $\Delta G$ ) to determine the most stable formulation. The results confirmed that linseed oil, with its high  $\alpha$ -linolenic acid content (55.25%), exhibited the highest susceptibility to oxidation. Conversely, palm oil blends demonstrated significantly improved oxidative stability due to their lower PUFA content and higher levels of palmitic and oleic acids. Among all tested samples, refined palm oil exhibited the lowest PV, AV, and TOTOX value, indicating superior resistance to oxidation. Compared to linseed oil alone, all palm oil blends showed greater stability, with refined palm oil blends offering the best oxida-

tive performance. The oxidative stability index (OSI) revealed that refined palm oil had the longest induction period, highlighting its resistance to oxidation. The stability ranking of the oils and blends followed this order: refined palm oil > 66R blend > crude palm oil > 33R blend > 66C blend > linseed oil > 33C blend. The refined palm oil blends, particularly the 33R and 66R blends, demonstrated significantly higher oxidative stability than crude palm oil blends, further confirming the positive effect of refining. The oxidation kinetics analysis showed that linseed oil had the highest temperature coefficient (Tc) and acceleration factor (Q10), indicating its rapid degradation under heat. Palm oil blends, especially those containing refined palm oil, exhibited the lowest Tc and Q10, signifying better thermal stability. The  $\Delta G$  values suggested that oxidation reactions occurred more readily in linseed oil compared to the palm oil blends. The Arrhenius equation analysis further demonstrated that refined palm oil blends had lower frequency factors (A), indicating a lower reaction rate and, consequently, improved stability. Overall, this study highlights that blending linseed oil with refined palm oil significantly enhances its oxidative stability. The refining process plays a crucial role in removing impurities and improving the stability of palm oil and its blends. The findings suggest that the refined palm oil blends, particularly the 33R and 66R formulations, offer a promising approach for improving the shelf-life and usability of linseed oil in food applications. These results provide valuable insights for the food industry, supporting the strategic formulation of stable, high-quality oil blends suitable for prolonged storage and diverse applications.

#### AUTHORSHIP CONTRIBUTION STATEMENT

M-T. Golmakani: Funding acquisition, Supervision, Project administration.

A. Soltani: Conceptualization, Data curation, Formal analysis, Investigation, Methodology.

S. Sahraeian: Formal analysis, Visualization, Software, Writing – review & editing.

#### DECLARATION OF COMPETING INTEREST

All authors consented to the publication of this work. The authors declare that they have no competing interests.

## ETHICAL STATEMENT

The authors confirm all relevant ethical guidelines have been followed. All methods were used in accordance with relevant guidelines and regulations. There are no human or animal subjects in this article.

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