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A kinetic study on the effect of short-term frying cycles on the properties of cold-pressed peanut oil

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The present study investigated the effect of frying cycles on the fatty acid profile and physicochemical properties of cold-pressed peanut oil during preparation of French fries. The quality indices monitored are acid value, peroxide value, iodine value, total polar materials, hunter 'L', 'b' value, total colour difference and fatty acid composition. Results showed that significant changes ($p<0.05$) were observed in quality indices. Fatty acid profiling showed gradual increase in saturated fatty acid content and decrease in unsaturated fatty acid content during repeated deep fat frying cycles. Kinetic modelling reveals that the change in acid value, and total polar materials followed first-order kinetic model whereas iodine value, hunter 'L', 'b' value and total colour difference followed zero-order kinetic model. The correlation of frying cycle with the oil quality indices showed that frying cycle has strong positive correlation with total colour difference (r: 0.9966) and strong negative correlation with hunter 'L' value (r: -0.9971).

Keywords: cold-pressed peanut oil, kinetic modelling, fatty acid profiling, GC-MS.

INTRODUCTION

Edible oil is an excellent reserve of energy and essential fatty acids which act as a carrier of fat-soluble vitamins in our body. Deep frying is one of the oldest and popular food preparations, which is a process of immersing food in oil at a high temperature of 150 to 200 °C [1]. The desirable flavour, colour and crispy texture makes deep-fat fried foods very popular [2, 3]. Physicochemical reactions such as thermo-oxidation, hydrolysis, polymerization, isomerisation or cyclization take place at the high temperature of the frying process, leading to the decomposition of frying oil and formation of oxidative compounds, which affects the quality of oil and fried product [4]. The decline in quality of frying oils can be monitored using quality parameters such as total polar materials (TPM), acid value (AV), peroxide value (PV), iodine value (IV), total colour difference (TCD), density and refractive index, etc. [5]. According to reports, cold-pressed oils are healthier than refined oils, since nutritive capacity of refined oil is lost after the refining process. Cold-pressed peanut oil (CPO) is pale yellow in colour and has the characteristic "nutty" flavour of roasted peanuts. It is highly nutritious due to high level of polyunsaturated fatty acids, tocopherols, phytosterols, carotenoids, chlorophylls and polyphenolics, as well as significant amount of resveratrol. The antioxidant and antimicrobial efficiency of resveratrol provides health benefits, such as the prevention of cardiovascular diseases, atherosclerosis and cancer [6]. The major fatty acids of peanut oil are palmitic, stearic, oleic, linoleic and arachidic acid. Linoleic and oleic acids constitute up to 75 % of peanut oil [7, 8]. Oleic acid contributes to higher stability of peanut oil over other oils rich in polyunsaturated fatty acids, *viz*. sunflower oil, safflower oil, etc. [9].

The rate of deterioration of frying oil and formation of breakdown products differs based on various factors [10]. The optimum frying life, performance and quality changes of edible oil during frying can be estimated using kinetic models and the knowledge of kinetics helps us to predict quality changes and to determine the threshold reject point [11]. The objectives of the present work were to evaluate the effect of frying cycles on the physico-chemical properties of CPO during frying experiments. The changes in physical and chemical parameters were modelled using kinetic equations and the relationship between the parameters and the frying cycle was also investigated.

EXPERIMENTAL

Frying experiment

CPO and fresh potatoes were purchased from the local market in Chennai, Tamil Nadu. Potatoes were washed, peeled and cut into strips (1 cm × 1 cm × 6 cm). The samples of the potato slices (200 g) were randomly selected for frying. The frying operation was carried out in an Orbit DF30 3.5 L stainless steel electric deep fryer. The frying pot was filled with 3 kg of fresh oil and the frying oil-to-product ratio was maintained at 20:1 (w/w). Frying was carried out at 180 °C for 7 min. One frying cycle includes deep

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of French fries for seven min and draining of oil in the frying basket for one min. Seven consecutive frying cycles were carried out and oil was collected after each frying cycle, stored in tight screw cap bottles at -20 °C till further analysis.

Physicochemical analysis of the oil samples

Fatty acid profiling

Fatty acid methyl esters (FAMEs) of the oil samples were prepared using a sodium methoxide-methanol solution [12]. FAMEs were analysed by GC-MS (Thermo Scientific, Trace 1300- TSQ 8000 Evo). Separation of fatty acid methyl esters was carried out on a TG 5-MS column (30 m × 0.25 mm I.D; 0.25 µm film thickness). The GC-MS interface temperature was maintained at 240°C. Helium was used as a carrier gas at a constant flow rate of 1.0 ml/min. 1 µl of sample was injected by split-less mode with the injector port at 240 °C. The column temperature programme was as follows: 55 °C, held for 2 min; 7.5 °C/min to 180°C, held for 15 min; 8 °C/min to 230 °C, held for 15 min. For MS detection, electron ionization with 70 eV was used, with scan range of 50 -500 amu, scan rate 0.2012 s^{-1}. The ion source temperature and transfer line temperature were at 240°C. Solvent cut of time/delay time of 2.3 min was used for detection. The peaks identification was according to retention times of the reference FAME standard. Peak areas were determined by ICIS peak detection algorithm.

Physicochemical properties

AV of the oil samples were determined by titration method [13] expressed as mg KOH/g of oil. For the determination of PV, 5 g of oil sample was mixed with 30 ml of acetic acid-chloroform (3:2 (v/v)). Then 0.5 ml of freshly prepared saturated potassium iodide solution was added followed by addition of 30 ml of distilled water. The liberated iodine was titrated with 0.01 N sodium thiosulfate solution using starch (1 %) as indicator. The values were expressed as meqO_2/kg of oil [14]. IV was determined by adding 10 ml of carbon tetrachloride and 25 ml of Wijs solution to one gram of oil. The solution was kept in dark for 30 min followed by addition of 15 ml of potassium iodide (10 %) and 100 ml of distilled water. The solution was then titrated with 0.1 N sodium thiosulfate solution using starch as an indicator. A blank titration was carried out without the oil samples [15]. TPM (%) of the frying oil samples were measured directly with a cooking oil monitor (Testo 270 cooking oil tester).

Colour measurements of the oil samples were carried out using a Hunter Lab Ultra-Scan Vis spectrophotometer. The colour values were expressed as L (whiteness or brightness/darkness), a (redness/greenness) and b (yellowness/blueness). An optically clear glass cell with fixed path length of 20 mm was used for measurements [16].

Kinetic modelling

During frying various complex reactions take place and change in quality parameters are analysed using kinetic equations [11]. Chemical reaction kinetics is applied to quantify individual attribute of an ideal food system in form of the general rate law [17]:

$$dP/dt = \pm k\, P_n$$

where k is the rate constant (1/min), t the reaction time (min), n the reaction order and P represents a quantitative value for a quality attribute.

The food quality changes are generally modelled as zero- and first-order rate reactions. The rate equations (Equations 1 and 2) are as follows [17, 18]:

$$\text{Zero-order equation: } P = P_0 \pm kt \quad (1)$$

$$\text{First-order equation: } P = P_0\, e^{\pm k.t} \quad (2)$$

where P_0 represents the value of a quality attribute at t=0. The coefficient of determination (r^2) was used as primary criterion to select the best fit of the tested mathematical model to the experimental data. The higher the value of r^2 the better the model was taken to fit [19].

Statistical analysis

The physicochemical properties of oil samples were analysed in triplicate and the fatty acid profile was analysed in duplicate. The results of the experiments were expressed as the mean ± standard deviation. The data were subjected to analysis of variance (ANOVA) and the Duncan's test for the 5 % significance level using the SPSS software (Version 21, IBM, New York, USA).

RESULTS AND DISCUSSION

Impact of frying cycles on fatty acid profile

The fatty acid composition, monounsaturated /saturated fatty acid ratio (m/s) and polyunsaturated/ saturated fatty acid ratio (p/s) of fresh oil and oil samples of initial and final frying cycle are represented in Table 1.

Table 1. Fatty acid composition (%) of fresh cold-pressed oil and at first and seventh frying cycle

Fatty acids	Fresh oil	First frying cycle	Seventh frying cycle
C14:0	0.504[a]	0.592[b]	1.160[c]
C16:0	20.440[a]	22.092[b]	32.942[c]
C18:0	0.174[a]	0.897[b]	1.198[c]
C20:0	3.313[a]	3.894[b]	4.606[c]
C22:0	0.217[a]	0.407[b]	0.546[c]
C24:0	0.142[a]	0.183[b]	0.428[c]
C14:1	0.156[c]	0.137[b]	0.107[a]
C16:1	1.418[c]	1.197[b]	0.883[a]
C18:1	46.586[c]	44.911[b]	40.794[a]
C18:2	25.234[c]	24.375[b]	16.760[a]
C18:3	1.799[c]	1.266[b]	0.532[a]
m/s	1.943	1.648	1.022
p/s	1.091	0.914	0.423

The values reported are mean values of duplicate experiments. Values with different superscripts within a row are significantly different (P<0.05). [C14:0-myristic acid; C16:0-palmitic acid; C18:0-stearic acid; C20:0-arachidic acid; C22:0-behenic acid; C24:0-lignoceric acid; C14:1-myristoleic acid; C16:1-palmitoleic acid; C18:1-oleic acid; C18:2-linoleic acid; C18:3-linolenic acid; m/s: monounsaturated/saturated fatty acid ratio; p/s: polyunsaturated/saturated fatty acid ratio].

A significant increase ($p<0.05$) in the saturated fatty acid content and decrease in the unsaturated fatty acids during the frying cycles was observed. The decrease in unsaturated fatty acids during repeated frying is due to oxidation and thermal degradation [20].

The decrease was particularly noticeable in polyunstaurated fatty acids contributing to their multiple unsaturated bonds' instability [21]. The fatty acids with double bonds are prone to oxidation due to the presence of π (pi) bonds [22]. The decrease in oleic and linoleic acid content of peanut oil during deep frying has been previously reported [9]. The m/s and p/s ratio of fresh oil was 1.943 and 1.091. The ratio decreased to 1.022 and 0.423 at the end of the seventh frying cycle.

Impact of frying cycles on AV

AV is used as an indicator of hydrolytic rancidity of vegetable oils [9], which is associated with the amount of free fatty acids present in the oil. During frying water, steam and oxygen initiate a series of chemical reactions in oil and food which form carbonyl compounds which are then oxidized to low molecular weight free fatty acids [23, 24]. As shown in Table 2, the AV of fresh CPO was found to be 1.309±0.030 mg KOH/g of oil, which is within the limit suggested by the Codex Alimentarius Commission [25].

Table 2. Quality indices of fresh CPO

S. No	Quality index	Values
1	AV	1.309±0.030
2	PV	4.290±0.587
3	IV	95.36±0.48
4	TPM	3.0±0.00
5	Hunter 'L' Hunter 'b'	83.685± 0.056 30.937±0.040

The values are expressed as mean ± standard deviation. AV – acid value (mg KOH/g of oil); PV – peroxide value ($meqO_2$/kg of oil); IV – iodine value (gI_2/100 g); TPM – total polar materials (%).

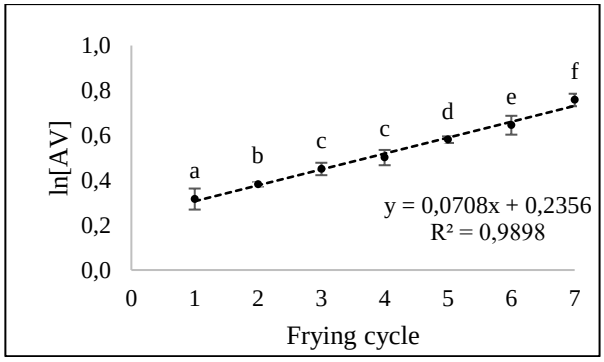

Figure 1. Change in AV with respect to frying cycles on a natural log scale. The dotted line represents first-order fit. The standard deviation is denoted as error bars. Values with different data labels are significantly different (P<0.05).

Fig. 1 shows the results of the kinetic study of the increase in AV of oil sample with each frying cycle. The AV of CPO increased significantly ($p<0.05$) with frying cycles. The AV of frying oil was reported to increase with the number of frying cycles and it is recognized that hydrolysis of oil/fat results in formation of free fatty acids, di-acyl glyceride, mono-acyl glyceride and glycerol [9, 26]. The first order kinetic model ($r^2 = 0.9898$) was found to be adequate in describing the increase in AV with respect to the frying cycles. AV showed positive correlation with frying cycles, PV, TPM, hunter 'b' value, TCD and it showed negative correlation with IV and Hunter 'L' value (Table 3).

Impact of frying cycles on PV

The PV is the measure of primary oxidation products i.e., hydro-peroxides formed by oxidation of oil during frying [9]. PV of oil also varies with respect to the storage time, temperature and contact with air of the oil samples. As shown in Table 2, the PV of fresh CPO (4.290±0.587 $meqO_2$/kg of oil) was also within the limit suggested by the Codex Alimentarius Commission [25]. A significant change of PV is noted but there are inconsistencies in the change during each frying cycle (Fig. 2). The inconsistencies are due to the unstable nature of

peroxides at high temperatures [27]. The peroxides break down to secondary oxidation products like carbonyl and aldehyde compounds causing the decrease in PV [28]. The similar phenomenon has been widely reported in the literature [9, 29]. PV showed positive correlation with frying cycles, AV, TPM, Hunter 'b' value, TCD and it showed negative correlation with IV and Hunter 'L' value (Table 3).

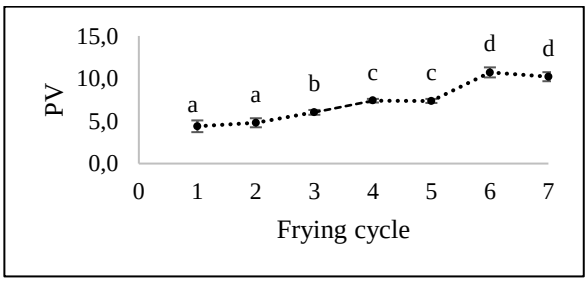

Figure 2. Change in PV with respect to frying cycles. The standard deviation is denoted as error bars. Values with different data labels are significantly different (P<0.05).

Impact of frying cycles on IV

IV, a measure of degree of unsaturation of the edible oils is used to determine the oxidative stability of oils[30]. The IV of fresh oil was found to be 95.36±0.48 g I_2/100 g (Table 2) and decreased significantly (p<0.05) with respect to frying cycles (Fig. 3). The decrease in IV is due to the oxidative and thermal degradation reactions which occurs during the deep frying[31]. The zero order kinetic model was found to be satisfactory in describing the change in IV (Fig. 3). IV showed positive correlation with hunter 'L' and negative correlation with frying cycles, AV, PV, TPM, hunter 'b' value, TCD (Table 3).

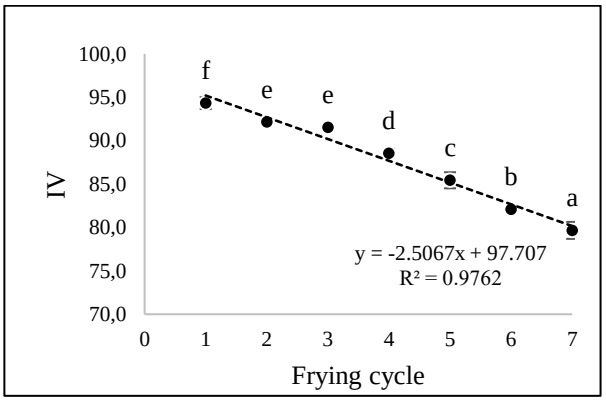

Figure 3. Change in IV with respect to frying cycles. The dotted line represents zero order fit. The standard deviation was denoted as error bars. Values with different data labels are significantly different (P<0.05).

Impact of frying cycles on TPM

TPM has been considered as one of the best indicators of the overall quality of frying oils since it represents the accurate estimation of the thermo-oxidative deterioration of frying oils by assessing the entire degraded components existing in the oil [26]. The factors which influence the formation of polar materials are fatty acid composition of oil, frying temperature, ratio of the surface oil area to oil volume in the fryer, food composition and turnover ratio [32]. The TPM of fresh CPO is 3.0 % (Table 2) and it increased significantly (p<0.05) with each frying cycle (Fig. 4). The increase in TPM is strongly related to the number of frying cycles and frying time [33]. The change in TPM with respect to frying cycles was best fitted using first-order kinetic model (Fig. 4). TPM showed positive correlation with frying cycles, AV, PV, Hunter 'b' value, TCD and it showed negative correlation with IV and Hunter 'L' value (Table 3).

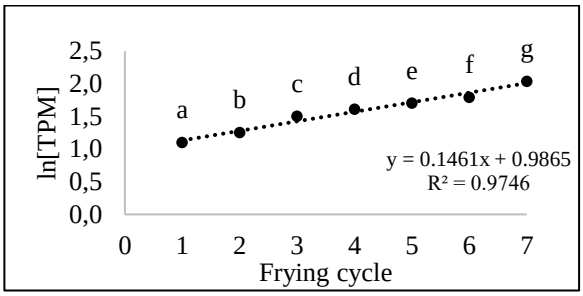

Figure 4. Change in TPM with respect to frying cycles on a natural log scale. The dotted line represents first order fit. The standard deviation is denoted as error bars. Values with different data labels are significantly different (P<0.05).

Impact of frying cycles on colour

Colour is the prominent physical property of the edible oils. The colour of oils darkens during frying which mainly depends on the temperature of deep frying, time of frying and the type of food fried. Typically the colour of edible oil changes from pale yellow to light brown and then dark brown during repeated frying [16, 24].When food is fried at high temperatures, chemical reactions such as thermal oxidation and polymerization of the unsaturated fatty acids lead to the formation of non-volatile decomposition products like polymers and non-polar compounds which contribute to the colour change [34]. The Hunter 'L' and 'b' values represent light/dark and yellow/blue colour of the sample. In the current study, the 'L' value reduced significantly (p<0.05) with frying cycles (Fig. 5). The change indicates darkening of the frying oil caused by oxidised products, products of Maillard reaction and chemical reactions of the oxidised products with

Maillard reaction products, charred food residues, leaching of pigments from food particles into the oil [16, 35, 36]. It has been reported that during frying of potatoes, reactions between sugar aldehyde groups and amino acids produce brown products [37]. The change of 'L' value was best fitted in zero-order kinetic model (Fig. 5). The Hunter 'b' value increased significantly ($p<0.05$) with frying cycles (Fig. 5). The increase in yellow colour might be due to the pigments present in the food particles, the colour induced from Maillard reactions, formation of the chroman-5,6-quinones by partial oxidation of vegetable oils [16, 36, 38]. The zero-order kinetic model was found to be adequate in fitting the Hunter 'b' value (Fig. 5).

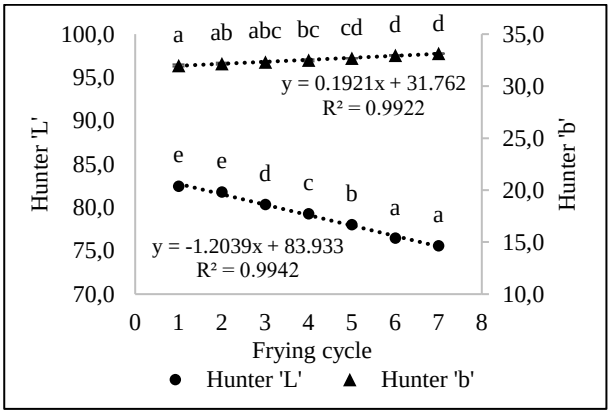

Figure 5. Change in hunter 'L' and 'b' with respect to frying cycles. The dotted line represents zero order fit. The standard deviation is denoted as error bars. Values with different data labels are significantly different ($P<0.05$).

The TCD represents, overall, that the oil darkness increased significantly ($p<0.05$) with each frying cycle and it was best fitted in zero-order reaction kinetic model (Fig. 6). The TCD following zero-order kinetic model has also been reported [10, 16, 39].

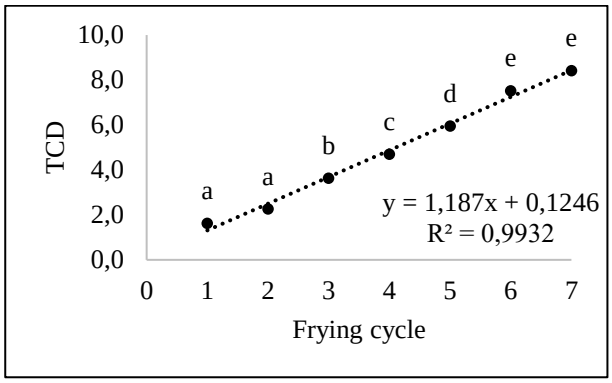

Figure 6. Change in yellowness with respect to frying cycles. The dotted line represents zero order fit. The standard deviation is denoted as error bars. Values with different data labels are significantly different ($P<0.05$).

CONCLUSION

In conclusion the current study reports the impact of frying cycles on the quality deterioration of CPO during frying of French fries. Our findings suggest that the frying cycles declined the quality of oil proven by increase in AV, PV, TPM and darkening of oil. Frying cycle showed strong correlation with the colour of the oil. Kinetic modelling showed change in AV, and TPM followed first-order kinetic model while IV, Hunter 'L', 'b' and TCD followed zero-order kinetic model. Fatty acid profile showed significant decline in unsaturated fatty acids. The marginal changes in the quality changes in CPO subjected to frying, indicated its stability as a frying medium.

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Table 3. Pearson correlation of quality indices of CPO

	Frying cycles	AV	PV	IV	TPM	Hunter 'L'	Hunter 'b'	TCD
Frying cycles		0.9867	0.9583	-0.9880	0.9849	-0.9971	0.9961	0.9966
AV	0.9867		0.9388	-0.9881	0.9923	-0.9849	0.9911	0.9862
PV	0.9583	0.9388		-0.9603	0.9260	-0.9711	0.9696	0.9717
IV	-0.9880	-0.9881	-0.9603		-0.9689	0.9896	-0.9948	-0.9912
TPM	0.9849	0.9923	0.9260	-0.9689		-0.9794	0.9810	0.9793
Hunter 'L'	-0.9971	-0.9849	-0.9711	0.9896	-0.9794		-0.9971	-0.9999
Hunter 'b'	0.9961	0.9911	0.9696	-0.9948	0.9810	-0.9971		0.9977
TCD	0.9966	0.9862	0.9717	-0.9912	0.9793	-0.9999	0.9977	

AV – acid value (mg KOH/g of oil); PV – peroxide value (meqO_2/kg of oil); IV – iodine value (gI_2/100g); TPM – total polar materials; TCD – total colour difference.

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