

Deep frying induced oxidative changes in edible oils used in commercial Chips manufacturing industries

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Abstract: Edible oils are an essential part of the diet and serve as the primary sources of dietary fatty acids. However, during commercial use, edible oils are often subjected to deep frying, compared to limited heating in household settings. The present study aimed to evaluate the changes in selected commercially used edible oils exposed to deep frying. The oils were collected from an industry where banana chips are manufactured. Various parameters indicating lipid peroxidation, such as TBARS, CD, and CT, were analyzed according to standard protocols. The fatty acid profile was examined using GC-MS based on the fatty acid methyl ester method. Results showed a significantly elevated level of early lipid peroxidation products, such as conjugated dienes and trienes, as well as thiobarbituric acid reactive substances. FTIR spectroscopy confirmed the cis-trans conversion in the oils. These changes were more pronounced in oils rich in unsaturated fats, whereas they were less evident in coconut and palm oils. Considering these findings, it is possible that the consumption of deep-fried oils may be harmful to health; however, further studies are necessary to determine the impact of consuming these oils.

Keywords: Coconut oil, Deep frying, Fatty acid profile, Physico-chemical changes, Rice bran oil, Sunflower oil.

1. Introduction

Edible oils are the important dietary components, which is the predominant source of different fatty acids. They are mainly used for cooking purposes, which involves rapid and repeated heating at higher temperatures. During the process of frying, the edible oils are exposed to high temperature usually above 160°C. Previously, Choe and Min [1] and Warner [2] have well described the chemistry of edible oils during thermal treatments, these include hydrolysis, oxidation and polymerization reactions. Changes associated with thermal treatment also determine the biological effects of these edible oils after consumption.

Thermal oxidation, generates large volume of epoxides and peroxides of saturated or monounsaturated nature, resulting in the increase in peroxide values, color viscosity and polymers of triglycerides. The primary lipid peroxidation products formed during the process of deep frying include peroxides, conjugated diene structures. Due to their reduced stability and high reactivity, they undergo auto-decomposition yielding highly toxic secondary oxidation products such as aldehydes and ketone bodies. Ammouche, et al. [3] have reported that thermal oxidation of the sunflower oil increases its peroxide value and free fatty acids content. Polymerization reactions are crosslinking of two or more triglycerides, which often takes place during thermal oxidation of oils. Further, cyclization (formation of cyclic structures such as CFAMs) and dimerization (formation of intra strand crosslinking between two different fatty acids) reactions are also taking place during the deep frying process of oils. Studies have observed elevated levels of triacylglycerol polymers and dimers in oils [4].

India is a country with diverse food types depending on the regional specificity, likewise, the edible oils and fats chosen for the culinary purposes also varies. The common oils that are used for commercial purposes in India include coconut oil, palm oil, sesame oil, rice bran oil and sunflower oil. Among these coconut and palm oils are saturated fatty acid rich edible oils, whereas sunflower, sesame and rice bran oil contain polyunsaturated and monounsaturated fatty acid rich oil. In the current study, we analyzed the representative edible oils used for commercial purposes in India and their deep fried forms.

2. Materials and Methods

2.1. Edible Oils Used in The Study

Coconut oil, palm oil, sunflower oil, rice bran oil, and Sesame oil were purchased from the local market and kept under refrigerated conditions in order to avoid auto-oxidation. Deep fried forms of these edible oils, used for frying purposes were collected from commercial chips manufacturers, whose identity is not disclosed.

2.2. FTIR analysis

The samples were placed on the diamond/ZnSe crystal of ATR (Attenuated total reflection)-FTIR spectrophotometer (Spectrum Two, PerkinElmer, Massachusetts, United States) and scanned between the wavelengths 3500- 750 nm^{-1} . The percentage transmission against the wavelength was plotted. The transmission intensity of each spectrum was determined using the FTIR system software and used for the interpretation of the data.

2.3. Fatty acid profiling using GC-MS method

The total fatty acids in edible oils were converted to fatty acid methyl esters (FAME) as per standard protocols [5] and the FAME composition was analyzed using GCMS. GC-MS analysis was carried out using a GC model-7890A (Agilent Technologies, USA). The column used for the study was DB-5 with dimensions 30 m x 0.25 mm x 0.25 μm . The MS detector used was Agilent, 5975C Inert XL MSD. The injection volume was 1.0 μL and the injection temperature was 250°C. The Column Oven initial Temperature was 60° C with an increment at a rate of 5°C/ min. The detector was set at a temperature of 250°C.

2.4. Biochemical Analysis

Changes in the lipid peroxidation indicators such as thiobarbituric acid reactive substances (TBARS) [6], conjugated diene (CD) as well as conjugated triene (CT) [6] were estimated as per the standard protocol.

3. Results

3.1. FTIR Spectroscopic Analysis of Fresh and Deep Fried Oils

FTIR spectra of CO and FCO showed striking differences, especially in the region around 458, 583, 1111, 1743, and 3500 cm^{-1} . Respective transmittance of CO at these regions was 0.940, 0.959, 0.737, 0.513 and 0.995. In FCO, the transmittance at 458, 583, 1111, 1743, and 3500 cm^{-1} was 0.912, 0.933, 0.762, 0.596, and 0.977 (Table 1).

Similar to CO, FTIR spectra of PO and FPO showed variations near the wavelength 458, 583, 1111, 1742, and 3500 cm^{-1} . Respective transmittance of CO at these regions was 0.966, 0.941, 0.699, 0.553 and 0.952. In FCO, the transmittance at 458, 583, 1111, 1743, and 3500 cm^{-1} was 0.892, 0.901, 0.781, 0.586, and 0.933 (Table 1).

In deep fried sesame oil (FGO), the FTIR spectral differences were mainly around 3500, 3008, 1744, 1377, 1236, 967, 721, and 455 cm^{-1} . The transmittance of MO at these regions was 0.991, 0.944, 0.626, 0.912, 0.864, 0.727, 0.821, and 0.948, respectively. The transmittance at the same regions in FGO was 0.985, 0.966, 0.664, 0.899, 0.857, 0.737, 0.859, and 0.933, respectively (Table 1).

FTIR spectra of SO and FSO showed distinct changes in the regions at 3500, 3008, 1377, 1234, 1161, 1099, 969, 585, and 461 cm^{-1} . In RO, the respective transmittance at these regions was 0.994, 0.949, 0.905, 0.862, 0.731, 0.823, 0.924, 0.945, and 0.943. In FRO, the transmittance at the same regions was changed to 0.970, 0.967, 0.885, 0.839, 0.717, 0.824, 0.897, 0.928, and 0.914, respectively (Table 1).

Table 1.

FTIR analysis of fresh and commercially deep fried oil; absorbance is provided within parenthesis along with the respective wavelength.

Edible oil	Wavelength (Absorbance)
Coconut oil	3500 (0.995), 1743 (0.513), 1111 (0.737), 583 (0.959), and 458 (0.940)
Fried coconut oil	3500 (0.977), 1743 (0.596), 1111 (0.762), 583 (0.933), and 458 (0.912)
Palm oil	3500 (0.952), 3008 (0.814), 1743 (0.553), 1111 (0.699), 583 (0.941), and 458 (0.966)
Fried Palm oil	3500 (0.933), 3008 (0.893), 1743 (0.586), 1111 (0.781), 583 (0.901), and 458 (0.892)
Sesame oil	3500 (0.996), 3008 (0.903), 1744 (0.599), 1377 (0.927), 1236 (0.920), 967 (0.702), 721 (0.814), and 455 (0.965)
Fried sesame oil	3500 (0.969), 3008 (0.951), 1744 (0.689), 1377 (0.843), 1236 (0.885), 967 (0.744), 721 (0.860), and 455 (0.919)
Rice bran oil	3500 (0.991), 3009 (0.946), 1744 (0.621), 1237 (0.843), 987 (0.834), and 455 (0.924)
Fried rice bran oil	3500 (0.987), 3009 (0.960), 1744 (0.624), 1237 (0.835), 987 (0.856), and 455 (0.923)
Sunflower oil	3500 (0.944), 3008 (0.921), 1377 (0.922), 1237 (0.742), 1161 (0.673), 1099 (0.881), 969 (0.907), 585 (0.951), and 461 (0.964)
Fried sunflower oil	3500 (0.910), 3008 (0.972), 1377 (0.867), 1237 (0.704), 1161 (0.601), 1099 (0.844), 969 (0.872), 585 (0.903), and 461 (0.907)

In RO and FRO, the striking differences were observed in the regions 3500, 3009, 1744, 1237, 987, and 455 cm^{-1} . The transmission intensity at these respective regions of SO was 0.991, 0.946, 0.621, 0.843, 0.834, and 0.924. The transmittance at the same regions in FSO was 0.987, 0.960, 0.624, 0.835, 0.856, and 0.923 (Table 1).

3.2. Fatty Acid Composition and Changes During Thermal Oxidation

The result indicated that the oil extracted from coconut contain predominantly lauric acid (50.77%). The other major fatty acids in the CO were myristic acid and palmitic acid with 19.2 and 8.87%. However, oxidative modifications during thermal treatment reduced the levels of lauric acid (49.2%), capric acid (5.9%) and caprylic acid (3.6%); on contrary, the long chain saturated fatty acids such as myristic (21.7%) and palmitic acid (11.5%) were increased (Table 2). In Sesame oil also, similar changes were noticed as shown in Table 2.

Table 2.

Fatty acid profile of fresh and commercially deep fried edible oils.

Edible oil	Capric acid	Lauric acid	Myristic acid	Palmitic acid	Stearic acid	Oleic acid	Linoleic acid	α -linoleic acid	Other
CO	6.15	50.77	19.2	8.87	3.01	6.3	1.8	-	C8:0- 3.9%
FCO	5.9	49.2	21.7	11.5	4.5	3.1	0.5	-	C8:0- 3.6
PO	-	-	1.1	41.3	4.4	42.3	10.9	-	
FPO	-	-	1.3	46.5	7.1	37.6	7.5	-	
GO	-	-	-	8.9	6.1	41.3	42.5	0.7	0.5%
FGO	-	-	-	12.7	9.7	39.2	37.4	0.3	0.7%
SO	-	-	-	6.8	3.1	26.1	62.9	0.2	0.9%
FSO	-	-	-	9.7	6.2	24.7	55.4	0	1.1%
RO	-	-	0.5	19.1	3.1	46.5	30.2	0.6	
FRO	-	-	0.8	24.4	4.5	42.7	27.4	0.2	

Sunflower oil had higher level of linoleic acid (63.5%) followed by oleic acid (27.5%) and the total saturated fatty acid content was 8.0%. Upon thermal oxidation, the linoleic acid concentration was

reduced to 53.7% and oleic acid content was elevated to 29.8%. Total saturated fatty acid content was also increased to 13.2% of the total (Table 2).

In rice bran oil, predominant fatty acids were linoleic acid (46.5%), palmitic acid (25.5%), stearic acid (13.0), and oleic acid (8.5%). Upon thermal oxidation, the composition was changed to linoleic acid (40.1%), palmitic acid (27.8%), stearic acid (13.6), and oleic acid (9.2%) (Table 2).

3.3. Biochemical Changes During Thermal Oxidation

Lipid peroxidation levels as indicated by TBARS were found to be 0.241 ± 0.035 nmoles/kg in CO, which was increased to 0.861 ± 0.091 nmoles/kg in FCO. Similarly, TBARS in MO was 0.365 ± 0.094 nmoles/kg, which elevated to 1.210 ± 0.115 nmoles/kg in FGO (Table 3). Lipid peroxidation level in sunflower oil was 0.299 ± 0.08 nmoles/kg and it elevated during thermal oxidation (FSO) to 1.01 ± 0.09 nmoles/kg. In Rice bran oil, the level was 0.229 ± 0.05 nmoles/kg and in FRO it elevated to 1.12 ± 0.08 nmoles/kg (Table 3).

Table 3.

Changes in the biochemical parameters in polar and non-polar fractions of fried oils.

Edible oil	Conjugated dienes	Conjugated trienes	TBARS
CO	0.181 ± 0.005	0.095 ± 0.012	0.241 ± 0.035
TCO	$0.278 \pm 0.012^{**}$	$0.185 \pm 0.027^{***}$	$0.861 \pm 0.091^{***}$
PO	0.192 ± 0.011	0.091 ± 0.022	0.244 ± 0.073
FPO	$0.299 \pm 0.014^{**}$	$0.188 \pm 0.017^{***}$	$0.881 \pm 0.025^{***}$
GO	0.245 ± 0.009	0.129 ± 0.016	0.311 ± 0.025
FGO	$0.420 \pm 0.021^{***}$	$0.448 \pm 0.029^{***}$	$1.178 \pm 0.031^{***}$
RO	0.233 ± 0.018	0.137 ± 0.010	0.302 ± 0.022
FRO	$0.481 \pm 0.022^{***}$	$0.699 \pm 0.013^{***}$	$1.655 \pm 0.121^{***}$
SO	0.276 ± 0.021	0.146 ± 0.020	0.299 ± 0.055
FSO	$0.562 \pm 0.027^{***}$	$0.506 \pm 0.021^{***}$	$1.454 \pm 0.041^{***}$

The values are represented as mean \pm SD of three independent experiments, each carried in triplicate. ** indicate $p < 0.01$ and *** indicate $p < 0.001$

Conjugated diene level in fresh CO was found to be 0.181 ± 0.005 mmol/kg, which increased considerably to 0.278 ± 0.012 in FCO. In MO, the CD level was 0.109 ± 0.007 mmol/kg, which increased to 0.359 ± 0.020 mmol/kg in FGO (Table 3). Sunflower oil had a CD level of 0.183 ± 0.01 mmol/kg and which was increased to 0.422 ± 0.07 mmol/kg in FSO. In RO, the conjugated diene content was 0.202 ± 0.03 mmol/kg. It was increased to 0.344 ± 0.06 mmol/kg in FRO (Table 3).

CT is another lipid peroxidation marker, which was found to be 0.095 ± 0.012 mmol/kg in CO, elevated to 0.185 ± 0.027 during thermal oxidation. In MO, the conjugated triene level was 0.158 ± 0.410 mmol/kg and increased in FGO to a level of 0.537 ± 0.068 mmol/kg (Table 3). In fresh SO, the CT level was 0.131 ± 0.01 mmol/kg and which increased to 0.234 ± 0.09 mmol/kg in FSO. The CT level of RO was 0.122 ± 0.04 mmol/kg and it increased upon thermal oxidation to 0.155 ± 0.03 mmol/kg (Table 3).

4. Discussion

Oils are chemically triglycerides, which contain three fatty acids attached to a glycerol moiety. The physical and chemical nature as well as their biological effect is dependent on the attached fatty acids. During the extraction as well as purification, there occur several changes in them. In addition, during the cooking procedures they undergo rapid oxidative modifications. In the present study, it has been observed that the edible oils undergo severe oxidative damages, especially oxidative reactions. The increased level of lipid oxidation structures such as conjugated diene, conjugated triene, thiobarbituric acid reactive substances and p-anisidine values, indicate the oxidative insults in edible oils. The degree of oxidative modifications increases with the unsaturation of oil; hence, rendering the polyunsaturated fat rich oil most susceptible to the thermal oxidation.

Compared to the unsaturated fatty acid rich edible oils, saturated fat rich coconut oil/ palm oil are found to have lesser level of oxidation products. However, there observed an increase in lipid peroxidation products to a moderate level. Considerable increase in the lipid peroxidation products indicated in terms of CD, CT and TBARS are observed in unsaturated fat containing oils such as sunflower, sesame and rice bran. FTIR spectra of these fried oils also supported these results as the reduced transmittance in the 3500 cm^{-1} region, which indicate the aldehyde or hydroperoxide groups, indicates increased accumulation of aldehyde groups. This is in accordance with the previous studies conducted by Moros, et al. [7] and Zahir *et al.* (2017). The aldehyde or peroxide content may be generated as a result of thermal oxidation of unsaturated fatty acids such as oleic acid and linoleic acid present in these edible oils. Stretches at 1111 , 1154 , and 1254 cm^{-1} indicate saturated acyl groups [8], in our study they remained unaltered thus indicating the intact saturated fat content. Vilela, et al. [9] identified the region at 3008 cm^{-1} as an indicative of cis- bonds, the change in these region may also indicated of cis-trans conversion. This may partially contributed to the observed reduction in unsaturated fatty acid profile of deep fried oil. Together with this, oxidative breakage of unsaturated fatty acids may also have contributed to the same. Different unsaturated fatty acid rich edible oils have increased oxidative modifications, as indicated by the lipid peroxidation indices of conjugated diene, triene and TBARS. This observation is supported by the reduced transmittance at 3500 cm^{-1} in FGO, which indicates increased hydroperoxide or aldehyde generation. Further, Ali, et al. [10] described the region at 3008 cm^{-1} as the possible cis- to trans- conversion, hence increased transmittance in FGO at this region possibly indicates the cis- to trans- conversion occurred during thermal oxidation. Region 1744 cm^{-1} is indicative of carbonyl compounds, which is found to be reduced in MO than FGO. Since the increased in carbonyl content found in MO may be the presence of iso-thiocynates, which got decreased during thermal oxidation. Similar observations have been made in FSO and FRO at the regions 3500 , 3008 , and 1744 cm^{-1} indicated the increased hydroperoxides and cis- to trans- conversion occurred during thermal oxidation. Further, the regions at 1160 and 1237 cm^{-1} indicate saturated acyl groups [8]. All the unsaturated fatty acids rich edible oils upon thermal oxidation (FGO, FSO and FRO) have shown reduced transmittance at these regions, which may due to the conversion of unsaturated acyl group to saturated form. This assumption has been well supported by the fatty acid composition analysis, where a reduction in the unsaturated fatty acid content with concomitant increase in saturation has been observed in sesame oil, sunflower oil as well as rice bran oil.

Further, previous reports have indicated that deep fried vegetable oils contain higher levels of cyclic fatty acid monomers (CFAM), especially in unsaturated edible oils [11]. It is thus expected that lipid oxidation in the sesame oil, sunflower oil and rice bran oil may have resulted in the formation of CFAM. The CFAMs formed in the deep fried are known to be unhealthy, they inhibit the pancreatic lipase activity in animals [12]. On the contrary, studies have reported that a diet rich in medium chain saturated and monounsaturated fatty acids enhance the enzymatic activity of lipases [13, 14].

Accumulation of lipid oxidation products are often recognized as a risk factor for many of the degenerative diseases including non-alcoholic fatty liver [15, 16] and colorectal cancers [17]. Studies have come up with the observations that deep fried foods especially using deep fried edible oils increase the risk for various metabolic and lifestyle associated diseases [18]. In animal models consumption of deep fried edible oils have also shown to induce hypertension [19], dyslipidemia and oxidative stress [20, 21], impaired glycerolipid metabolism as well as gut microbiota [22].

Thus, it is expected that, due to the higher levels of polymerization or oxidation products in deep fried oils, they may induce deleterious changes. However, further independent investigations on each of the edible oil using animal models are necessary to understand the actual effects of these deep fried edible oils on the extent of toxic effects in animal body and metabolism.

Transparency:

The authors confirm that the manuscript is an honest, accurate, and transparent account of the study; that no vital features of the study have been omitted; and that any discrepancies from the study as planned have been explained. This study followed all ethical practices during writing.

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