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Optimizing the stability and antioxidant potential of used edible oils with rosemary residue powder enrichment

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ABSTRACT

This study explores the potential of enriching used edible oils with distillation residual rosemary powder (DRRP) to enhance their quality and stability. Physico-chemical and thermal analyses were performed to assess the oils' chemical quality indices. The results revealed that enrichment with 1%, 5%, and 10% DRRP significantly minimized acid index degradation, maintaining stability even after 11 days of storage. Additionally, the enriched oils demonstrated improved compliance with food quality standards. Thermal analysis further showed that DRRP enrichment markedly enhanced resistance to thermal degradation at elevated temperatures compared to virgin oil (positive control) and untreated used oil (negative control). These findings underscore the protective properties of DRRP, presenting a promising strategy to extend the shelf life of used oils in the food industry while promoting sustainable practices.

Keywords: used oils, antioxidant activity, chemical quality indices, oil quality, thermal stability, preservation.

INTRODUCTION

Vegetable oil is an indispensable ingredient in the human daily life diet, not only due to its sensory attributes when used as a cooking medium, but also as a supply energy to maintain body normal temperature (Xu et al., 2015). Vegetable oils provide essential fatty acids (EFAs) which serve as carriers for liposoluble vitamins, act as precursors for steroid hormones and prostaglandins synthesis (Li et al., 2016), and play a significant role in health protection and disease prevention. The consumption edible vegetable oils (EVOs) have been rising steadily due to urbanization, per capita income rise, and shifts to obesogenic diets around the world (Joe, 2018). Throughout oil cooking process, there is a reverse transfer of water vapor from food to oil and eventually to atmosphere, as elucidated by (Hubbard and Farkas, 1999) repeated use oil can lead to degradation, resulting in undesirable changes in their physicochemical and nutritional properties, when used in cooking, oil serves to enhance the food, color, and fragrance. However, elevated temperature and prolonged cooking duration can not only degrade unsaturated fatty acids and essential active substances but also trigger the oils oxidation, 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 (Angelini et al., 2003; Okamura et al., 1994). 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 prepared dishes 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 becomes crucial. One promising approach is to enrich these oils with plant extracts rich in natural antioxidants. Rosemary (*Rosmarinus officinalis)* is a plant widely used in cooking for its aroma and taste, but it also has well-documented antioxidant properties. Distillation residues Romain powder, generally considered as waste products, have an interesting potential as a source of antioxidants to improve oils stability. This study focuses on assessing the effect of enriching used oils with residual rosemary powder on their stability and antioxidant activity. To this end, a series of physico-chemical and thermal analyses were carried out on samples of virgin oil, used oil(positive control) and oil enriched with different concentrations of residual rosemary powder (negative control). Obtained results will enable us to better understand the impact of this enrichment on the properties of the oils, and to identify the optimum concentrations for obtaining best results in terms of stability and antioxidant activity. The findings of this study could have important implications for the preservation of reused edible oils, offering a sustainable and natural approach to extending their shelf life while preserving their nutritional quality. In addition, these results could also contribute to promoting a responsible approach to the management of aromatic plant residues, valuing them as sources of beneficial antioxidants for the food industry. With this in mind, this introduction will lay the foundations for the present work by outlining the general context of cooking oils, the problems associated with their repeated use, and the aims of the study to assess the effectiveness of enriching used oils with residual rosemary powder to improve their stability and antioxidant activity.

MATERIALS AND METHODS

Biological material

The plant biomass to be utilised was obtained from the distillation residues of essential oils (*Rosmarinus officinalis*). The plant material was imported from an industrial plant (HERBADIS) in Ain Taoujtate. The depleted biomass of essential oils was quickly transferred to the laboratory of applied organic chemistry. The samples were dried in an oven until they reached constant weight. Thereafter, the biomass was gradually ground in grinding cycles to reduce the particle size. After grinding, a fine powder was retained by sieving through a 0.1 mm stainless steel filter. The powder was stored in a humidity-controlled chamber.

Treatment of waste oil

In order to answer the research question about residual restoration waste oil quality, we developed the following experimental design:

- A negative control batch consisting of used oil.
- A positive control batch consisting of virgin oil.
- Three batches of used oil, when each treated with rosemary powder at the following concentrations: 1%, 5%, 10%. The 15% was discarded as it gave unsatisfactory results.

The treated and untreated oils were initially shaken (100 rpm) during the storage period according to the method developed by (Zhang, 2017). Daily analyses were performed to monitor the evolution of the restitution of the used oil. Against this background, biochemical analyses were carried out to determine qualitative and quantitative indices in accordance with international standards (AOAC).

Biochemical analysis

Acid index

The acid index (AI), a measure of the free fatty acids resulting from triglyceride hydrolysis, is generally regarded as one of the most important parameters for representing oil quality. The acidity values were determined using a standard titration method according to AOCS Cd 3d-63 (AOCS. Acid Value Official Methods and Recommended Practices of the AOCS, 1999). In brief, 5 g of sample was completely dissolved in 50 ml of a previously neutralized ethyl ether/ethanol solution (2:1, v/v). An ethanolic solution of 0.05 M potassium hydroxide was used as a standard reagent until a phenolphthalein endpoint was reached (the phenolphthalein color persisted for at least 30 s). The AI value was expressed in milligrams of potassium hydroxide required to neutralize the free fatty acids present in 1 g of oil sample (mg KOH/g). The AI value was calculated as follows (Lion, 1955):

$$VI\left(\frac{\text{mg}}{\text{g}}\right) = 56.1 \times V \times \frac{c}{m} \quad (1)$$

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

Iodine index

This index measures the overall degree of unsaturation of a fat by determining the number of grams of iodine bound to the double bonds in 100 g of lipid. Highly saturated animal fats have iodine indices of about 45, while vegetable oils have indices of up to 150 (Azoulay, 1968). The acidity index was determined using a standard titration method according to AOCS Cd 3d-63 (AOCS. Acid Value Official Methods and Recommended Practices of the AOCS, 1999). 0.001 g of the sample was placed in a 250-ml flask, and then 15 ml of carbon tetrachloride and 25 ml of Wijs reagent were added to the solution. After settling and shaking, the whole thing was wrapped in black paper and allowed to stand for one hour. Finally, we titrated with 0.1 N sodium thiosulfate solution until the yellow color was almost gone due to the iodine. Add a few drops of starch and continue the titration until the blue-violet color disappears and the solution becomes transparent. we Carry out a blank titration in the same.

Refractive index

The measurement of the refractive index is initiated after the refractometer has been properly calibrated according to the manufacturer's instructions. A small amount of the sample is taken with a dropper and placed on the prism of the refractometer. The index value is then displayed on refractometer screen.

Physical analysis

Viscosity analysis

Before starting the viscosity development, viscometer was calibrated according to the manufacturer's instructions. At room temperature, a sufficient quantity of the product was removed using a piston and injected into viscometer measuring chamber. The viscometer is equipped with a thermometer to ensure temperature stability. The central cylinder is immersed in oil and set in rotation. oil resistance to rotation is proportional to the viscosity, which is displayed on the dial (Azoulay, 1968)

Absorbance analysis

Absorbance was measured against a control (hexane) corresponding to 100% transmittance in a range from 200 to 800 nm (Pigni et al., 2010).

Analysis of antioxidant activity

The protocol described is known as the β-carotene/linoleic acid assay and is used to assess the lipid extracts antioxidant activity (Dapkevicius et al., 1998), The β-carotene/linoleic acid test was carried out according to the method described by (Kelen and Tepe, 2008), with modifications (Tween 80 instead of Tween 40). A detailed explanation of the steps follows: Two milligrams of β-carotene are dissolved in 1 ml of chloroform to obtain a β-carotene solution. 2 mg linoleic acid and 200 mg Tween 80 are added to a flask. The chloroform is evaporated under vacuum at a temperature of 40 °C, leaving a solution containing linoleic acid, Tween 80 and β-carotene. One hundred milliliters of oxygen-saturated distilled water are added to this solution at a flow rate of 30 ml/min for 30 minutes while stirring vigorously. This forms an emulsion. From this new solution, 2.5 ml are taken and transferred to a test tube. Into this tube, 350 µl of extract at a concentration of 0.5 mg/mL is added. The extract may be a natural extract containing antioxidant compounds. The resulting emulsion system is incubated in the dark for 48 hours. For reference purposes, the same process is repeated for the synthetic antioxidant, butylated hydroxytoluene (BHT), at a concentration of 0.5 mg/ml, together with a blank tube as a control. After incubation, sample absorbance is measured at a wavelength of 490 nm using a spectrophotometer. Relative antioxidant activity (RAA) is calculated using the following formula:

$$AAR(\%) = (A\ \text{sample})/(A\ BHT) \times 100 \quad (2)$$

where: A sample is the absorbance of the sample testedand $A\ BHT$ is the absorbance of BHT. Relative antioxidant activity (RAA) is used to compare the efficacy of the extract tested with that of BHT, a synthetic antioxidant commonly used as a reference. A higher percentage of AAR indicates a higher antioxidant activity of the tested extract (Merouane et al., 2015).

Thermal analysis gradient (TAG)

To investigate the thermal characteristics of samples, we examined mass changes over the temperature range from 100 °C to 500 °C using a Nabertherm furnace. The analysis yielded curves describing the progression of mass loss as a function of temperature (Nabih et al., 2021).

RESULTS AND DISCUSSIONS

Acid value

This quality parameter is an indicator of free fatty acids concentration; a low AI value indicates an oil of good quality (De Medina et al., 2011). The Figure 1 illustrates the evolution of acid number as a function of storage time, expressed in days. From this Figure 1 we can see that used frying oil has a relatively high initial acid number of 8.5 mg KOH/g, suggesting that the oil has undergone some degradation during repeated use. However, this acid number shows a minimal and constant evolution over time, indicating that degradation remains relatively stable after the 11th day of storage. Enrichment with residual rosemary powder shows interesting results. enriched oil with 1% residual rosemary powder shows a lower initial acid number than used oil, with a value of 7.3 mg KOH/g. This suggests that the addition of residue rosemary powder has a positive effect on oil stability. enriched Oil with 5 DRRP also shows a lower initial acid number, suggesting a dose-dependent effect. Enrichment with 5%DRRP leads to a greater improvement in oil stability than enrichment with 1%. Used oil treated with 10% DRRP shows an even lower initial acid number, with a starting value of 6.6 mg KOH/g. This indicates that 10% DRRP enrichment has a more pronounced effect on the reduction of used oil acidity than lower enrichment levels. Furthermore, the presence of a stepwise acid evolution number after day 4 of storage suggests a sequential mechanism involving several families of antioxidant molecules. This observation reinforces the idea that the residual rosemary powder acts by neutralizing the fatty acids released over time, helping to reduce the oil's acidity and prolong its stability. It is important to note that according to Codex Alimentarius standards, (Fao, 1983) the acid value of vegetable edible oils should be between 2.2–7.26. In conclusion, enrichment of used oils with residual rosemary powder has a significant effect on oil stability, as demonstrated by the evolution of the acid number over time. The results suggest that the effect is dose-dependent, with a more pronounced improvement in oil stability at higher enrichment levels. The presence of a sequential mechanism involving several families of antioxidant molecules confirms the effectiveness of rosemary powder in neutralizing released fatty acids and maintaining reused oil quality. These findings reinforce the value of enriching used oils with residual rosemary powder to improve their stability and antioxidant value. Compounds present in rosemary distillation residues, such as carnosic acid, carnosol, phenolic acids, flavonoids, and other antioxidants, play an important role in upgrading the quality of used edible oils. The antioxidant compounds present in rosemary residues help stabilize the acid number of used edible oils. Acid index is a measure of the amount of free fatty acids present in oil, which is an indicator of its deterioration. Antioxidants help neutralize free fatty

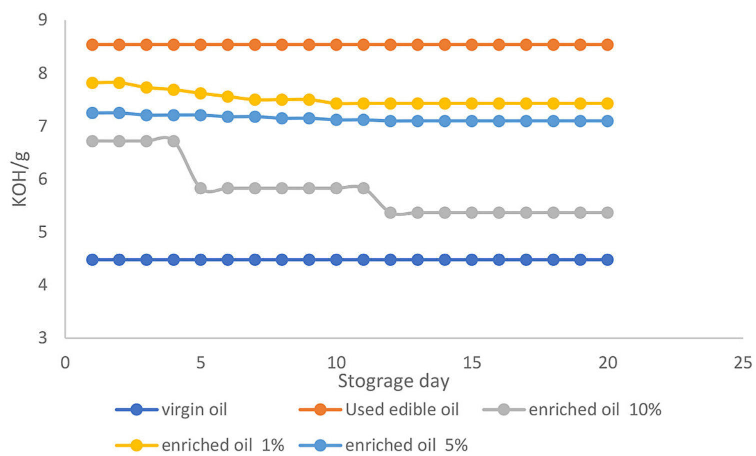

Figure 1. Acid index evaluation

acids, reducing the increase in acid number over time. several reports also showed that some plant extracts are able to delay the lipid oxidation in fish or meat products during refrigerated storage, thus prolonging the shelf life and improving the quality (Benlemlih et al., 2012; Ding et al., 2015).

Iodine value

The iodine value is an important parameter for assessing the amount of unsaturation in oils fatty acids. A high value indicates the presence of more double bonds, making the oil more susceptible to degradation under heat and frying cycles effect. (Matthäus et al., 2014) The results show that virgin oil has a relatively high initial iodine value of 95 mg/100 g. However, after several frying cycles, the iodine value decreases to 84.36 mg/100 g, indicating double bonds degradation in oil's fatty acids, probably due to oxidation and polymerization. For oil treated with 1% of distillation residual rosemary powder, a gradual increase in iodine index was observed from day 9, reaching a value of 87.13 mg/100 g on day 15, which remained constant thereafter. This increase in iodine value suggests that the 1% enrichment helps preserve certain unsaturation's in oil's fatty acids, probably due to the antioxidants present in rosemary powder. For oil treated with 5% rosemary powder, there was also a gradual increase in iodine index until day 14, when it reached a value of 89.46 g/100 g, then stabilized at 91.12 mg/100 g. This more marked increase in the iodine index for the 5% enriched oil indicates that this concentration has a more pronounced effect on the preservation of unsaturation's in the oil's fatty acids. Finally, for the oil treated with 10% DRRP, the iodine index increased until day 10, reaching a value of 87.13 g/100 g, then underwent a more marked increase towards day 13 to reach 92.26 g/100 g, remaining constant thereafter. These results show that enrichment to 10% DRRP preserves more unsaturation in the fatty acids, thus approaching the iodine value of virgin oil. The results obtained in this study are consistent with other research that has investigated the effect of enriching oils with antioxidant-rich plant extracts. Previous studies, such as those conducted by (Matthäus et al., 2014) on canola oil enriched with rosemary extract or (Zahran et al., 2015) on soybean and sunflower oil enriched with olive leaf extract, have also shown a preservation of unsaturation in fatty acids, translated into an increase in iodine value. It is important to note that according to Codex Alimentarius standards (1983), the iodine value of vegetable edible oils must be between 92 and 102 g/100 g. obtained results here show that enrichment with rosemary powder makes used oil more compliant with these standards, by increasing the iodine value and thus preserving a certain amount of unsaturation in the fatty acids. In conclusion, the results concerning the iodine index (Figure 2) highlight the beneficial effects of enriching used oils with residual rosemary powder. Enrichment preserves unsaturation in fatty acids, probably thanks to antioxidants present in rosemary powder. Rosemary extracts, containing phenolic compounds, exert antioxidant abilities via their free radical scavenging abilities to interrupt lipid oxidative chain reaction in food products (Shah et al., 2014). This preservation is reflected in an increase in the iodine value, bringing used oil closer to virgin oil in terms of stability and unsaturated fatty acid composition. These results

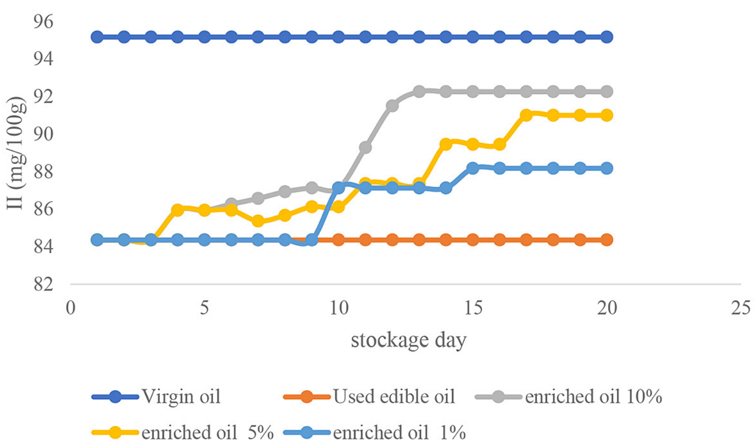

Figure 2. Iodine index evaluation

reinforce the interest of enrichment with DRRP to improve the quality and stability of reused edible oils. Certain compounds, such as carnosol and carnosic acid, are known to preserve unsaturation in the fatty acids of oils. Unsaturation are double bonds in fatty acids that are sensitive to oxidation. By preserving this unsaturation, oils retain their nutritional profile and stability.

Refractive index

The refractive index results show that used edible oil after frying has a higher refractive index (1.478) than virgin oil (1.463). These values remain constant throughout the storage period. This observation is consistent with previous studies cited in the literature, which also found that used frying oils have higher refractive indices than fresh oils. This is often due to oils degradation during repeated cooking. For oil enriched with 1% and 5% of DRRP, the refractive index starts at 1.478, then gradually decreases to 1.468 (for 1% DRRP) and 1.466 (for 5% DRRP) on the 16th day of storage. These values remain constant thereafter. This gradual decrease in refractive index suggests that enrichment with these percentages of rosemary powder has a beneficial effect on oil stabilization, probably due to the antioxidants provided by rosemary, such as flavonoids and phenolic derivatives, which block oil oxidation for oil enriched with 10% DRRP, refractive index decreases steadily throughout the storage days, from 1.478 to 1.466 on day 16, then stabilizes. These gradual decreases, leading to an index value close to that of virgin oil, clearly demonstrates the effectiveness of 10% DRRP enrichment in improving oil stability. It is important to note that according to the Codex Alimentarius standards (1983), the refractive index of vegetable edible oils must be between 1.463 and 1.478 The results obtained here show that enrichment with DRRP makes used oil more compliant with these standards, bringing it closer to acceptable refractive index values. obtained results in this study are consistent with other similar research carried out on oil enrichment with rosemary extracts. Previous work, notably by (Mioara VARGA, 2017) on sunflower oil enriched with rosemary extract during frying, has also shown improvements in refractive index, corroborating the benefits of enrichment for oil stability, so results concerning the refractive index of oils enriched with residual rosemary powder clearly demonstrate the benefits of this technique for improving used oils stability. Enrichment reduces the refractive index and makes the oils more compatible with food standards. The beneficial effect of rosemary powder on oil stabilization is supported by the presence of natural antioxidants in rosemary, which protect oils against oxidation. These results reinforce the interest of enrichment with residual rosemary powder to improve the quality and nutritional value of reused edible oils. The compounds present in rosemary distillation residues can help improve the rheological properties of used oils. For example, the increase in viscosity and stabilization of refractive index due to enrichment with antioxidant compounds may indicate an improvement in oil stability (Figure 3).

Viscosity analysis

fixed oils viscosity gives a very clear idea of its quality (Che Man and Jaswir, 2000a) It is an essential rheological factor in the management of fluid substances such as vegetable oils. Extensive

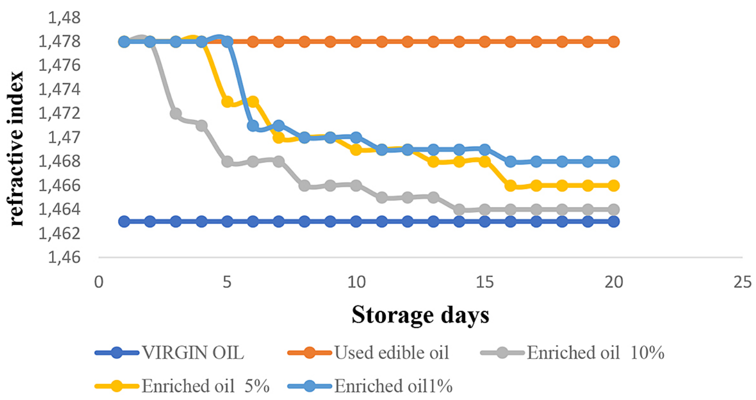

Figure 3. Refractive index evaluation

research confirms that temperature significantly influences fluid products viscosity, leading to a decrease in viscosity as temperature increases (Singh et al., 2009). The viscosity of fixed oils gives a very clear idea of their quality (Che Man and Jaswir, 2000). This notion was introduced in this study to investigate the impact of adding DRRP. tested concentrations were (1%DRRP, 5% DRRP, 10% DRRP). Generally speaking, the fall in viscosity is divided into two phases, a rapid phase which ends on average at 52 °C, and a slow phase which ends at 105 °C. used oil in our study had a minimum viscosity (7.2 mP/s) at the start, and gradually decreased until it reached a final value that was always the lowest. Used oil treated with 1% DRRP starts with a value of 7.8mP/s) and 25 °C and stabilizes at a value (4.3 mP/s), at which point it can be seen that the addition of 1% DRRP does not sufficiently improve used oil viscosity and still remains lower than virgin oil. used oil treatment with 5% DRRP resulted in a marked improvement in used oil rheological behavior over a temperature range from 25 °C to 52 °C, after which treated oil with 5% DRRP joined treated oil with 1% DRRP, Treating used oil with 10% DRRP significantly improved the rheological behavior of used oil, whose viscosity became much higher than that of virgin oil (8.4 mP/s to 9.4 mP/s) at start-up. At this level, rapid phase became longer (from 25 to 70 °C). During this temperature range, the used oil, supplemented with 10% of the residual distillation powder, has a higher viscosity than virgin oil. After 70 °C, the treated and virgin oils have the same viscosity values, and after 94 °C virgin oil viscosity is lower than that used oil treated with 10% DRRP. These results are consistent presence of these phases (2 for used and treated oil and 3 for virgin oil) shows that there are several oil protection mechanisms, molecules that protect at low temperatures, molecules that protect at medium temperatures and molecules that protect at high temperatures. This Table 1 confirms the logarithmic nature of viscosity behavior as a function of temperature. The high values of the weighting coefficients confirm mathematical models validity obtained from the experimental results The leading coefficient of the mathematical model for used oil has a value of -3.89, higher than that for virgin oil. The addition of 1% DRRP and 5% DRRP doses significantly increased this coefficient, in parallel with the constant of the two models (6.21 and 6.86). The addition of 10% DRRP residual powder reduced the model coefficient to below that of virgin oil, of course with a further reduction in model coefficient to match that of virgin oil. From this result (Fig. 4 and Table 1) we can notice that 10% of the residual powder from the distillation of the rosemary plant significantly restores used oil. rheological character.

Antioxidant activity

The choice of using the β-carotene/linoleic acid test to assess the antioxidant activity of oil is suitable for only substance. This test is widely recognized for assessing antioxidant ability to protect lipids against oxidation, as it measures the oil's ability to inhibit β-carotene discoloration, a natural antioxidant. Antioxidant activity of virgin and used edible oil: The results show that virgin oil has a low percentage of antioxidant activity (17%), indicating that it contains limited levels of natural antioxidants. Afte frying process, antioxidant activity increases slightly to 25%, which could be attributed to antioxidants present in the vegetable matrices used for cooking. Effect of enrichment with (DRRP): the results clearly show that enrichment of treated oils with residual rosemary powder progressively increases antioxidant activity. The addition of just 1% DRRP results in a significant 36% increase in antioxidant activity, and this increases further with the addition of 5% DRRP, reaching 54%. Finally, the antioxidant activity of oils enriched with 10% DRRP reaches 64%, approaching the value of the reference antioxidant, BHT (96%). These results clearly demonstrate that the enrichment process with residual rosemary powder is crucial for

Table 1. Mathematical model of oils

Viscosity	Mathematical model	Weighting factor
Used oil (negative control)	$Vuo = -3.89\ln(T) + 20.06$	$R^2 = 0.96$
1% DRRP	$V1\% = -0.07 \ln(T) + 6.22$	$R^2 = 0.94$
5% DRRP	$V5\% = -0.08 \ln(T) + 6.87$	$R^2 = 0.90$
10% DRRP	$V10\% = -4.78\ln(T) + 24.25$	$R^2 = 0.98$
Virgin oil (positive control)	$Vvo = -4.26\ln(T) + 20.93$	$R^2 = 0.99$

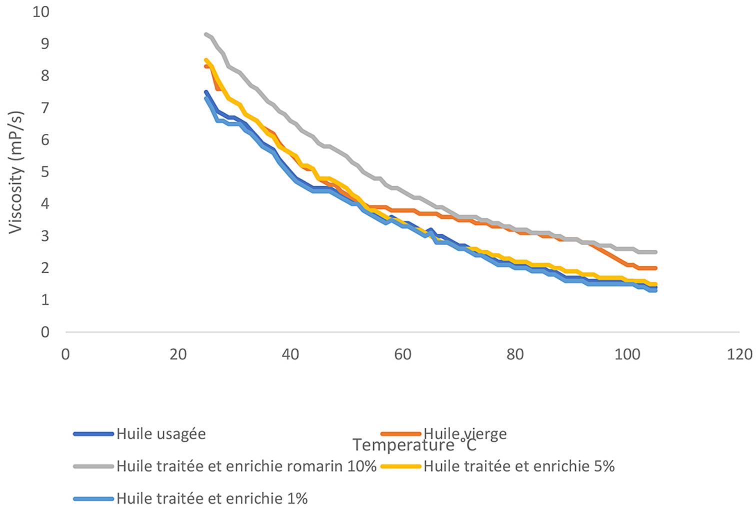

Figure 4. Viscosity analysis

increasing the antioxidant activity of reused oils. These results are related to antioxidant activity examined by the β-carotene bleaching test of the Turkish varieties Halhalı, Eğriburun, Haşebi, Karamani and Saurani ranges from 10 to 75%, of which the Saurani variety has the highest effect (Arslan and Schreiner, 2012). The antioxidant activity of olive oil is due to its richness in antioxidants, particularly phenolic compounds (Benlemlih et al., 2012), which can exert a significant antioxidant effect even in vivo (Bonanome et al., n.d.). This significant increase in antioxidant activity can help protect lipids from oxidation and prevent the formation of harmful free radicals during repeated cooking. Enriching reused oils with DRRP represents a promising approach to improving their quality and stability, while increasing their natural antioxidant content. This could help extend their shelf life, reduce the formation of undesirable compounds and improve their nutritional profile, making reused oils safer and healthier for consumption. In conclusion, the results obtained highlight the effectiveness of enriching edible oils with DRRP to increase their antioxidant activity. These results suggest that enrichment may be a promising strategy for improving the quality of reused oils and making them richer in natural antioxidants, thus contributing to their safer and more beneficial use in culinary practices. However, further studies may be needed to better understand the mechanisms underlying antioxidant activity of DRRP, and to optimize enrichment concentrations to achieve a balance between antioxidant activity and the organoleptic characteristics of enriched oils. Compounds present in rosemary distillation residues, such as carnosic acid, carnosol, phenolic acids, flavonoids, and other antioxidants, play an important role in upgrading the quality of used edible oils. The compounds present in rosemary distillation residues are rich in natural antioxidants. Antioxidants work by neutralizing free radicals and preventing the oxidation of fatty acids and other sensitive compounds in edible oils. This slows down the degradation of used oils, prevents the formation of undesirable compounds and prolongs their shelf life (Figure 5).

Thermal analysis gradient

The results obtained from thermal analysis gradient (TAG) studied oils as a function of temperature (Fig. 6). The findings are highly instructive, revealing essential information about the oil's stability under different conditions. Firstly, it is reassuring to note that all the oils are stable between 0 and 250 °C, indicating that they are resistant to the relatively high temperatures commonly used in cooking. However, above 250 °C, the oils show differential degradation, with treated oils behaving differently from control oils. This highlights the importance of understanding how applied treatments can influence oil stability. The results for virgin oil are remarkable. It undergoes a significant degradation of 70% of its matter at a temperature of 430 °C. This information is crucial to understanding the heating limits of this oil. Used edible oil, on the other hand, undergoes even greater degradation, reaching 96% at a temperature of 450 °C. This indicates that repeated use of the oil results in considerable degradation of its components. However, the introduction of

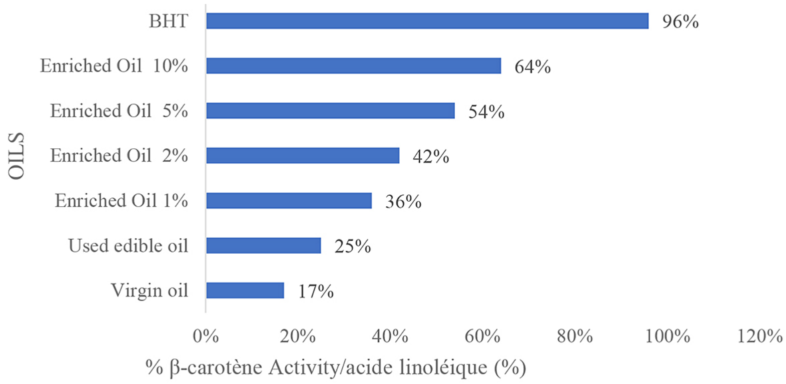

Figure 5. Antioxidant activity

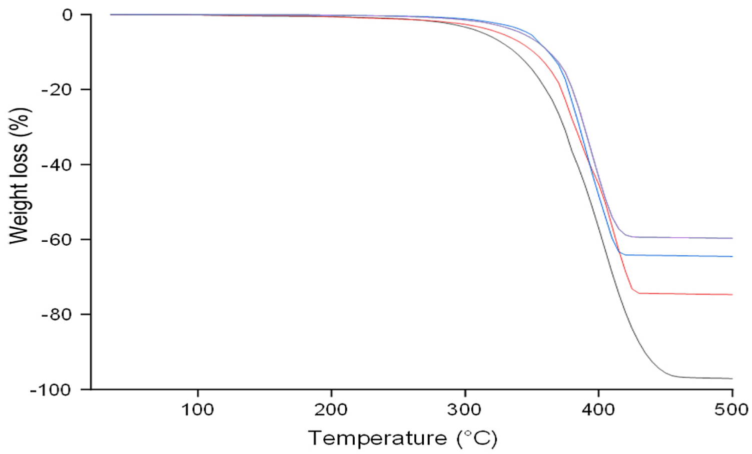

Figure 6. TAG analysis

rosemary powder extract into used edible oil is showing promising results. When the oil is enriched to 1%, it offers no significant protection against degradation, but at concentrations of 5% and 10%, rosemary powder extract appears to act as an effective protective agent. The 5%-enriched oil shows a degradation of only 60% at 430 °C, while the 10%-enriched oil shows an even lower degradation of 58%. This shows that enrichment with rosemary extract at these concentrations can actually preserve used edible oil against thermal degradation at high temperatures. It would be interesting to explore the underlying mechanisms behind the protective efficacy of rosemary extract at these concentrations. Rosemary's antioxidant properties could play a key role in this protection by neutralizing free radicals and preventing oil components degradation. In summary, these results highlight the importance of choosing appropriate methods for oil preservation, and underscore the potential of rosemary powder extract as an effective protective agent for preserving used oils stability. These findings could have practical applications in the food industry to improve the shelf life and quality of oils used in cooking.

The antioxidant properties and stabilization of used edible oils through enrichment with rosemary compounds can help bring them into compliance with food standards in terms of acidity index, iodine index and other quality parameters. In short, the compounds present in rosemary distillation residues act as antioxidant and protective agents, helping to preserve the quality and stability of used edible oils. By enriching used oils with these compounds, it is possible to extend their useful life, reduce the effects of degradation and make the oils more suitable for safe and beneficial culinary use.

CONCLUSIONS

The results obtained in this study highlight the beneficial effect of enriching used edible oils with residual rosemary powder. This technique has significant potential for improving the stability and quality of reused oils. The main conclusions of this study are as follows: The high initial acid number of used frying oil suggests degradation during repeated use. However, the acid number stabilizes after the 11th day of storage, indicating relatively stable degradation. Enrichment with rosemary powder reduces the acid number and makes the oil more compliant with food standards. The iodine value decreases over time, indicating a degradation of unsaturations in the fatty acids of reused oils. Enrichment with rosemary powder preserves unsaturations and improves oil stability. Used oils have a higher refractive index than virgin oils, indicating degradation during repeated cooking. Enrichment with rosemary powder reduces the refractive index, bringing the oils closer to acceptable values. Used oil treated with 10% DRRP has a higher viscosity than virgin oil, demonstrating the effectiveness of enrichment in restoring the rheological behavior of used oil. Enrichment with rosemary powder increases the antioxidant activity of reused oils, protecting lipids against oxidation. Oil enriched with 5% DRRP and 10% DRRP showed reduced thermal degradation compared to used oil, indicating better stability at elevated temperatures. Overall, the results suggest that adding residual rosemary powder to used oils improves their stability and quality, while increasing their natural antioxidant content. This approach may benefit the food industry by extending the shelf life of reused oils, reducing undesirable compounds and improving their nutritional profile.

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REFERENCES

1. Abedi, E., Sahari, M. A., Barzegar, M. & Azizi, M. H. (2016). Designing of high voltage electric field for soybean and sunflower oil bleaching. *Innovative Food Science and Emerging Technologies, 36*. https://doi.org/10.1016/j.ifset.2016.06.015
2. Angelini, L. G., Carpanese, G., Cioni, P. L., Morelli, I., Macchia, M. & Flamini, G. (2003). Essential oils from Mediterranean Lamiaceae as weed germination inhibitors. *Journal of Agricultural and Food Chemistry, 51*(21). https://doi.org/10.1021/jf0210728
3. AOCS. (1999). Acid value Official methods and recommended practices of the AOCS. 5th Ed. Champaign, IL.
4. Arslan, D. & Schreiner, M. (2012). Chemical characteristics and antioxidant activity of olive oils from Turkish varieties grown in Hatay province. *Scientia Horticulturae, 144*. https://doi.org/10.1016/j.scienta.2012.07.006
5. Aydar, A. Y., Bagdatlioglu, N. & Köseoglu, O. (2017). Effect of ultrasound on olive oil extraction and optimization of ultrasound-assisted extraction of extra virgin olive oil by response surface methodology (RSM). *Grasas y Aceites, 68*(2). https://doi.org/10.3989/gya.1057162
6. Benlemlih, M., Ghanam, J. & Joyeux, H. (2012). Polyphénols d'huile d'olive, trésors santé! polyphénols aux actions antioxydantes, anti-inflammatoires, anticancéreuses, anti-vieillissement et protectrices cardio-vasculaires. *MedicatriX*.
7. Bonanome, A., Pagnan, A., Biffanti, S., Opportuno, A., Sorgato, F., Dorella, M., Maiorino, M. & Ursini, F. (1992). *Effect of dietary monounsaturated and polyunsaturated fatty acids on the susceptibility of plasma low density lipoproteins to oxidative modification*. http://ahajournals.org
8. Che Man, Y. B. & Jaswir, I. (2000). Effect of rosemary and sage extracts on frying performance of refined, bleached and deodorized (RBD) palm olein during deep-fat frying. *Food Chemistry, 69*(3), 301–307. https://doi.org/10.1016/S0308-8146(99)00270-8
9. Che Man, Y. B. & Jaswir, I. (2000b). Effect of rosemary and sage extracts on frying performance of refined, bleached and deodorized (RBD) palm olein during deep-fat frying. *Food Chemistry, 69*(3), 301–307. https://doi.org/10.1016/S0308-8146(99)00270-8
10. Dapkevicius, A., Venskutonis, R., Van Beek2, T. A. & Linssen3, J. P. H. (1998). Antioxidant activity of extracts obtained by different isolation procedures from some aromatic herbs grown in Lithuania. In *J Sci Food Agric 77*.
11. Ding, Y., Wang, S. Y., Yang, D. J., Chang, M. H. & Chen, Y. C. (2015). Alleviative effects of litchi (*Litchi chinensis* Sonn.) flower on lipid peroxidation and protein degradation in emulsified pork meatballs. *Journal of Food and Drug Analysis, 23*(3), 501–508. https://doi.org/10.1016/J.JFDA.2015.02.004
12. Ed. Azoulay, Paris. (1968). Manuel d'analyse des corps gras.
13. Fao. (1983). Alinorm 99/17 joint FAO/WHO food standards programme codex alimentarius commission twenty-third session report of the sixteenth session of the codex committee on fats and oils.
14. García-Moreno, P. J., Guadix, A., Gońez-Robledo, L., Melgosa, M. & Guadix, E. M. (2013). Optimization of bleaching conditions for sardine oil. *Journal of Food Engineering, 116*(2). https://doi.org/10.1016/j.jfoodeng.2012.12.040
15. Gibbins, R. D., Aksoy, H. A. & Ustun, G. (2012).

Enzyme-assisted aqueous extraction of safflower oil: Optimisation by response surface methodology. *International Journal of Food Science and Technology, 47*(5). https://doi.org/10.1111/j.1365-2621.2012.02940.x

16. Hajji Nabih, M., El Hajam, M., Boulika, H., Hassan, M. M., Idrissi Kandri, N., Hedfi, A., Zerouale, A. & Boufahja, F. (2021). Physicochemical characterization of cardoon "cynara cardunculus" wastes (Leaves and stems): A comparative study. *Sustainability (Switzerland), 13*(24). https://doi.org/10.3390/su132413905
17. Hubbard, L. J. & Farkas, B. E. (1999). Method for determining the convective heat transfer coefficient during immersion frying. *Journal of Food Process Engineering, 22*(3). https://doi.org/10.1111/j.1745-4530.1999.tb00481.x
18. Joe, P. (2018). Global edible vegetable oil market trends. *Biomedical Journal of Scientific & Technical Research, 2*(1). https://doi.org/10.26717/bjstr.2018.02.000680
19. Kelen, M. & Tepe, B. (2008). Chemical composition, antioxidant and antimicrobial properties of the essential oils of three Salvia species from Turkish flora. *Bioresource Technology, 99*(10), 4096–4104. https://doi.org/10.1016/J.BIORTECH.2007.09.002
20. Li, X., Kong, W., Shi, W. & Shen, Q. (2016). A combination of chemometrics methods and GC-MS for the classification of edible vegetable oils. *Chemometrics and Intelligent Laboratory Systems, 155*. https://doi.org/10.1016/j.chemolab.2016.03.028
21. Lion Ph. (1955). *Travaux pratiques de chimie organique*.
22. Matthäus, B., Pudel, F., Chen, Y., Achary, A. & Thiyam-Holländer, U. (2014). Impact of canolol-enriched extract from heat-treated canola meal to enhance oil quality parameters in deep-frying: A comparison with rosemary extract and TBHQ-fortified oil systems. *JAOCS, Journal of the American Oil Chemists' Society, 91*(12). https://doi.org/10.1007/s11746-014-2561-8
23. Merouane, A., Noui, A., Medjahed, H., Nedjari Benhadj Ali, K. & Saadi, A. (2015). Activité antioxydante des composés phénoliques d'huile d'olive extraite par méthode traditionnelle. *International Journal of Biological and Chemical Sciences, 8*(4), 1865. https://doi.org/10.4314/ijbcs.v8i4.45
24. Ghiduruş, M., Varga, M. (2017). Natural antioxidants used in frying oils to minimize the accumulation of toxic compounds. *AgroLife Scientific Journal, 6*.
25. Okamura, N., Haraguchi, H., Hashimoto, K. & Yagi, A. (1994). Flavonoids in Rosmarinus officinalis leaves. *Phytochemistry, 37*(5). https://doi.org/10.1016/S0031-9422(00)90434-5
26. Paul Singh, R., Heldman, D. R., New York, L. & Diego San Francisco, S. (2009). *Introduction to Food Engineering, Fourth Edition*.
27. Pigni, N. B., Berkov, S., Elamrani, A., Benaissa, M., Viladomat, F., Codina, C. & Bastida, J. (2010). Two New alkaloids from *Narcissus serotinus* L. *Molecules, 15*(10). https://doi.org/10.3390/molecules15107083
28. Popescu, A., Stoian, E. & Şerban, V. (2019). Oil seeds crops cultivated area and production in the EU-28-trends and correlations, 2008–2018. *Scientific Papers Series Management, Economic Engineering in Agriculture and Rural Development, 19*.
29. Sánchez De Medina, V., Priego-Capote, F., Jiménez-Ot, C. & Luque De Castro, M. D. (2011). Quality and stability of edible oils enriched with hydrophilic antioxidants from the olive tree: The role of enrichment extracts and lipid composition. *Journal of Agricultural and Food Chemistry, 59*(21). https://doi.org/10.1021/jf2020528
30. Shah, M. A., Bosco, S. J. D. & Mir, S. A. (2014). Plant extracts as natural antioxidants in meat and meat products. *Meat Science, 98*(1), 21–33. https://doi.org/10.1016/J.MEATSCI.2014.03.020
31. Xu, H. Y., Zhu, L. R., Dong, J. E., Wei, Q. & Lei, M. (2015). Composition of Catalpa ovata seed oil and flavonoids in seed meal as well as their antioxidant activities. *JAOCS, Journal of the American Oil Chemists' Society, 92*(3). https://doi.org/10.1007/s11746-015-2595-6
32. Zahran, H. A., El-Kalyoubi, M. H., M. M., K., And & Abdel-Razek, A. G. (2015). Improving oils stability during deep-fat frying using natural antioxidants extracted from olive leaves using different methods. *Middle East Journal of Applied Sciences, 5*(1).
33. Zhang, Y. (2017). Comparison of antioxidant activity of rosemary extract in peanut oil and flaxseed oil in relation to their fatty acids compositions. *International Journal of Food Processing Technology, 4*(2). https://doi.org/10.15379/2408-9826.2017.04.02.02