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Strengthening the quality standards of used edible oils through the incorporation of distillation thyme powder

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ABSTRACT

This study investigates an innovative approach to improving the quality and stability of used frying oils by enriching them with distillation residual thyme powder (DRTP). A series of physico-chemical and thermal analyses were performed to evaluate the effect of DRTP enrichment on essential quality parameters. The results demonstrate that incorporating thyme powder at concentrations of 1% (DRTP), 5%(DRTP), and 10%(DRTP) significantly slowed acid degradation over a 10-day storage period, ensuring that the oils remained within acceptable food quality standards. Furthermore, the enriched oils exhibited superior thermal stability at elevated temperatures compared to both virgin oil (positive control) and untreated used oil (negative control). These findings highlight the potent antioxidant and protective properties of thyme, which not only enhance oil quality but also extend the shelf life of recycled cooking oils. This sustainable strategy paves the way for the effective reuse of used frying oils in the food industry, offering economic and environmental benefits.

Keywords: used frying oils, antioxidants, oil preservation, thermal stability, thyme powder.

INTRODUCTION

Edible plant oil is abundant in triglycerides and comprises fatty acids along with various micronutrients such as tocopherols, phospholipids, sterols, and carotenoids (Sabikhi and Kumar, 2012). It finds extensive applications in the culinary, food, pharmaceutical, and cosmetic industries (Seneviratne et al., 2009). The global demand for vegetable oil exhibited an annual growth rate of 5.14% from 2020 to 2025 (Kai et al., 2015). In comparison to animal oils, vegetable oils are favored by consumers for their renewable nature, high quality, and absence of cholesterol (Fiori, 2009). The production of vegetable oils is contingent upon the yield of oil crops. Over the past three decades, global oil crop production has surged by 240%. Between 2008 and 2018, the leading oilseeds in global production volume were rape, followed by sunflower seed, soybean, and others (Sharma and Gupta, 2006). Anterior studies have indicated that vegetable oil preparation methods primarily encompass organic solvent extraction (Ayas and Yilmaz, 2014; Wong et al., 2014) cold and hot pressing (Snyder et al., 1984; Yazan et al., 2011). It is noteworthy that, with the continuous advancement of technology, novel extraction methods such as supercritical CO_2 fluid extraction (Liu et al., 2009; Tian et al., 2013), solvent-assisted ultrasound extraction (Chan and Ismail, 2009; Eikani et al., 2012), enzymatic-assisted aqueous extraction (Bhosle and Subramanian, 2005; Durmaz and Gökmen, 2019), pulsed electric field extraction (García-Moreno et al., 2013), and microwave-assisted extraction(Abedi et al., 2016), among others, have emerged. In comparison to traditional extraction methods, these innovative techniques offer advantages such as higher oil extraction rates, shorter extraction durations, and lower solvent consumption India stands as the world's largest importer and the third-largest consumer of edible oil, (Serjouie et al., 2010). In 2007, the annual per capita consumption of edible oil in India was reported to be 11 million tonnes and 11.5 kg, respectively (Data source: Ministry of Agriculture, Government of India). The widespread use of oil, particularly for the popular culinary technique of deep-frying, is notable in both industrial and domestic food preparations, contributing to increased fat consumption,(Gertz et al., 2000). Deep frying, a widely embraced gastronomic method in India, involves simultaneous heat and mass transfer during processing. This technique entails immersing the entire food item in hot oil, typically at a temperature around or exceeding 180 °C (Hubbard and Farkas, 1999), as well as (Debnath et al., 2003, 2009). Throughout this cooking process, there is a reverse transfer of water vapor from the food to the oil and eventually to the atmosphere, as elucidated by (Hubbard and Farkas, 1999) repeated use of oil can lead to degradation, resulting in undesirable changes in their physicochemical and nutritional properties, Sunflower oil, soybean oil, palm oil, rapeseed oil, and peanut oil are commonly employed in cooking practices (Sharma and Gupta, 2006). When used in cooking, oil serves to enhance the taste, color, and fragrance of food. However, the elevated temperature and prolonged cooking duration can not only degrade unsaturated fatty acids and essential active substances but also trigger the oxidation of oils, resulting in the formation of primary or secondary oxidation products (Popescu et al., 2019). In traditional culinary preparations, oil plays a key role in crafting vegetable salads, stir-fries, pan-frying, and deep-frying. Vegetable salads typically require a small amount of unheated oil. Stir-frying involves a small quantity of oil but at a high temperature. Pan-frying typically demands slightly more oil than stir-frying, but with lower cooking temperatures and longer durations. In the case of deep-frying, researchers commonly note that food is coated with a batter before being immersed in a substantial volume of oil, heated to around 180 °C (Aydar et al., 2017; García-Moreno et al., 2013; Gibbins et al., 2012). Stir-frying stands out as the most traditional and prevalent method applied in Chinese cuisine (Abedi et al., 2016). These changes not only affect the quality of dishes prepared with these oils but can also have a negative impact on health due to formation of potentially toxic compounds during cooking at high temperatures. In this context, the search for solutions to improve the stability and quality of used oils is crucial. One promising approach is to enrich these oils with plant extracts rich in natural antioxidants Thyme (*Thymus vulgaris*), which is highly valued for its aroma and flavor, is also known for its well-documented antioxidant properties. The distillation residues of thyme essential oils(Angelini et al., 2003; Okamura et al., 1994), often considered as waste products, have an interesting potential as a source of antioxidants to improve the stability of reused edible oils. This study investigates how the enrichment of used oils with thyme powder residues affects their stability and antioxidant activity. For this purpose, a series of physico-chemical and thermal analyses were carried out on samples of virgin oil, used oil and oil enriched with different concentrations of thyme residues. The results obtained will allow us to better understand the effects of this enrichment on the properties of oils and to determine the optimal concentrations to obtain the best results in terms of stability and antioxidant activity. The results of this study could have significant implications for the preservation of reused edible oils, offering a sustainable and natural approach to extending their shelf life while maintaining their nutritional quality. These results could also help to promote responsible use of aromatic plant residues by valuing them as a source of beneficial antioxidants for the food industry. This introduction lays the foundation for the present research by outlining the general context of edible oils, the challenges associated with their repeated use, and the objectives of the study to evaluate the efficacy of fortifying used oils with thyme powder residues to improve their stability and antioxidant activity

MATERIALS AND METHODS

Biological material

The plant biomass used for valorization was derived from the distillation residues of essential oils obtained from *Thymus vulgaris*. This material was sourced from HERBADIS, an industrial unit located in Ain Taoujtate. The exhausted biomass was promptly transported to the Applied Organic Chemistry Laboratory, where it was oven-dried to a constant weight. The dried biomass was then ground in successive cycles to reduce particle size and sieved through a 0.1 mm stainless steel filter to produce a fine powder. The resulting powder was stored in a humidity-controlled chamber for further use.

To address the research objective of restoring the quality of used frying oils, the following experimental design was implemented:

- A negative control batch consisting of untreated used oils.
- A positive control batch consisting of virgin oil.
- Three experimental batches of used oils treated with (DRTP) at concentrations of 1%, 5%, and 10%.

Treated and untreated oil samples were agitated at 100 rpm during the storage period, following the method outlined in [28]. Daily analyses were conducted to monitor the progression of oil quality restoration. Biochemical assessments were performed to evaluate the qualitative and quantitative indices of the oils in compliance with international standards (AOAC).

Biochemical analysis

Acid value

The acid number (AN), which indicates the free fatty acid content resulting from triglyceride hydrolysis, is widely recognized as a key parameter for assessing oil quality and its changes during storage. The acid value (AV) was determined using the standard titration method outlined in AOCS Cd 3d-63 (AOCS, 1999). In this procedure, 5 g of the oil sample were fully dissolved in 50 ml of a previously neutralized ethyl ether/ethanol mixture (2:1, v/v). A 0.05 M ethanolic potassium hydroxide solution served as the standard titrant, and phenolphthalein was used as the indicator. The titration continued until the endpoint was reached, signaled by the persistence of the phenolphthalein color for at least 30 seconds. The acid value was expressed as the milligrams of potassium hydroxide (mg KOH) required to neutralize the free fatty acids in 1 g of the oil sample. The value was calculated using the following formula (Lion, 1955):

$$VA\ (\text{mg/g}) = 56.1 \times V \times c/m \quad (1)$$

where: V is the volume in milliliters of volumetric potassium hydroxide standard solution used; c is the concentration in moles per liter of volumetric potassium hydroxide standard solution used; and m is mass in grams of the test sample of oils.

Iodine index

The iodine index measures the degree of unsaturation in fats by quantifying the amount of iodine (in grams) that reacts with the double bonds in 100 g of lipid. Saturated animal fats typically exhibit iodine indices around 45, while vegetable oils can have values reaching up to 150 (Azoulay, 1968.). To determine the iodine value, a standard titration method was used. A 0.001 g sample was placed in a 250 ml flask, followed by (AOCS, 1999) the addition of 15 ml of carbon tetrachloride and 25 ml of Wijs' reagent. The flask was sealed, shaken, and wrapped in black paper to shield it from light, then allowed to stand for one hour. After this, 20 ml of a 10% potassium iodide solution and 150 ml of distilled water were immediately added to the mixture. Titration was performed using a 0.1 N sodium thiosulfate solution until the yellow iodine color almost disappeared. A few drops of starch indicator were then added, and the titration continued until the blue-violet color completely disappeared, leaving the solution transparent. A blank titration was conducted using the same procedure.

The iodine value was calculated using the following formula:

$$IV = [(V_0 - V)/P] \times 126.9 \times N \quad (2)$$

where: V_0 – volume (in ml) of 0.1 N sodium thiosulfate required for the blank titration; V – volume (in ml) of 0.1 N sodium thiosulfate required for the sample titration; P – weight of the sample (g); N – normality of the sodium thiosulfate solution (0.1 N).

Refraction index

Once the refractometer is calibrated according to the manufacturer's instructions, the measurement of the refractive index begins. A small amount of the sample is collected using a dropper and carefully placed onto the prism of the refractometer. The refractive index value is then displayed directly on the device's screen.

Physical analysis

Viscosity analysis

Before measuring viscosity evolution, the viscometer was calibrated following the manufacturer's guidelines. At room temperature, an appropriate amount of oil was sampled using a flask and injected into the viscometer's measuring chamber. To ensure temperature stability, the viscometer was equipped with a thermometer. The central cylinder, immersed in the oil, was set to rotate, and the oil's resistance to this rotation – proportional to its viscosity – was measured and displayed on the instrument's dial (Manuel d'analyse de La Viscosite Fungilab, n.d.)

Absorbance analysis

Absorbance was measured using a control sample of hexane, set to correspond to 100% transmittance. Measurements were taken over a wavelength range of 200 to 800 nm to assess the optical properties of the sample (Pigni et al., 2010).

Analysis of antioxidant activity

The described protocol is the β-carotene/linoleic acid assay, a method used to evaluate the antioxidant activity of lipid extracts [34]. The procedure is based on the approach outlined by Kelen and Bektas (2008) [35], with modifications such as the use of Tween 80 instead of Tween 40. Below is the step-by-step explanation:

- Preparation of β-carotene solution:
  - Dissolve 2 mg of β-carotene in 1 ml of chloroform to create a β-carotene solution.
- Preparation of reaction mixture:
  - Add 2 mg of linoleic acid and 200 mg of tween 80 to a flask containing the β-carotene solution.
  - Evaporate the chloroform under vacuum at 40 °C, leaving behind a mixture of linoleic acid, Tween 80, and β-carotene.
- Formation of emulsion:
  - Add 100 ml of oxygen-saturated distilled water to the mixture at a flow rate of 30 ml/min over 30 minutes with vigorous stirring to form an emulsion.
- Sample preparation:
  - Take 2.5 ml of the emulsion and transfer it to a test tube.
  - Add 350 µl of the test extract (at 0.5 mg/mL concentration). The test extract is a natural extract potentially containing antioxidant compounds.
- Reference and control:
  - Repeat the same process for a synthetic antioxidant, butylated hydroxytoluene (BHT), at a concentration of 0.5 mg/mL, and include a blank tube as a control.
- Incubation:
  - Incubate the emulsion systems in the dark for 48 hours to prevent light-induced oxidation.
- Absorbance measurement:
  - After incubation, measure the absorbance of each sample at 490 nm using a spectrophotometer.

Calculation of relative antioxidant activity (*RAA*). Use the formula:

$$RAA(\%) = (A\ sample / A\ BHT) \times 100 \tag{3}$$

where: *A sample* – absorbance of the tested extract, *A BHT* – absorbance of the reference antioxidant, BHT.

The relative antioxidant activity (*RAA*) compares the efficacy of the tested extract to that of BHT, a widely used synthetic antioxidant. A higher *RAA* percentage indicates greater antioxidant activity of the extract (Merouane et al., 2015).

Thermal analysis (TGA)

To investigate the thermal properties of the samples, we analyzed mass changes within a temperature range of 100 °C to 500 °C using a Nabertherm furnace. The resulting data produced curves that illustrate the relationship between mass loss and temperature, providing insights into the thermal stability and decomposition behavior of the samples (Nabih et al., 2021).

RESULTS AND DISCUSSIONS

Acidity index

The acidity index (AI) is a critical quality parameter used to measure the concentration of free fatty acids in oil, with lower values indicating better oil quality [38]. Monitoring the evolution of AI during storage provides important insights into the stability of oils, especially those that have been used for frying and subsequently enriched with residual thyme powder. Initially, used frying oil displays a high AI value of 8.5 mg KOH/g, reflecting some degradation from repeated use. However, after the 11th day of storage, the AI shows minimal and stable changes, suggesting that the degradation process has slowed.

Enriching the oil with varying percentages of thyme powder yields notable results. Oil treated with 1% (DRTP) has a lower initial AI (7.82 mg KOH/g), suggesting a beneficial effect on stability. At 5%, a dose-dependent improvement is observed, with a more significant stabilization effect. At 10%(DRTP), the AI is further reduced to 6.65 mg KOH/g, indicating a substantial reduction in oil acidity. The observed stepwise changes in AI after day 5 suggest a sequential antioxidant mechanism, with different antioxidant molecules acting at different stages, which helps reduce acidity and improve stability.

The findings align with Codex Alimentarius standards (1983), which set the acceptable AI range for vegetable edible oils between 2.2 and 7.26 mg KOH/g [39]. The results show that (DRTP) helps bring used oils closer to this standard, supporting its potential for improving the quality of reused oils.

In conclusion, enriching used oils with (DRTP) significantly enhances stability, as demonstrated by the controlled evolution of the AI over time. The stability improvement is dose-dependent, and the sequential reduction in AI confirms that thyme powder is effective in preserving the quality of used oils. These findings emphasize the potential of thyme powder as a sustainable solution for improving the stability and antioxidant capacity of used oils.

Chemical mechanism of enrichment (DRTP) contains antioxidants such as flavonoids and polyphenols. These compounds act by donating electrons to neutralize free radicals generated during the thermal degradation of used oil. Free radicals are reactive molecules that can trigger lipid oxidation, which leads to the degradation of oil. By neutralizing these free radicals, thyme's antioxidants prevent the oxidation of the oil's lipids, preserving its chemical stability.

In addition to preventing oxidation, thyme's antioxidants also neutralize free fatty acids, which contribute to the increase in acidity. This neutralization reduces the oil's acidity, as indicated by the lower AI values in enriched oils.

Thus, enriching oils with thyme powder works chemically by introducing antioxidant compounds that stabilize free radicals, inhibit oxidation, and reduce acidity. These actions collectively preserve the oil's stability, extend its shelf life, and improve its nutritional quality (Figure 1).

Diode index

The iodine value (IV) is a crucial indicator for assessing the degree of unsaturation in oils, which directly relates to their susceptibility to heat and degradation during frying cycles. Oils with higher iodine values contain more double bonds in their fatty acids, making them more prone to oxidation and degradation (Matthäus et al., 2014). The results of this study highlight the significant impact of enriching used oils with thyme powder on their iodine value over time.

Initially, virgin oil has a high iodine value of 95 mg/100g, indicating a substantial proportion of unsaturated fatty acids. However, after undergoing several frying cycles, this value drops to 84.36 mg/100g, reflecting the degradation of double bonds due to oxidation and polymerization under high-temperature conditions. Enriching used oils with (DRTP) at different concentrations leads to noticeable effects on the iodine value, suggesting a protective effect on the unsaturated fatty acids.

- At 1% (DRTP), the iodine value increases gradually from day 10, stabilizing at 88.32 mg/100g by day 15. This indicates that thyme's antioxidants help preserve the unsaturations in fatty acids.

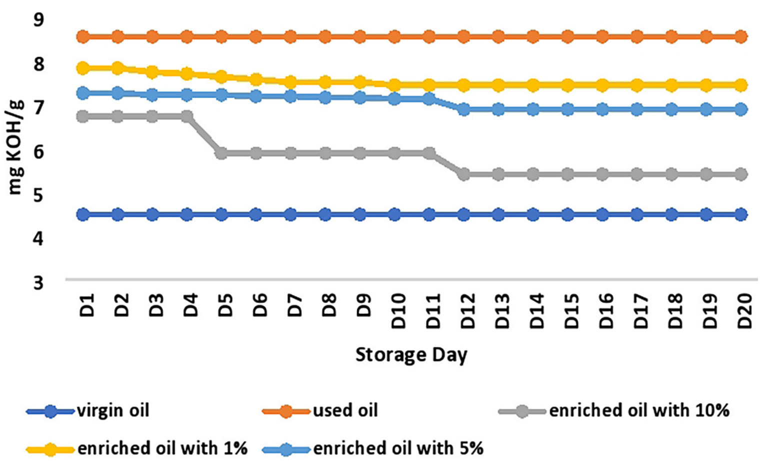

Figure 1. Evaluation of acid value

- At 5% (DRTP), the iodine value rises more significantly, reaching 89.51 mg/100 g by day 10. This shows a more pronounced effect at this concentration, further preserving the unsaturated bonds in the oil.
- At 10% (DRTP), the iodine value stabilizes at 92.26 mg/100g from day 13 onwards, approaching the iodine value of virgin oil. This suggests that the higher concentration of thyme powder provides substantial protection to the double bonds in fatty acids, maintaining the oil's stability.

The results align with Codex Alimentarius standards (1983), which stipulate that the iodine value for vegetable oils should be between 92 and 102 g/100g. Our findings demonstrate that thyme powder enrichment allows used oil to meet these standards, thus improving its quality and stability. The results also echo previous research, such as studies on canola oil enriched with rosemary extract, and on soybean and sunflower oils enriched with olive leaf extract (Zahran et al., 2015), which similarly showed improved preservation of unsaturations in fatty acids, as reflected by an increase in iodine value.

Chemical mechanism of enrichment – the observed changes in iodine value can be explained through the chemical mechanisms underlying the action of (DRTP) antioxidants, such as flavonoids and polyphenols. These compounds possess antioxidant properties that allow them to neutralize free radicals generated during thermal degradation, thus stabilizing the unsaturated fatty acids and preventing further oxidation.

High initial iodine value of virgin oil (95 mg/100g): This indicates a high proportion of unsaturated fatty acids, which are more reactive and susceptible to oxidation during frying cycles.

Decrease in iodine value after several frying cycles (84.36 mg/100g): The reduction in iodine value shows that the unsaturated bonds are breaking down due to oxidation and polymerization during repeated frying, which lowers the oil's quality.

- Effect of 1% (DRTP) enrichment (88.32 mg/100g): The gradual increase in iodine value indicates that thyme's antioxidants are helping to preserve the unsaturated bonds in the oil, likely by neutralizing free radicals and reducing oxidation.
- Effect of 5% (DRTP) enrichment (89.51 mg/100g): The more significant increase in iodine value reflects a dose-dependent effect, with a higher concentration of antioxidants better preserving unsaturation.
- Effect of 10% (DRTP) enrichment (92.26 mg/100g): The stabilization of the iodine value at 92.26 mg/100g suggests that the higher concentration of thyme powder effectively maintains the unsaturated bonds, keeping the oil close to the quality of virgin oil.

Compliance with Codex Alimentarius standards (92–102 mg/100g): The results demonstrate that thyme powder enrichment enables used oil to meet the iodine value standards, preserving unsaturation and improving overall oil stability.

In conclusion, the enrichment of used oils with (DRTP) plays a significant role in preserving the unsaturation in fatty acids, as demonstrated by the increase in iodine value. This preservation of unsaturations enhances the stability of used oils, improving their quality and aligning them more closely with the standards for virgin oils. The antioxidant compounds in (DRTP), such as flavonoids and polyphenols, are key to this process, making thyme powder a promising solution for improving the stability and quality of reused edible oils (Figure 2).

Refractive index

The refractive index is a valuable parameter in evaluating the stability of edible oils, particularly those that have been used for frying and then enriched with (DRTP). The refractive index of oils provides insight into their chemical composition, with higher values often indicating increased oxidation and degradation.

Initial refractive index changes

The refractive index of used oil after frying (1.478) is higher than that of virgin oil (1.463), which aligns with the observation that repeated heating and frying cause oil degradation. The increased refractive index reflects the formation of more oxidized compounds, such as polar compounds and polymers, during frying.

Effect of distillation thyme powder enrichment (DRTP): when oils are enriched with (DRTP), the refractive index decreases progressively over time, indicating improved stability. Specifically:

- At 1% (DRTP), the refractive index decreases from 1.478 to 1.469 by day 14, continuing to 1.465 by day 20, suggesting that thyme powder aids in stabilizing the oil.

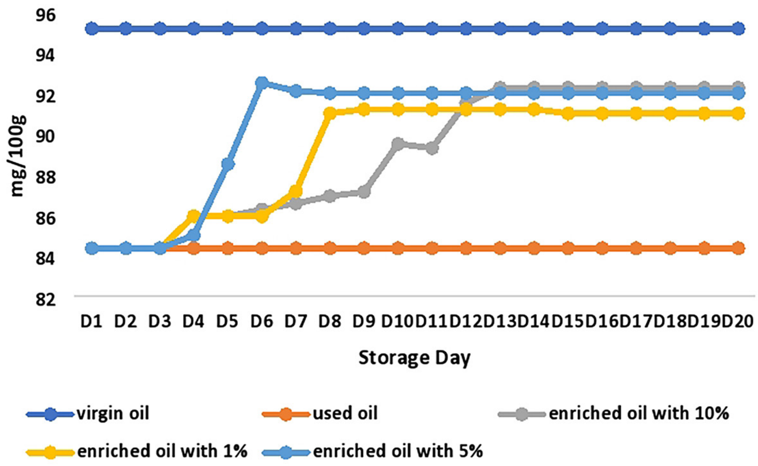

Figure 2. Evaluation of diode Index

- At 5% (DRTP), the refractive index decreases from 1.478 to 1.467 by day 14, with a slight stabilization thereafter.
- At 10% (DRTP), the refractive index decreases to 1.466 by day 16, then stabilizes at 1.465, similar to the values seen with 5% thyme enrichment.

These changes demonstrate the effectiveness of thyme powder in reducing the oxidation and degradation that contribute to the elevated refractive index of used oil.

Compliance with food standards – the Codex Alimentarius (1983) standards stipulate that the refractive index of vegetable edible oils should be between 1.463 and 1.478. The results show that distillation thyme powder enrichment brings the refractive index of used oils closer to the lower end of this acceptable range, improving their quality and making them more compliant with these standards.

Comparison with other studies – these findings are consistent with other research, such as that by [42] on sunflower oil enriched with rosemary extract. Similar studies have shown that enrichment with antioxidant-rich extracts can improve the refractive index, demonstrating the broader applicability of plant-derived antioxidants for improving oil stability.

Chemical mechanism of enrichment – the improvement in the refractive index of used oils enriched with thyme powder can be attributed to the antioxidant properties of thyme's bioactive compounds, including flavonoids and polyphenols. Here's the proposed mechanism:

Antioxidant action – the flavonoids and polyphenols in thyme powder act as antioxidants, which interact with free radicals formed during oxidation. These free radicals are highly reactive species that can damage the oil by initiating chain reactions that lead to lipid degradation.

Free radical neutralization – thyme's antioxidants donate electrons to free radicals, effectively neutralizing them and reducing their reactivity. This process prevents free radicals from attacking the lipids in the oil, which would otherwise lead to oxidation and the formation of unwanted oxidized compounds.

Prevention of lipid oxidation – oxidation in oils is a chain reaction that leads to the breakdown of unsaturated fatty acids and the formation of harmful compounds. By neutralizing free radicals, thyme's antioxidants interrupt this chain reaction, thus reducing the formation of oxidized products and preserving the oil's chemical structure.

Reduction in refractive index – the high refractive index of used oil after frying is often a result of the formation of oxidized compounds that increase the oil's density and change its optical properties. By preventing oxidation, (DRTP) helps reduce the refractive index, bringing the oil closer to its initial or acceptable state.

The study confirms that enriching used oils with (DRTP) offers significant benefits in improving their stability, as reflected by the decrease in refractive index. This process appears to help stabilize the oil, making it more compliant with food standards and extending its usability. The antioxidant compounds in thyme powder play a critical role in this improvement by neutralizing free radicals and preventing oxidation. As a result, enriching oils with (DRTP) not only preserves their quality but also enhances their nutritional value,

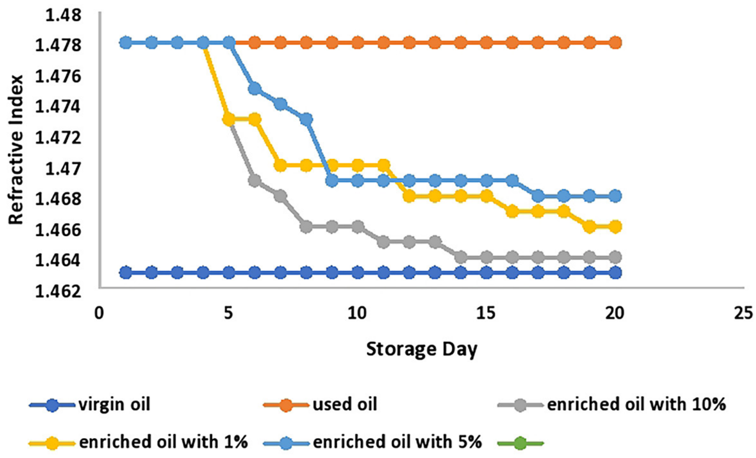

Figure 3. Evaluation of refractive index

making it a promising strategy for improving the quality of reused edible oils (Figure 3).

Viscosity analysis

The viscosity of oils is a key parameter in determining their quality and behavior under different conditions, particularly in vegetable oils. Viscosity provides valuable insights into the flow characteristics and stability of oils during use, especially when subjected to heat or repeated frying cycles. (Che Man and Jaswir, 2000) The study you're describing focuses on how the addition of thyme essential oil distillation residues (at concentrations of 1%, 5%, and 10%) affects the rheological properties (flow behavior) of used oils.

Effect of temperature on viscosity – as expected, the viscosity of oils generally decreases with an increase in temperature. This is a common observation for oils and is due to the decrease in molecular interactions and friction between oil molecules as they gain kinetic energy at higher temperatures.

The study identified two phases of viscosity change:

- Rapid phase – the viscosity decreases rapidly up to around 50 °C.
- Slow phase – the decrease slows down after 50 °C, continuing until 102 °C.

Used oil behavior – the initial viscosity of used oil (7.5 mP/s at 25 °C) was relatively low, and it continued to decrease with rising temperature, indicating the degradation of the oil's rheological properties due to repeated use, oxidation, and polymerization of fatty acids.

Effect of thyme powder enrichment:

- 1% (DRTP) – this concentration slightly improved the viscosity of used oil from 7.5 mP/s to 4.4 mP/s at 25 °C. However, it was not enough to bring the viscosity closer to that of virgin oil, showing only minimal improvement.
- 5% (DRTP) – this concentration resulted in a more significant improvement in the viscosity of the used oil, particularly in the temperature range of 25 °C to 52 °C. After this temperature, the viscosity of the 5% enriched oil began to resemble that of the 1% treated oil, indicating that higher levels of thyme powder are more effective in improving the oil's stability at lower temperatures.
- 10% thyme powder – this concentration showed a marked improvement in viscosity, even surpassing that of virgin oil. The viscosity increased from 8.4 mP/s to 9.4 mP/s at the start, showing an initial advantage over virgin oil. The 10% enrichment maintained superior viscosity performance up to temperatures around 94 °C, where the viscosity of virgin oil dropped below that of the enriched oil.

Temperature-dependent mechanisms

The results suggest the operation of several protective mechanisms at different temperature ranges:

- Low temperature (25 °C to 50 °C) – protective molecules in the thyme powder likely stabilize the oil's viscosity, preventing the rapid degradation typical in used oils.
- Medium temperature (50 °C to 70 °C) – the thyme compounds may act more effectively in

this range, enhancing viscosity by providing additional stability to the oil's structure.

- High temperature (70 °C and above) – at higher temperatures, the antioxidants and other compounds in (DRTP)continue to prevent oxidation and polymerization, helping to maintain the oil's fluidity and prevent excessive degradation.

Chemical mechanisms at play – the enrichment of oils with (DRTP) likely involves multiple chemical mechanisms that contribute to improving the oil's stability and rheological properties:

Antioxidant action (DRTP) – contains flavonoids, polyphenols, and other antioxidants that can neutralize free radicals formed during the degradation of the oil. By preventing the oxidation of unsaturated fatty acids, these antioxidants reduce the formation of oxidized compounds, which could otherwise increase viscosity.

Stabilization of oil structure – the compounds in thyme powder may help stabilize the molecular structure of the oil by preventing the formation of polymers and other high-molecular-weight compounds that increase viscosity. This results in a more fluid and stable oil under heat.

Thermal protection – the thermal stability of the oil is enhanced by the presence of thyme's bioactive compounds. These compounds likely form protective layers or interact with the oil's molecular components to prevent excessive breakdown at higher temperatures, thus maintaining better rheological properties.

Reduction of polymerization – as used oil undergoes repeated frying cycles, it undergoes polymerization, which leads to an increase in viscosity. The antioxidants in thyme powder may help to inhibit polymer formation, thus reducing the increase in viscosity and keeping the oil more fluid.

The study provides compelling evidence that enriching used oils with thyme powder at concentrations of 5% and 10% can significantly improve the oil's viscosity and stability across a range of temperatures. This improvement likely stems from the antioxidant properties of thyme, which prevent oxidation and polymerization, and the presence of compounds that help stabilize the oil's molecular structure at different temperature ranges. The use of thyme powder thus appears to offer a promising strategy to enhance the quality and performance of used oils, bringing them closer to the properties of virgin oils and improving their stability during storage and cooking (Fig. 4).

Antioxidant activity

The β-carotene/linoleic acid test used to assess the antioxidant activity of oil is an effective method for evaluating the oil's ability to protect against oxidation. This test measures the ability of the oil to prevent the discoloration of β-carotene, a natural antioxidant, which is directly related to its capacity to neutralize free radicals and slow down lipid oxidation. The findings of this study underscore the importance of enriching oils with thyme powder, which enhances their antioxidant properties and helps improve the overall quality and stability of reused oils.

Virgin oil – the initial antioxidant activity of virgin oil was low (17%), which is expected since virgin oil typically contains low levels of natural antioxidants compared to oils enriched with plant extracts.

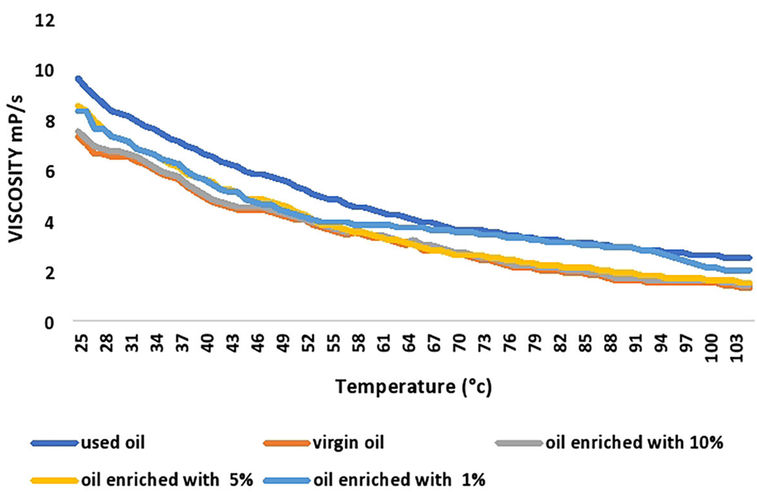

Figure 4. Evaluation of viscosity

After frying – the antioxidant activity of used oil increased to 25% after frying, possibly due to the presence of antioxidants from the food being cooked. However, this increase was modest and did not significantly enhance the oil's overall stability.

- The addition of 1% (DRTP) led to a notable increase in antioxidant activity to 52%, suggesting the first positive effects of thyme's antioxidants on oil stability.
- 5% (DRTP) further increased the antioxidant activity to 64%, highlighting a dose-dependent enhancement.
- At 10% (DRTP), the antioxidant activity reached 76%, approaching the reference value of BHT (96%), a common synthetic antioxidant. This demonstrates thyme powder's significant ability to improve the oxidative stability of used oils.

The study also referenced other oils, such as olive oil, known for its high levels of phenolic compounds [44, 45], which also exhibit significant antioxidant activity. Similarly, thyme powder enriched oils show significant antioxidant potential, which can help in the prevention of lipid oxidation and formation of harmful free radicals.

DRTP is rich in flavonoids, polyphenols, and other antioxidant compounds such as thymol and carvacrol. These compounds neutralize free radicals – reactive molecules that contribute to lipid oxidation.

Antioxidants like flavonoids and polyphenols work by donating electrons to free radicals, thereby stabilizing these reactive species and preventing them from initiating the oxidation process that can degrade the oil.

By neutralizing free radicals, the antioxidants in thyme powder prevent lipid oxidation, which would otherwise lead to the formation of undesirable compounds that could affect the oil's flavor, viscosity, and nutritional value.

The study clearly demonstrates that enriching oils with distillation thyme powder significantly boosts their antioxidant activity, making them more resistant to oxidation and thus preserving their quality over multiple cooking cycles.

This approach could be a promising strategy to improve the stability, safety, and health benefits of reused oils. By enhancing the natural antioxidant content of cooking oils, (DRTP) helps extend their shelf life and prevent the formation of harmful compounds during repeated use.

Further research is recommended to optimize the concentrations of (DRTP), balancing antioxidant activity with the organoleptic characteristics (such as taste, odor, and color) of the enriched oils. This will help ensure the oils remain acceptable to consumers while benefiting from enhanced stability and safety.

In conclusion, the use of (DRTP) as an enrichment in edible oils is a promising and effective method for enhancing antioxidant activity, improving the quality and stability of reused oils (Figure 5).

Thermal analysis

The thermal analysis of oils across a range of temperatures from 0 to 240 °C reveals important insights into the stability of edible oils under cooking conditions. These results are significant for understanding the behavior of oils during

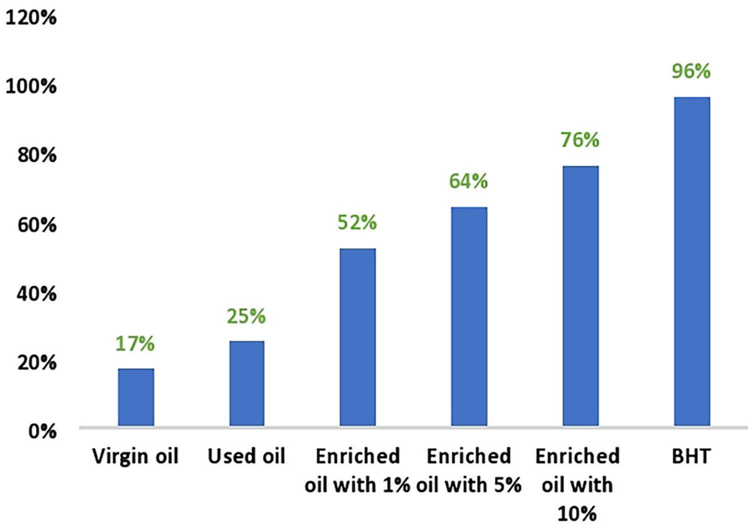

Figure 5. Evaluation of antioxidant activity

heating and their vulnerability to thermal degradation. Here's a breakdown of the findings:

In the 0 to 240 °C range, all oils maintained robust stability, indicating that typical cooking temperatures do not significantly affect the oils' quality. This is reassuring, as most cooking processes fall within this range, ensuring that the oils retain their functional properties during everyday use.

However, beyond 240 °C, a distinctive degradation pattern emerges, particularly with virgin oil. Virgin oil undergoes a 72% degradation at 430 °C, highlighting the heating limitations of this oil. This suggests that while virgin oil is relatively stable at lower temperatures, it begins to break down significantly at higher, extreme temperatures.

Used edible oil demonstrates an even more pronounced 98% degradation at 450 °C, revealing substantial deterioration due to repeated use. This indicates that used oils are more prone to thermal breakdown as they have already undergone changes during previous cooking cycles, making them less stable under high heat.

The introduction of DRTP to used oil shows promising results in mitigating thermal degradation. At a 1% enrichment level, the protective effect is not significant, suggesting that this low concentration of thyme extract does not sufficiently safeguard the oil against thermal damage.However, when the oil is enriched with 5% and 10% (DRTP), there is a clear improvement in thermal stability. At 5% thyme extract, the degradation is reduced to 60% at 420 °C, showing a moderate protective effect. This means that the oil retains more of its integrity at higher temperatures compared to the untreated used oil.

The 10% DRTP shows an even greater reduction in degradation to 56% at 430 °C, highlighting the enhanced protective efficacy of higher concentrations of thyme powder. This suggests that the antioxidant properties of thyme extract play a critical role in preserving the oil's stability by neutralizing free radicals and preventing the thermal breakdown of oil components.

The antioxidants in (DRTP), particularly flavonoids, polyphenols, and thymol, are key to its protective action. These compounds neutralize free radicals that are generated during the heating process, thus preventing the oxidation and polymerization of the oil's fatty acids, which are typically responsible for degradation at elevated temperatures. By donating electrons to the free radicals, thyme's antioxidants stabilize the reactive species and inhibit the chain reaction of oxidation, helping to preserve the structural integrity of the oil even at higher temperatures.

These findings highlight the significant potential of DRTP as a natural preservative for used edible oils, especially in commercial and industrial frying processes where oils are subjected to high temperatures repeatedly.

By enriching oils with thyme powder, it may be possible to extend their shelf life, improve their stability, and reduce the formation of harmful compounds typically associated with repeated heating. This can lead to healthier, more stable oils for use in cooking.

The results could have practical applications in the food industry, where reused oils are common, and protecting oil quality can reduce waste, improve

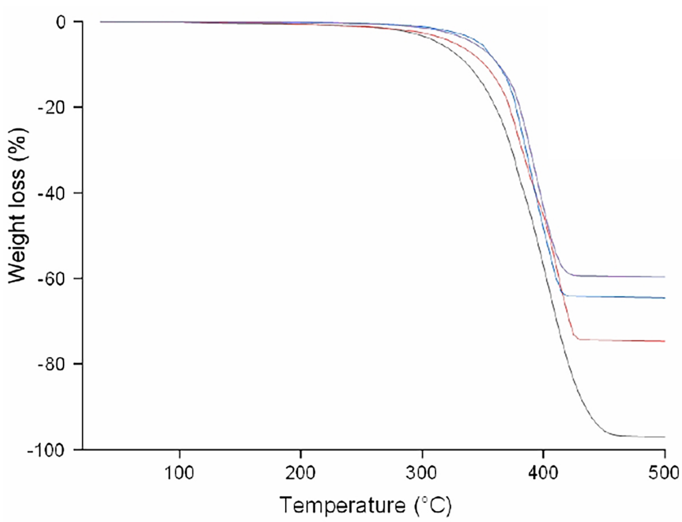

Figure 6. Evaluation of TGA

product consistency, and ensure the safety of cooking oils for both consumers and foodservice operators.

In summary, the study demonstrates that DRTP has a protective effect against thermal degradation in oils, especially when used at 5% and 10% concentrations. The antioxidant properties of thyme effectively reduce thermal damage, preserving the quality and stability of used oils. This makes thyme powder an excellent candidate for enhancing the shelf life and thermal stability of reused oils in cooking and food preparation processes. Further exploration and optimization of (DRTP) concentrations could lead to more widespread applications in food safety and oil preservation (Fig.6).

CONCLUSIONS

The findings of this study underscore the positive impact of supplementing used edible oils with residual thyme powder, presenting a significant potential for enhancing the stability and quality of reused oils. The key takeaways from this research are as follows: The initially high acid number in used frying oil suggests degradation through repeated use. However, the acid number stabilizes after the 10th day of storage, indicating relatively stable degradation. Incorporating rosemary powder reduces the acid number, aligning the oil more closely with food standards. The iodine value decreases over time, signaling the degradation of unsaturations in the fatty acids of reused oils. Enriching with thyme powder preserves unsaturations and enhances oil stability. Used oils exhibit a higher refractive index than virgin oils, indicating degradation during repeated cooking. Enrichment with thyme powder reduces the refractive index, bringing the oils within acceptable values. Oil treated with 10% thyme powder demonstrates a higher viscosity than virgin oil, showcasing the effectiveness of enrichment in restoring the rheological behavior of used oil. DRTP boosts the antioxidant activity of reused oils, safeguarding lipids from oxidation. Oils enriched with 5% and 10% rosemary powder show decreased thermal degradation compared to used oil, indicating improved stability at elevated temperatures. Overall, the results suggest that the addition of residual thyme powder to used oils enhances their stability and quality, concurrently elevating their natural antioxidant content. This approach holds promise for the food industry by extending the shelf life of reused oils, minimizing undesirable compounds, and enhancing their nutritional profile. Further studies could delve into the mechanisms of action of thyme powder and optimize enrichment concentrations to strike a balance between antioxidant activity and the organoleptic characteristics of enriched oils. In conclusion, enriching used oils with residual thyme powder emerges as a promising strategy for enhancing the quality of reused edible oils.

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