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Introducing wild almond oil as a healthier alternative for frying purposes

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ABSTRACT

Wild almond (*Amygdalus scoparia*) is an underutilized source of oil with potential for use in many food applications. In the current study, the applicability of this oil was investigated as a frying medium for par-fried potatoes. Free fatty acid (FFA) content, peroxide value (PV), *p*-anisidine value (PAV), conjugated diene (CD) content, and total polar (TP) compounds of the oil were monitored over a 16-h deep-fat frying process. FFA level changed from 0.04 to 0.21%, PV from 1.0 to 3.5 meq O_2/kg oil, PAV from 3.0 to 35.3%, CD content from 0 to 26 mmol/L, and TP content from 2.0 to 22.0%. However, none of these quality parameters exceeded their required standard levels for frying oil. Overall, wild almond oil showed great potential to be used as a frying oil. Further studies are needed to fully discover such potential under different conditions applied for the various fried products.

Keywords: Oxidative stability; Physicochemical properties; Potato strips; Quality parameters; Smoke point

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1. Introduction

Many processed foods may undergo a light frying stage before it is served by the consumers. Deep-fat frying has also been paid exclusive attention to a wide range of food products. Type and quality of the oil used for frying, the applied temperature, the length of time for the frying and the level of antioxidants added to the oil can impact the stability of the oil during the frying (Keshavarz and Moslehishad, 2020). As the demand for the oil supply is increasing, finding new sources of oil is a mandate to meet such demands.

Wild almond (*Amygdalus scoparia*), which is grown in Iran and a few surrounding countries, is a novel source of edible oil. Previous studies from the current laboratory have considered different aspects of the extraction process and application of the extracted oil from wild almond in different food products. A relatively higher ratio of monounsaturated fatty acids (MUFA) to polyunsaturated fatty acids (PUFA) in the wild almond oil (WAO) (3.24-3.86) (Hojjati et al., 2016) makes this oil more stable against oxidative stresses.

Studies on different aspects of frying are still of major interest among Food scientists. In a recent work, the physicochemical properties and oxidative stability of corn oil were investigated in infrared-based and hot air-circulating cookers (Kim et al., 2022). Considering the application of the frying oils, the oxidative stability of fish crackers after frying in a blend of soybean and tea seed oils was investigated over a 12-week period (Prabsangob and Benjakul, 2019). Also, the changes in the physicochemical properties of the oil blend after 8 h of frying over 3 d of frying process were evaluated.

The objectives of this study were to investigate the applicability of hexane-extracted WAO as a potential source of frying oil. The changes in the physicochemical properties including iodine value (IV), free fatty acid (FFA) content, saponification number (SN), peroxide value (PV), *p*-anisidine value (PAV), totox value, conjugated dienes (CD) value, refractive index (RI), polar compounds, oxidative stability, smoke point, and color, as well as the stability of the WAO, were investigated over a 16-h deep-fat frying process at 170 °C.

2. Material and Methods

A general overview of the process applied in the current study is provided in Fig. 1. Wild almond was obtained from the Fars province of Iran and stored in a cold room at 4 °C before the oil was extracted. To extract oil, pulverized wild almond kernels were soaked in cold *n*-hexane as solvent for 4 h at ambient conditions, after which the residue was re-extracted once more (for about 2 h) using fresh solvent and then combined with the first extract, and then total solvent was separated using a vacuum rotary-evaporator. The extracted oil was stored in dark containers at -18 °C until used in the

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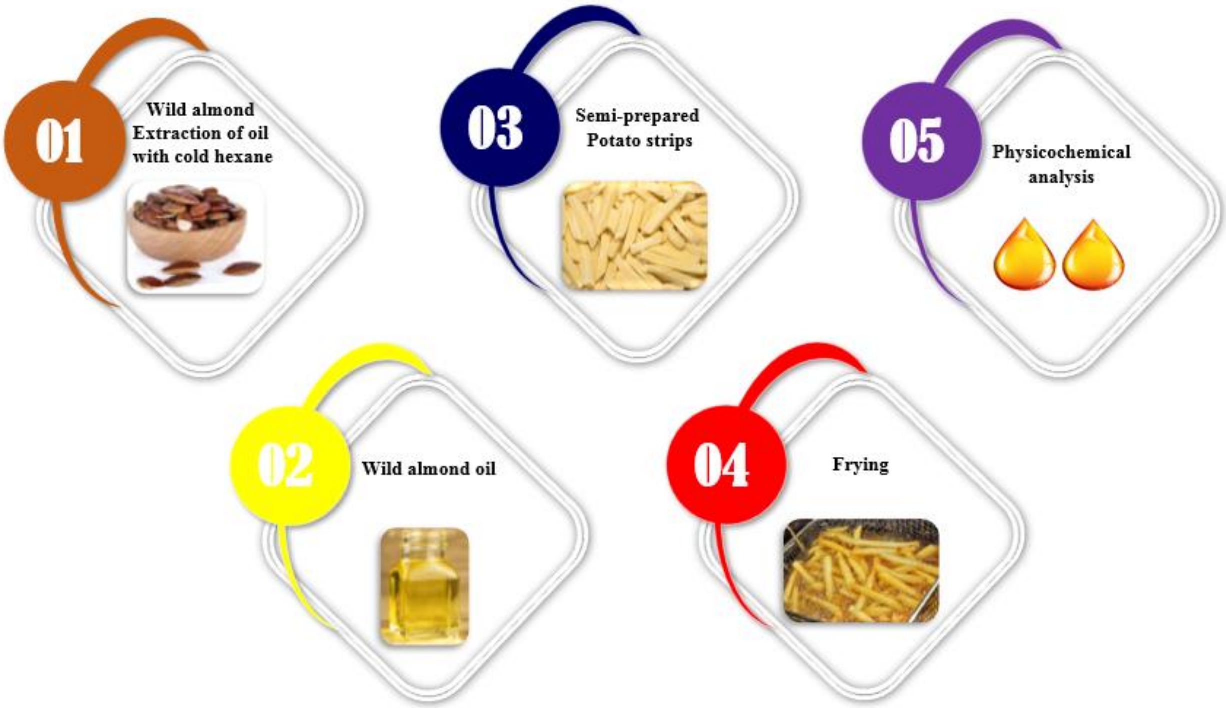

Fig. 1. General overview of the procedures applied in the current study.

experiment. The reference oil was a combination of sunflower, soybean, and canola oils and palm olein and palm super olein, which was purchased immediately after production and stored at -18 °C in a dark place until they were used in the frying process. All chemicals used in the present study were of analytical grade and purchased from Merck Chemical Company (Darmstadt, Germany).

2.1. Frying process

A model F13235 electric fryer from Delonghi (Italy) equipped with a thermostat and a stainless-steel lattice basket was used for frying. WAO (1.3 L) was poured into the fryer and the temperature was adjusted to 170 °C. For each frying stage, 40 g of the semi-prepared potato strips from Paris Food Company (Mobarakeh, Isfahan, Iran) was fried at 170 °C for 7 min. The frying process was continued for 16 h consecutively and using a metallic syringe, 80 g of oil was taken every 2 h and after cooling it under ambient conditions it was flushed with a flow of nitrogen gas and maintained at -18 °C in a dark place until analytical measurements were conducted. During the frying process, no fresh oil was added to the fryer.

2.2. Determination of fatty acid profile

A modified method from Jung and Choi (2016) was used to determine FA compositions of the oil samples. To prepare the methyl esters of FA, one drop of oil and 2.0 mL of 1.0 N methanolic potassium hydroxide were mixed and incubated at 50°C for 30 min. Then, to terminate the trans-esterification reaction, 1.0 mL distilled water was added along with 2.0 mL hexane to recover the methyl ester components and the mixture was shaken and allowed to stand for several min so that the aqueous and organic layers were separated from each other. Part of the upper (hexane) layer was taken and dried by anhydrous sodium sulfate (0.2 g). A gas chromatography system (7890A, Agilent Technologies Inc., Wilmington, DE) equipped with a CPSill-88 column (100 m × 250 µm id × 0.2 µm film thickness) and a flame ionization detector was used to determine FA composition of the oil samples. Both injector and detector temperatures were set at 250 °C. The oven temperature started at 190 °C, which was held for 10 min, and then rose (at a rate of 5 °C/min) to 235 °C, where it was held for 10 min before the analysis was stopped.

2.3. Acid value

The acid value (AV) of the oil samples were measured according to the procedure described by Prabsangob and Benjakul (2019) with some modifications. For each sample, 7.0 g of oil was mixed with 75 mL ethanol and titrated (in the presence of phenolphthalein) by 0.1-N NaOH solution. AV was determined (in mg NaOH/g oil) using Eq. (1).

$$\mathrm{AV}=\frac{\mathrm{mL\ NaOH} \times \mathrm{N} \times 28.2 \times 1.99}{\mathrm{g\ of\ sample}} \tag{1}$$

where N is the normality of the NaOH solution.

2.4. Peroxide value

The procedure reported by Jeon et al. (2016) with some modifications was used to measure PV of the oil samples. For each test, one drop of oil was dissolved in 50 mL of a mixture of acetic acid and *iso*-octane (2:3 ratio, v/v, respectively). After adding 0.5

mL potassium iodide solution (saturated) followed by stirring for 1.0 min and adding 30 mL water, it was titrated by 0.1 N sodium thiosulfate solution until clear yellow solution was reached. Then, after adding 0.5 mL sodium dodecyl sulfate solution (10%, w/v) and 0.5 mL starch reagent and seeing the blue color, the titration was extended until such blue color disappeared. Titration of the blank solution was performed in the absence of an oil sample. PV was determined using Eq. (2):

$$\mathrm{PV}= \frac{(\mathrm{S}-\mathrm{B}) \times \mathrm{N} \times 1000}{\text{g of sample}} \tag{2}$$

where S is the volume of the sodium thiosulfate solution (in mL) for the sample and B is that of the blank. N is the normality of thiosulfate solution.

2.5. p-Anisidine value and Totox value

PAV was determined using the method reported by Kim et al. (2021) with some modifications. For each analysis, 2.0 g oil was placed in a 25-mL volumetric flask and *iso*-octane was added to the mark. Then, in two different test tubes, one tube was poured 5.0 mL of oil in *iso*-octane as the solvent, and in the other tube 5.0 mL of solvent without the oil. For each tube, 1.0 mL PAV solution (0.25 g reagent dissolved in 100 mL glacial acetic acid) was added and shaken. The PAV was determined after the reaction was over by incorporating the spectrophotometric absorbances of the sample (As) and blank (Ab) at 350 nm in Eq. (3), where m is the weight of the oil sample. The Totox value was determined by adding the PAV and twice the PV (Veronezi & Jorge, 2018).

$$\mathrm{PAV} = \frac{25 \times (1.2\ \mathrm{As}-\mathrm{Ab})}{\mathrm{m}} \tag{3}$$

2.6. Total polar compounds and conjugated diene value

The amount of total polar compounds was determined according to the method reported by Kmiecik et al. (2018) with slight modifications. For each sample, 500 mg oil was poured into a 5.0-mL volumetric flask and was taken to the volume using toluene, and after thorough mixing, 1.0 mL was placed on a silica column and it was eluted by three consecutive volumes (1.0, 3.5 and 3.5 mL, respectively) of a mixture of *iso*-hexane and *di*-*iso*-propyl ether at 85:15 ratio (v/v, respectively). The solvent was then evaporated using a flow of nitrogen gas and total amount of polar compounds (C_p) was determined using the following equation:

$$\mathrm{C_p}= \frac{\mathrm{Ws}-\mathrm{Wn}}{\mathrm{Ws}} \tag{4}$$

where W_s is the weight of initial sample and W_n is the weight of the nonpolar part.

The CD value was measured according to the method reported by Hwang et al. (2019) with slight modifications. The oil samples were diluted at a ratio of 1:100 (v/v) using *iso*-octane and then their absorbance levels were recorded using a spectrophotometer at 234 nm.

2.7. Oxidative stability

The oxidative stability was measured based on the method reported by Patra et al. (2022) using a Rancimat 679 Metrohm instrument (Herisau, Switzerland). For each sample, 2.5 g oil was used at 110 °C and the air was flown at a rate of 20 L h^{-1}. The oxidative stability was reported as the length of time (h) required for the major oxidation to start.

2.8. Iodine value

The IV was measured according to the method applied by Rincón et al. (2019) with slight modifications. For each sample, 500 mg oil was dissolved in 10 mL chloroform and after adding 25 mL Wijs solution (one molar iodine monochloride solution in acetic acid), the mixture was placed in dark for 30 min. Then, 10.0 mL of a 15%-potassium iodide solution was added and after mixing, 100 mL distilled water was added and it was titrated using 0.1-N sodium thiosulphate in the presence of 1% starch solution in water until the blue color disappeared. The IV was determined using Eq. (5):

$$\mathrm{IV}= \frac{[(\mathrm{B}-\mathrm{S})\times \mathrm{N}\times 12.69]}{\text{Sample weight (g)}} \tag{5}$$

where, B is the volume (mL) of sodium thiosulphate solution required for the titration of the blank and S is that for the sample. N is the normality of sodium thiosulphate solution. The procedures explained by Patterson (2011) was applied to determine the theoretical iodine value of wild almond and also that of the reference oil based on the obtained FA composition data.

2.9. Saponification number

The SN was determined according to Zamanhuri et al. (2020). In a 250-mL Erlenmeyer flask, 5.0 g oil was mixed with 50 mL 1-M ethanolic potassium hydroxide solution (56.1 g of potassium hydroxide in one liter ethanol) and connected to a condenser and after refluxing for 60 min, titration was performed using a 0.5-N hydrochloric acid solution. The SN was obtained using Eq. (6):

$$\mathrm{SN}= \frac{(\mathrm{B}-\mathrm{S})\times 28.05}{\mathrm{w}} \tag{6}$$

where B is the volume (mL) of hydrochloric acid used for the blank and S is for the oil sample. W is the weight of the oil sample (in g) used for the test.

2.10. Refractive index and Lovibond color

RI was determined according to AOCS Cc 7-25 method (AOCS, 2017) using a model G refractometer from Abbe (Carl-Zeiss, Germany). The Lovibond color was measured according to AOCS Cc 13e-92 method (AOCS, 2017) using a Lovibond tintometer (Model F, Greenwich, England).

2.11. Statistical approaches

Data analysis was performed using SPSS version 19 software. All tests were carried out in triplicate and the data were reported as mean ± standard deviation. The differences between the measured parameters of fresh WAO and those of the fresh reference oil were evaluated using a one-way analysis of variance using Duncan's mean comparison test at a confidence level of 95%.

Table 1. Major characteristics of wild almond oil (WAO) and those of a reference frying oil used in this study

Parameters	WAO	The reference oil
Free fatty acid content (%)	0.041±0.001^{a}	0.076±0.002^{b}
Iodine value (Determined by Hanus method)	91.3±2.6^{a}	80.3±2.1^{b}
Iodine value (Determined using fatty acid profiles of the oil samples)	96.2±0.1^{a}	83.8±2.1^{b}
Saponification number	188±4^{b}	204±5^{a}
Peroxide value	1.06±0.15^{b}	1.50±0.20^{a}
P-anisidine value	3.33±0.15^{a}	3.20±0.14^{b}
Totox value	5.45	6.20
Conjugated diene value (mmol L^{-1})	0.30±0.02^{b}	0.50±0.03^{a}
Refractive index	1.466±0.000	1.463±0.000
Total polar compounds (w/w)	2.33±0.13^{b}	3.10±0.17^{a}
Oil stability (h)	16.0±0.3^{b}	18.0±0.4^{a}
Smoke point (°C)	234.7±2.6^{b}	241.6±2.9^{a}
Soap	Insignificant	Insignificant
Color		
Red Lovibond	3	3
Yellow Lovibond	32	20
Blue Lovibond	0	1

$^{a,\,b}$: different letters in each row indicate a significant difference between the means ($p < 0.05$).

3. Results and Discussion

Table 1 presents the major characteristics of WAO and those of the reference oil used for the frying process. The FFA levels were 0.041% for WAO and 0.076% for the reference oil. According to Barrera-Arellano et al. (2018), the desired level of FFA in frying oils is 0.03-0.05%. FFAs disrupt the frying process due to the reduced smoke point and increased foaming level in the oil (Sarwar et al., 2016). Despite its low FFA content, WAO has a lower smoke point than the reference oil (Table 1). During the frying, FFAs can also be converted into new FFAs due to the hydrolysis (Yahya et al., 2019). In addition, FFAs reduce the surface tension between the air and the oil allowing more oxygen to enter the oil and increase the oxidation rate of the oil (Sahasrabudhe et al., 2019).

The PAV is another parameter that indicates the rate at which the production of secondary oxidation products (aldehydes, ketones, and alcohols) is formed from the primary oxidation products (Nagy et al., 2016). Therefore, the value of this index can be utilized to evaluate the progress of the oxidation in the oils. The PAV for WAO is slightly greater than what was measured for the reference oil (Table 1). The yellow color of Lovibond is somewhat indicative of the amount of the aldehydes in the oil (Strieder et al., 2017). Thus, the oxidation of WAO has progressed more than that in the reference oil due to the decomposition of primary oxidation products into their by-products.

The level of total polar compounds in WAO (2.33%, w/w) was at a lower level than that of the reference oil (3.10%). Polar compounds are not limited to aldehydes and ketones (Kasprzak et al., 2020).

The RI value can be utilized to monitor the development of the isomerization in an oil (Ayorinde et al., 2019). It is declined when the *cis* isomers are converted into *trans* isomers (Mikheev and Ershov, 2019). Freshly extracted WAO and the reference oil were not significantly different ($p < 0.05$) in some of the initial measured parameters, such as SN (Table 1). The amounts of soap in both oils were negligible (Table 1).

Table 2 provides the measured FA profiles for both WAO and the reference oil. Oleic acid is the major FA in both oils. However, the amount of oleic acid in WAO (68.45%) is much higher than that in the reference oil (40.09%). After oleic acid, linoleic acid in WAO and palmitic acid in the reference oil are the second major FAs. The reference oil with 34.91% SFA was expected to be more resistant to heat and oxidation than WAO with 12.47% SFA. According to a study by Melo et al. (2019), samples of a mixture of sunflower oil and tiger-nut oil had the highest thermal stability due to the high levels of SFA. However, it should be noted that the SFA level is not the only criterion for the thermal stability of the oil. The type of UFA and their unsaturation levels can impact the oxidation rate. The presence of natural antioxidants, especially α-tocopherol, can stabilize oils containing UFA, which are more susceptible to thermal degradation (Wang et al., 2019).

Table 2. The fatty acid profiles of WAO and the reference oil used in the current study.

Fatty acids	WAO	Reference oil
Myristic acid	BT	1.24±0.69
Palmitic acid	9.79±0.17	29.39±2.30
Palmitoleic acid	0.78±0.13	BT
Stearic acid	2.68±0.18	4.29±0.81
Oleic acid	68.45±0.71	40.09±1.44
Linoleic acid	18.31±0.86	22.24±0.97
Linolenic acid	BT	2.52±0.59
SFA	12.47±0.01%	34.91±2.17%
UFA	87.54±0.02%	64.84±1.82%
MUFA	69.23±0.83%	40.09±1.44%
PUFA	18.31±0.86%	24.75±0.38%
The ratio of UFA to SFA	7.02±0.01	1.86±0.17
The ratio of MUFA to SFA	5.55±0.06	1.15±0.11
The ratio of MUFA to PUFA	3.79±0.22	1.62±0.03

MUFA: monounsaturated fatty acids; PUFA: polyunsaturated fatty acids; SFA: saturated fatty acids; UFA: unsaturated fatty acids.
BT: Below the GC threshold level.

MUFA contributes to about 69.23% of the FA in WAO (Table 2), which is at much higher level than that in the reference oil (40.09%). Based on the data presented in Table 2, the ratio of MUFA/PUFA for WAO and the reference oil were at 3.79 and 1.62 levels, respectively. Due to the somewhat higher level of PUFA in the reference oil, a higher CD value was also expected (Table 1). However, it is not yet possible to know (based on such data) whether WAO has appropriate thermal and oxidative stabilities compared to the reference oil.

3.1. Changes in the quality attributes

Results of the epidemiological studies show that the excessive use of oils containing SFA can lead to cardiovascular diseases (Silva et al., 2019). Accordingly, based on the current study (Table 2), WAO seems to be a more suitable oil for the frying process due to its lower SFA than the reference oil specified here. Arslan et al. (2017) reported that the formation of FFA in the blend of sunflower and palm oils during 8 h of the frying process was lower than that obtained for pure palm oil. But, the formation patterns of polar compounds in both oils were similar. In addition, the degradation of tocopherols, which have antioxidative properties, in the ordinary sunflower oil was more pronounced than that in the modified sunflower oil. They specifically mentioned that it was not possible to comment on the thermal stability of an oil by relying solely on its FA composition. The following sections consider the changes occurring in the 16-h of deep-fat frying process for WAO as compared to a reference frying oil.

3.2. FFA level

Fig. 2 shows the pattern of changes in the FFA level in WAO during 16 h of frying at 170 °C. According to these data, the amount of FFA increases throughout the entire frying process. Such changes are consistent with the findings of other studies as follows. Manzoor et al. (2022) observed that AV increases during 20 and 25 h of frying in mustard oil. Such observations have also been reported for the rapeseed oil, high-oleic sunflower oil and cottonseed oil by Xu et al. (2020). Ajala and Ghavami (2020) showed that increasing the frying time leads to an increase in the FFA level. Song et al. (2017) justified that part of the changes in the acid content during such frying conditions was due to the presence of carbonyl groups in the polymeric or oxidative products. According to the standards available for the disposal time of the frying oils (Keshavarz and Moslehishad, 2020), WAO from this study did not exceed the limits proposed by such standards after 16 h of exposure to heat at 170 °C.

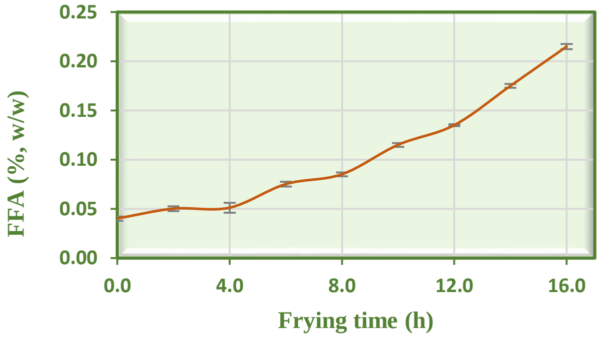

Fig. 2. Changes in the free fatty acid (FFA) content in the wild almond oil (WAO) over 16 h of frying in this study.

3.3. Peroxide value

The changes in the PV for WAO during the 16 h of frying at 170 °C are shown in Fig. 3. The PV increased for the first 8 h of the frying process, after which no such changes were observed. Most studies related to the stability of the oil indicate an increasing trend in the production of peroxide compounds as the frying continues. Shanker and Debnath (2019) reported that the PV of soybean oil increased for the first 18 h when used in a deep-fat frying process at 173 ± 2 °C followed by a decrease after that. Arslan et al. (2017) reported an increase in the PV in the first 4 h of heating for cottonseed oil and the first 6 h of heating for palm oil. However, no changes were observed in the PV at the latest stages of the heating process. According to Codex Alimentarius (2017), the maximum level of PV allowed for frying vegetable oils is 5 meq O_2/kg of oil. For the WAO, the highest level of PV was obtained at ~3.9 meq O_2/kg oil (Fig. 3), which was within the limit allowed for the frying oils. The PV was ~3.6 meq O_2/kg oil at the end of the frying.

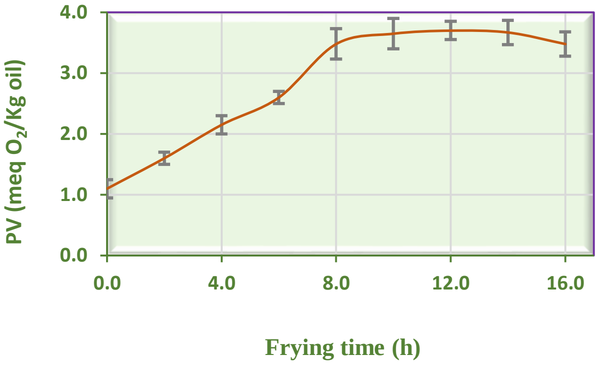

Fig. 3. Changes in the peroxide value (PV) of WAO over a 16-h of frying.

3.4. The p-anisidine value

Fig. 4 shows the changes in the PAV of WAO over the 16 h of frying in the current study. A consistent increase in the PAV is observed during the entire frying process for 16 h. The rate of this increase was slightly higher after the 10th h of frying when the PV started to level off or decrease (Fig. 3). Due to the instability of peroxide compounds, the extent of oxidation cannot be estimated solely on the basis of changes in the PV. Therefore, the use of a complementary parameter that measures the amounts of such compounds resulting from the breakdown of the peroxides can be helpful. PAV is such a parameter for measuring the levels of the secondary compounds in the oxidation process due to the breakdown of hydroperoxides (primary oxidation products). The point at which the steepening of the PAV (Fig. 4) and the flattening of the PV (Fig. 3) occurs in the frying process with WAO (i.e., the 8th-10th h) can be considered a point to be investigated further as a disposal point of this oil. As was the case in the current study, De Boer et al. (2018) observed that exactly at the time when the PV of frying oil started to decline, the PAV started to increase at a higher rate.

Liu et al. (2021) reported that the changes in the PAV of soybean oil during a deep-fat frying process followed an increasing linear pattern and its value reached 115.7 after 10 h of continuous heat at 170 °C. But the PAV levels were 43 and 77 for palm olein and corn oil, respectively. The PAV level of 35.0 for WAO in this study over a continuous heating at 170 °C can be considered a positive criterion for this oil. Xu et al. (2020) reported PAV levels of 91.0-107.5 for rapeseed oil, 49.8–67.8 for high-oleic sunflower oil and 89.7–128.7 for cottonseed oil after 12 h of frying.

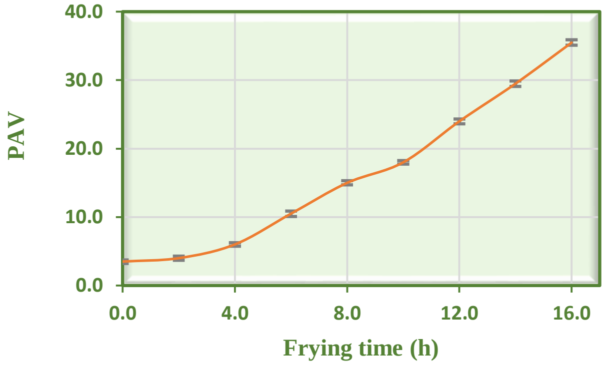

Fig. 4. Changes in the *p*-anisidine value (PAV) of WAO over 16 h of frying.

3.5. Changes in the polar compounds and conjugated diene value

Fig. 5 shows the changes in the level of polar compounds in WAO during 16 h of frying process at 170 °C. As frying proceeded, the level of polar compounds was also increased and reached 24% after such a period. According to Padovan et al. (2020), the maximum frying temperature is 180 °C and the maximum percentage of polar compounds should be between 24 and 27%. Therefore, the obtained values from the current study over 16 h of frying at 170 °C was below such maximum levels. However, Mba et al. (2016) reported that the level of polar compounds in the canola oil heated for 16 h at 190 °C exceeded the allowed limit for these compounds. For fresh oils, the typical amounts of polar compounds are suggested to be within 0.40 and 6.4% (Mba et al., 2016). For WAO in this study, the level of polar compounds in the oil before the frying was 2.3% (w/w), which is within the acceptable range specified above. In a study by Tarmizi et al. (2019), thermal stabilities of four types of frying oils were investigated and the results showed that the amounts of initial polar compounds in these oils varied from 3.7 to 7.5%.

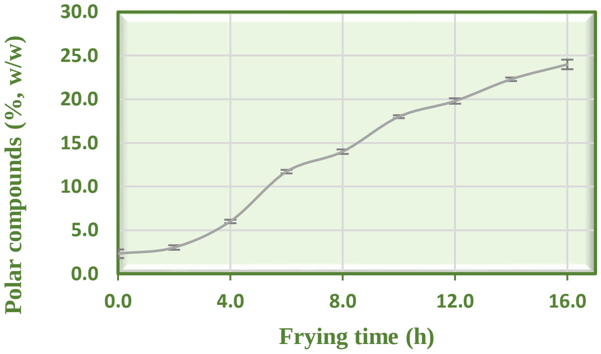

Fig. 5. Changes in the level of polar compounds in WAO during 16 h of frying.

Polar compounds include a set of chemicals that arise from the oxidation, polymerization and hydrolysis of FFA, mono- and di-acylglycerols, aldehydes, ketones, dimers, trimers etc. (Zhou et al., 2019). All the parameters influencing the rates of these reactions during the frying process can lead to differences in the levels of these compounds for different oils.

Changes in the level of CD produced in WAO during the 16 h of frying at 170 °C are shown in Fig. 6. As the frying proceeded, the amount of CD was also increased to a value as high as 27 mmol/L. Isomerization followed by the production of CDs may occur after the separation of hydrogen from the carbon chain in the fats having two or more double bonds (Wang et al, 2020). These compounds may then form hydroperoxides and later break down into secondary oxidation products or enter polymeric reactions from various pathways to form high-CD polymers (Hashem et al., 2020). The value of CD for WAO was obtained at 27 mmol/L after 16 h of frying at 170 °C.

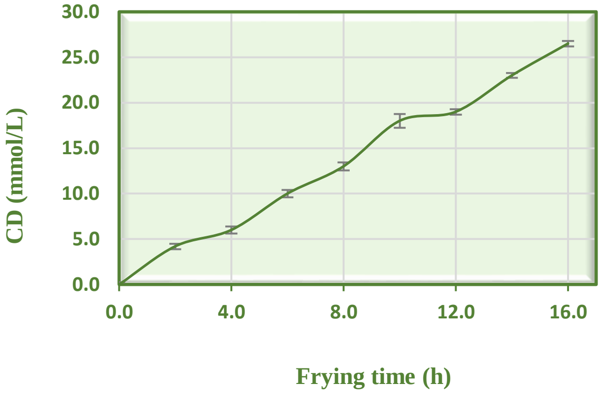

Fig. 6. Changes in the level of conjugated dienes (CD) in WAO over 16 h of frying.

4. Conclusion

WAO was evaluated for its nutritional as well as physicochemical properties and after getting positive feedback as to its potential for use as a frying medium, its properties were evaluated over a 16 h of deep-fat frying process. All measured parameters (such as FFA level, CD value, PAV) increased as the frying continued except for the PV, which increased only for the first 8 h of the process, after which it appeared to be decreasing. The results showed that despite major differences in the FA profiles (compared to a commercial frying oil), wild almond oil can be used as a frying oil without major adjustments in the overall oil properties. A lower level of SFA in the wild almond oil makes this oil a healthier alternative for frying purposes.

Conflict of interest

The authors declare that there is no conflict of interest.

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