RESEARCH ARTICLE

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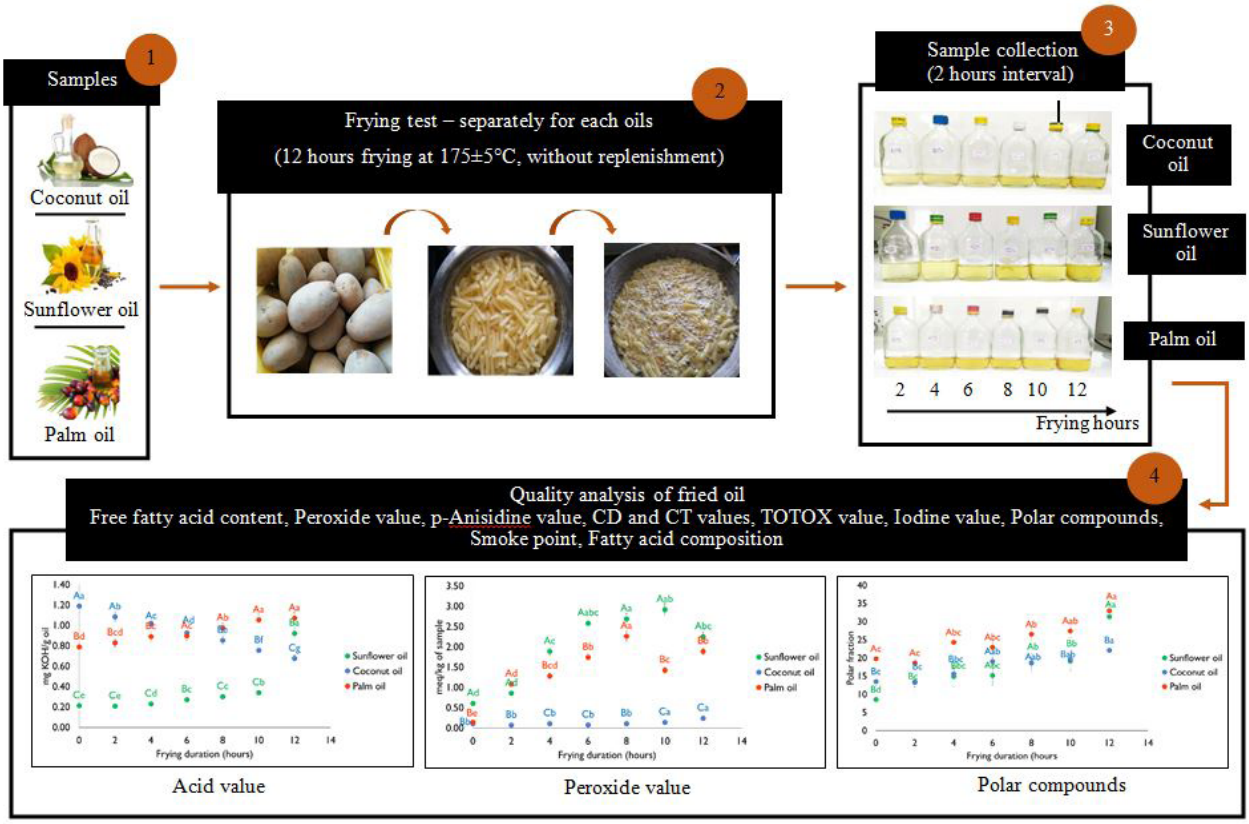

Research Highlights

- The stability of coconut oil, sunflower oil, and palm oil during frying was explored.
- Frying experiments were conducted by deep-frying potatoes for 12 hours
- Coconut oil had the highest stability and sunflower oil had the lowest stability
- *Trans* fat formation was observed in palm oil and sunflower oil

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Comparative study on stability of coconut oil, sunflower oil and palm oil during continuous deep frying

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Abstract: This study was carried out to evaluate the stability of three oils, namely, coconut oil, sunflower oil and palm oil during continuous deep frying. An experiment was conducted by frying potato slices at 175±5 °C for 15 minutes (one cycle) using different oils. The same oils were reused for frying over a period of 12 hours and samples were collected after every two hours of frying. The oil samples were tested for chemical and physical changes and oxidative stability during frying. Rates of increase in acid value, *p*-anisidine value and peroxide value were higher in sunflower oil (0.06 mg NaOH /g oil/hour, 8.03/hour and 0.13 meq/kg/hour, respectively) than in other oils. Smoke points of all oils decreased significantly over time, with the highest reduction in palm oil (208.5 to 172 ºC). Total polar compounds increased significantly during frying in all samples. Major reductions were observed in the linoleic acid content in palm oil and sunflower oil. *Trans* fat content did not increase significantly in coconut oil, whereas, a slight increase was observed in palm oil (1.14 to 1.21%) and sunflower oil (0.61 to 0.72%). In conclusion, coconut oil showed the highest stability whereas sunflower oil showed the lowest stability during continuous deep frying.

Keywords: Continuous deep frying, Frying cycle, Oxidative stability, Total polar compounds, *Trans* fat.

INTRODUCTION

Edible oils play a crucial role in the human diet as a source of energy and essential fatty acids. Frying is one of the most frequently used culinary practices around the world. Presently, fried food consumption is increased due to the rapid expansion of fast-food centers, changing lifestyles, and the higher number of working women. Even though deep frying can produce foods with good sensory qualities such as pleasant aroma, flavour and crispiness, it causes various chemical and physical changes in the fried food and frying oil. Major chemical reactions occurring in the frying oil during deep frying include hydrolysis, oxidation and polymerization, which are detrimental to the quality of the fried food as well as frying oil (Aniołowska *et al*., 2016; Asadi and Farahmandfar, 2020). These reactions are responsible for the production of harmful substances which affect the quality of oil and causes several adverse health effects on human (Syed, 2016).

Coconut oil, palm oil and sunflower oil are widely used for frying in Sri Lanka. Repeated use of frying oil is the major factor that affects the fried food quality (Jiang *et al*., 2019). Some European nations have published specific regulations regarding frying oils, however, there are no such regulations related to the quality of fried foods or for frying oils in Sri Lanka. Repeated use of oil for deep frying is a common practice in Sri Lanka in restaurants, street vending as well as in the home kitchen. Repeated use of frying oils negatively affects health and increases the risk for cardiovascular diseases due to the harmful compounds produced during frying (Ambreen *et al*., 2020; Grootveld *et al*., 2020). *Trans* fats are unhealthy fats that are produced during frying (Mekonnen *et al*., 2020). Repeated use of oil can accelerate the formation of *trans* fats, which are absorbed into the food. The increased risk of cardiovascular diseases and other human ailments is associated with dietary *trans* fats (Boateng *et al*., 2016). In this background, this study was designed to compare the stability of coconut oil, sunflower oil and palm oil during repeated use for continuous deep frying. This study will provide information on how long the selected oils can be used for frying continuously without negatively affecting the quality of the oil, and thus the quality of the fried food.

MATERIALS AND METHODS

Materials and Reagents

Oils and potatoes were purchased from local shops in Jaffna. All analytical grade chemicals and standards for Gas Chromatography (Supelco 37 component FAME mix and mixture of *trans* isomers of linoleic acid) were purchased from Merck, Germany.

Frying Experiment

Preparation of potato: Fresh potatoes were cleaned, peeled and sliced (0.5×0.5×3 cm^3) using a mechanical slicer. They were submerged in water at room temperature for 5 minutes and drained for 5 minutes.

Deep frying: All three oil samples were used separately to carry out the frying experiment. A steel frying pan with a maximum capacity of 2 L was used. The ratio of potato: oil (kg: L) was maintained at 1:3 throughout the frying. The volume of oil was measured before each frying cycle

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begins and the amount of potato slices to be fried in the next frying cycle was decided depending upon the volume of oil remaining. Oil (1.5 L) was poured into the frying pan and heated to 175±5 °C. Raw potato slices were introduced to the oil and frying was continued for 15 minutes. After the chips were taken out, the oil was heated again for 2 minutes before the next frying cycle. Constant oil temperature (175±5 °C) was maintained throughout frying by monitoring the temperature using a thermocouple. This process was conducted for three consecutive days (4 hours of continuous frying per day) which gave a total of 12 hours of frying. The same oil was reused without replenishment. Used oil samples were stored at room temperature to be used for frying the next day. About 40 mL of oil sample was collected after every two hours of frying. Collected oil samples were cooled to room temperature (28 °C) and stored at -20 °C in sealed glass bottles after flushing with nitrogen till analysis within a week. Frying experiment was conducted two times for each type of oil and the samples were analyzed in triplicates.

Analysis of Frying Oil

The initial quality of oils and quality of frying oil after every 2 hours of frying were assessed by determining the following parameters.

Determination of acid value: AOAC official method Ca 5a-40 (AOCS, 2009) was employed to determine acid value. Briefly, the oil sample (3-5 g) was dissolved in 25 mL of ethanol. The dissolved sample was boiled in a water bath for 30 minutes and titrated against 0.1 N NaOH solution and the acid value was calculated as a percentage.

Determination of Peroxide value: Peroxide value of the sample was determined using the spectrophotometric method (Hornero-Méndez *et al*., 2001). Briefly, the sample (0.05 g) was dissolved in 2 mL of chloroform/ acetic acid solution (2:3 v/v). Then, 200 µL of Fe (II) solution, 2 mL of deionized water and 4 mL of diethyl ether were added. The aqueous phase (1 mL) was transferred to another tube and mixed with 50 µL of saturated ammonium thiocyanate solution and 2 mL of deionized water. After 10 min, absorbance at 470 nm and 670 nm was measured against blank (deionized water) using UV visible spectrophotometer (Thermo Fisher Scientific, Evolution 200). A calibration curve was prepared using Fe (III). Results were calculated as meq/kg of the sample using the following equation.

$$\text{Peroxide value} = \frac{As470nm - Ab470nm}{55.84 \times 2 \times m \times W} - \frac{As670nm - Ab670nm}{55.84 \times 2 \times m \times W}$$

Where As470 nm is the absorbance of the sample at 470 nm; Ab470 nm is the absorbance of the control at 470 nm; As670 nm is the absorbance of the sample at 670 nm; Ab670 nm is the absorbance of the control at 670 nm; m is the slope of the Fe (III) calibration curve; 55.84 is the atomic weight of Fe; 2 is the factor to convert meq of Fe to meq of peroxide and W is the sample Weight (g).

Determination of p-anisidine value: p-Anisidine value of the samples was determined according to AOCS Official method Cd 18-90 (AOCS, 2009). Sample (1±0.05 g) was measured into a 25 mL volumetric flask and volumed up with 2,2,4-trimethylpentane. Sample solution (5 mL) and solvent (5 mL) were added separately into the test tubes. *p*-Anisidine reagent (1 mL) was added to each tube. After 10 minutes, absorbance of the sample was measured at 350 nm, using a solvent tube as blank. *p*-Anisidine value was calculated using the following equation.

$$\text{p-Anisidine value} = \frac{25 \times (1.2\ As - Ab)}{m}$$

As - absorbance of the solution after reaction with the *p*-anisidine reagent; Ab - absorbance of the solution; m - sample weight (g).

Determination of Total Oxidation (TOTOX) values: The following equation was used to determine TOTOX values for oil samples.

$$\text{TOTOX value} = (2 \times \text{peroxide value}) + \text{p-Anisidine value}$$

Determination of conjugated diene (CD) and conjugated triene (CT) values: CD and CT values were determined by following the standard method (IUPAC, 1987) with slight modifications. The oil sample (1±0.05 g) was dissolved in 25 mL of 2,2,4-trimethylpentane. Then the absorbance was measured at 235 nm for CD and 270 nm for CT using UV-Visible spectrophotometer (Thermo Fisher Scientific, Evolution 200). The results were calculated as E_1% using following equation;

$$E1\% = \frac{A\lambda}{C \times 1}$$

A_λ - absorbance measured at either 235 nm or 270 nm; C - concentration of oil solution (g/100 mL); l - path length of the cuvette (cm)

Determination of iodine value: The Iodine value was determined using AOAC Official method 993.2 (AOAC, 1998). The oil sample (0.1-3 g) in a 500 mL conical flask was dissolved completely in 10 mL chloroform and Wijs solution (25 mL) was added and stored in dark for 30 minutes. Subsequently, 20 mL potassium iodide (15%) solution and distilled water (100 mL) were added and titrated with 0.1M standard sodium thiosulfate solution while shaking vigorously until the yellow colour almost disappeared. The starch solution was added and titrated until the blue colour completely disappeared. The iodine value was calculated as g of iodine per 100 g of oil.

Determination of polar compounds: The column chromatography method (IUPAC, 1992) was employed to separate the polar and nonpolar fractions of the frying oils. For the separation of polar and nonpolar compounds, a mixed solution of light petroleum ether and diethyl ether, 90/10 (v/v) (solvent 1) and diethyl ether (solvent 2) were used as solvents. The oil sample (1±0.1 g) was dissolved in solvent 1 in 10 mL volumetric flask. Sample was introduced into the column and the nonpolar fraction was eluted using the elution solvent 1 (60 mL) and the polar fraction was eluted using solvent 2 (250 mL). The flow rate was maintained at 1.5 mL/min. The mass fraction of polar compounds was calculated as percentage.

Determination of smoke point: The AOCS Official method Cc 9a-48 (AOCS, 1997) was used to determine the smoke point of oils. A portion of each oil was taken in the boiling tube and heated until it started producing smoke at which point the temperature was noted using a thermocouple.

Determination of fatty acid profile: Gas chromatography was used to analyze the fatty acid profile of the samples. Fatty acid methyl esters (FAMEs) were prepared according to AOAC Official Method 996.06 (AOAC, 2001). FAMEs were analyzed by injecting 1 µL into GC, equipped with a Flame Ionization Detector (FID) and a fused silica capillary column (100 m length, 0.250 mm diameter, 0.20 µm film) (Model - Agilent technologies 7890B GC system) using nitrogen as carrier (20 mL/min). The split ratio was 30:1. Injector and detector temperatures were maintained at 260 °C. The initial column oven temperature was 140 °C for 5 minutes and increased to 240 °C at the rate of 4 °C/ min, then maintained at that temperature for 10 minutes. Authentic standards such as Supelco 37 component FAME mix and mixture of *trans* isomers of linoleic acid were used to identify the fatty acids.

Statistical Analysis

The frying experiment was carried out in duplicates and the samples were analyzed in triplicates. The data were expressed as the mean ± standard deviations of the mean using Microsoft excel 2013. Analysis of variance (ANOVA) was conducted using Statistical Analysis System (SAS), version 6.0.10. Two factor Completely Randomized Design model was used for the analysis. Duncan's Multiple Range Test was used to compare the treatment means at $p<0.05$. Spearman's correlation coefficients between chemical parameters were calculated using Microsoft Excel 2013.

RESULTS AND DISCUSSION

The changes in chemical and physical parameters of sunflower oil, coconut oil and palm oil during continuous deep frying for up to 12 hours are shown in Table 1. The presence of moisture from the food and high heat during frying accelerate the hydrolysis of oils (Park *et al*., 2020). In the presence of steam, free fatty acids and acidic compounds of low molecular weight formed during lipid oxidation increase the hydrolysis process (Nayak *et al*., 2016) thus increasing the free fatty acid content (Ben Hammouda *et al*., 2018; Ben Hammouda *et al*., 2017). Initial acid values of all three oils were significantly different ($p<0.05$). Sunflower oil and palm oil exhibited a significant increase ($p<0.05$) in acid value during frying whereas coconut oil showed a significant decrease ($p<0.05$) in acid value. Even though the coconut oil had a significantly ($p<0.05$) higher initial acid value than sunflower and palm oil, the reduction in the acid value of coconut oil indicates the loss of free fatty acids or less hydrolysis with increasing frying hours. Coconut oil contains a relatively high proportion of shorter chain fatty acids than the other two oils. Therefore, easily volatile short chain fatty acids may have been lost by volatilization due to high temperature during frying (Liu *et al.,* 2020) also be neutralized by the composition of the food being fried (Man *et al*., 1999). The rate of increase in the acid value of sunflower oil and palm oil was 0.06 and 0.02 and decreasing rate of coconut oil was 0.04 mg NaOH /g oil /hour. Based on the rate of increase in acid value, hydrolysis rate was high in sunflower oil than palm oil.

Peroxide value is used as a measure of primary oxidation whereas, *p*-anisidine value and CD and CT values are used as the measures of secondary oxidation products. TOTOX value is used as a measure of total oxidation that includes primary and secondary oxidation products. The peroxide value, *p*-anisidine value and TOTOX values increased significantly ($p<0.05$) with the frying hours in all three oils indicating an increase in primary and secondary oxidation products with frying hours. As shown in Table 1, the initial peroxide value, *p*-anisidine value and TOTOX value of sunflower oil, coconut oil and palm oil were different. It indicates that the initial oxidation level and quality of all oils were different. The rate of increase in the peroxide value of sunflower oil, coconut oil and palm oil were 0.13, 0.01 and 0.14 meq/kg/hour of frying, respectively. Sunflower oil showed a significant reduction ($p<0.05$) in peroxide value from ten hours of frying, whereas palm oil showed a significant ($p<0.05$) reduction from eight hours of frying, however, then it increased. Peroxides, the primary oxidation products, are unstable compounds and decompose into secondary oxidation products such as hydrocarbons, epoxy compounds, aldehydes, ketones, and organic acids (Dermiş *et al*., 2012). Thus, it could be the reason for the reduction in the peroxide value after a particular time. It is reported that the peroxide value of palm oil increased from 7.08 to 15.48 meq/kg during frying chicken nuggets at 170-180 °C for 3 minutes every 10 cycles per day, repeated for 10 days (Park and Kim, 2016). Another study reported that as the frying cycle increased, only *p*-anisidine increased proportionally, while the peroxide value rose and fell during frying, the same pattern observed in most deep frying studies (Trivedi *et al*., 2017) as observed in the present study. Even though the peroxide value increased with frying as in the present study, the differences in the results could be due to the differences in the initial quality of the oil and frying conditions.

The rate of increase in the *p*-anisidine value of sunflower oil, coconut oil and palm oil were 8.03, 0.19 and 8.05/hour of frying, respectively. Rates of increase in peroxide value, *p*-anisidine value and TOTOX value were low in coconut oil than in sunflower oil and palm oil. A clear indication that the oxidation of coconut oil during frying was lower than that of sunflower oil and palm oil. When the degree of unsaturation of fatty acids increases, the rate of oxidation also increases (Paul *et al*., 1997). Coconut oil has the highest degree of saturation followed by palm oil and sunflower oil. These results confirm that palm oil and sunflower oil are more susceptible to oxidation during frying at high temperatures than coconut oil. Similarly, Ben Hammouda *et al.* (2018), reported that *p*-anisidine value of olive pomace oil increased by frying potato at 180 °C. Oleic acid and linoleic acids are the major unsaturated fatty acids present in palm oil and sunflower oil that are easily oxidized. Linoleic acid is less stable to oxidation than oleic acid (Paul *et al.,* 1997). Accordingly, in this study also sunflower oil showed a higher level of oxidation than palm

Table 1: Changes in chemical and physical parameters of sunflower oil, coconut oil and palm oil during continuous deep frying.

Parameters	Oil	Frying duration (hours)						
		0	2	4	6	8	10	12
Acid value (mg NaOH /g oil)	SO	0.21 ± 0.01^{Ce}	0.21 ± 0.03^{Ce}	0.23 ± 0.02^{Cd}	0.27 ± 0.01^{Bc}	0.30 ± 0.04^{Cc}	0.34 ± 0.02^{Cb}	0.92 ± 0.02^{Ba}
	CO	1.19 ± 0.02^{Aa}	1.09 ± 0.02^{Ab}	1.02 ± 0.01^{Ac}	0.93 ± 0.00^{Ad}	0.85 ± 0.02^{Be}	0.76 ± 0.01^{Bf}	0.68 ± 0.01^{Cg}
	PO	0.79 ± 0.01^{Bd}	0.83 ± 0.03^{Bcd}	0.89 ± 0.01^{Bc}	0.90 ± 0.03^{Ac}	0.98 ± 0.03^{Ab}	1.05 ± 0.02^{Aa}	1.07 ± 0.02^{Aa}
Peroxide value (meq/kg)	SO	0.61 ± 0.02^{Ad}	0.86 ± 0.23^{Ad}	1.89 ± 0.03^{Ac}	2.59 ± 0.35^{Aabc}	2.69 ± 0.63^{Aa}	2.92 ± 0.43^{Aab}	2.25 ± 0.11^{Abc}
	CO	0.11 ± 0.02^{Bb}	0.07 ± 0.05^{Bb}	0.11 ± 0.02^{Cb}	0.08 ± 0.03^{Cb}	0.11 ± 0.03^{Bb}	0.14 ± 0.05^{Ca}	0.24 ± 0.06^{Ca}
	PO	0.15 ± 0.06^{Be}	1.09 ± 0.14^{Ad}	1.28 ± 0.08^{Bcd}	1.74 ± 0.04^{Bb}	2.26 ± 0.20^{Aa}	1.43 ± 0.05^{Bc}	1.89 ± 0.05^{Bb}
p-anisidine value	SO	3.04 ± 0.34^{Be}	37.50 ± 2.52^{Ad}	54.20 ± 4.46^{Ac}	76.62 ± 1.58^{Ab}	91.43 ± 2.08^{Aa}	101.61 ± 12.35^{Aa}	99.41 ± 7.78^{Aa}
	CO	0.12 ± 0.05^{Cd}	0.72 ± 0.02^{Cc}	0.92 ± 0.09^{Cc}	0.85 ± 0.06^{Cc}	1.70 ± 0.04^{Cb}	1.93 ± 0.33^{Bb}	2.46 ± 0.34^{Ba}
	PO	6.16 ± 0.69^{Ag}	18.41 ± 1.59^{Bf}	39.29 ± 1.04^{Be}	52.85 ± 0.54^{Bd}	68.45 ± 1.28^{Bc}	92.66 ± 0.5^{Ab}	102.87 ± 2.11^{Aa}
TOTOX value	SO	4.27 ± 0.30^{Be}	39.45 ± 2.82^{Ad}	58.01 ± 4.42^{Ac}	81.39 ± 1.62^{Ab}	97.24 ± 3.53^{Aa}	106.97 ± 12.17^{Aa}	103.80 ± 7.93^{Aa}
	CO	0.32 ± 0.07^{Cd}	0.92 ± 0.00^{Cc}	1.17 ± 0.13^{Cc}	1.03 ± 0.04^{Cc}	1.95 ± 0.06^{Cb}	2.27 ± 0.32^{Bb}	2.88 ± 0.41^{Ba}
	PO	6.45 ± 0.56^{Ag}	20.58 ± 1.32^{Bf}	41.86 ± 1.19^{Be}	56.33 ± 0.47^{Bd}	72.98 ± 0.87^{Bc}	95.52 ± 0.41^{Ab}	106.65 ± 2.01^{Aa}
CD value (ε1% 1cm λ235)	SO	0.18 ± 0.01^{Be}	0.21 ± 0.01^{Acd}	0.22 ± 0.03^{Bd}	0.25 ± 0.03^{Ac}	0.26 ± 0.06^{Ac}	0.27 ± 0.03^{Aa}	0.25 ± 0.01^{Ab}
	CO	0.11 ± 0.02^{Ba}	0.11 ± 0.01^{Ba}	0.10 ± 0.00^{Ca}	0.11 ± 0.02^{Ba}	0.10 ± 0.01^{Ba}	0.13 ± 0.01^{Ba}	0.12 ± 0.00^{Ba}
	PO	0.27 ± 0.04^{Aa}	0.23 ± 0.02^{Aa}	0.28 ± 0.03^{Aa}	0.26 ± 0.03^{Aa}	0.27 ± 0.03^{Aa}	0.27 ± 0.02^{Aa}	0.25 ± 0.01^{Aa}
CT value (ε1% 1cm λ270)	SO	0.67 ± 0.01^{Ac}	0.72 ± 0.00^{Bab}	0.71 ± 0.02^{Aab}	0.72 ± 0.01^{Aa}	0.72 ± 0.03^{Ab}	0.72 ± 0.00^{Bab}	0.73 ± 0.01^{Aab}
	CO	0.06 ± 0.00^{Cd}	0.07 ± 0.00^{Ccd}	0.07 ± 0.00^{Bbc}	0.07 ± 0.00^{Bbc}	0.07 ± 0.00^{Bbc}	0.08 ± 0.01^{Cb}	0.09 ± 0.01^{Ba}
	PO	0.59 ± 0.02^{Bb}	0.75 ± 0.01^{Aa}	0.81 ± 0.11^{Aa}	0.71 ± 0.02^{Aa}	0.73 ± 0.01^{Aa}	0.77 ± 0.00^{Aa}	0.73 ± 0.01^{Aa}
Iodine value (g Iodine /100g oil)	SO	138.90 ± 3.65^{Aab}	140.91 ± 3.47^{Aa}	134.22 ± 1.16^{Abc}	133.54 ± 3.08^{Abc}	130.16 ± 4.88^{Ac}	126.44 ± 2.27^{Ad}	125.03 ± 3.24^{Ad}
	CO	9.62 ± 0.22^{Ca}	10.02 ± 0.15^{Ca}	9.89 ± 0.32^{Ca}	9.79 ± 0.05^{Ca}	9.99 ± 0.22^{Ca}	9.86 ± 0.16^{Ca}	9.96 ± 0.01^{Ca}
	PO	36.25 ± 0.58^{Ba}	36.99 ± 1.55^{Ba}	36.51 ± 3.64^{Ba}	37.44 ± 0.96^{Ba}	36.04 ± 2.42^{Ba}	37.49 ± 0.27^{Ba}	35.82 ± 1.31^{Ba}

Total polar compounds (%)	SO	8.57±0.66^{Bd}	13.52±0.74^{Bc}	14.91±1.94^{Bbc}	15.24±1.74^{Abc}	18.62±3.71^{Ab}	19.15±2.46^{Bb}	31.41±1.12^{Aa}
	CO	13.55±2.62^{Bc}	13.24±0.06^{Bc}	15.69±1.90^{Bbc}	19.10±2.70^{Aab}	18.75±2.34^{Aab}	19.56±0.32^{Bab}	22.13±0.37^{Ba}
	PO	19.81±1.58^{Ac}	18.67±0.39^{Ac}	24.32±0.49^{Abc}	22.98±3.95^{Abc}	26.54±2.89^{Ab}	27.46±3.01^{Aab}	33.00±3.17^{Aa}
Smoke point (℃)	SO	239±1.41^{Aa}	235±4.24^{Aab}	236±2.83^{Aab}	234.5±0.71^{Aab}	233.5±2.12^{Aab}	231±1.41^{Abc}	226.5±2.12^{Ac}
	CO	174±1.41^{Ca}	168±2.83^{Cb}	169.5±0.71^{Cab}	170±2.83^{Cab}	166.5±3.54^{Cb}	168.5±0.71^{Bb}	168.5±0.71^{Bb}
	PO	208.5±0.71^{Ba}	201±1.41^{Bb}	190±0.00^{Bc}	177.5±2.12^{Bd}	179.5±0.71^{Bd}	169.5±0.71^{Be}	172±2.83^{Be}

Values are means ± standard deviation for triplicate analyses. Different superscripts letters (A-C) show the significant difference ($p < 0.05$) between the means of each oil type (column). Different superscripts letters (a-g) show the significant difference ($p < 0.05$) between the means of each frying hours (row).

(Abbreviation: SO - Sunflower oil, CO - Coconut oil, PO - Palm oil)

oil, as palm oil contains a higher amount of oleic acid and less amount of linoleic acid, whereas, sunflower oil contains 48.3-74% of linoleic acid.

As shown in Table 1, the CD value of coconut oil and palm oil was not significantly different (p>0.05) throughout the frying. Sunflower oil showed a significant increase (p<0.05) in CD value with frying hours. It indicates that oxidation of coconut oil and palm oil was lower than in sunflower oil. Less CD value of coconut oil indicates that it is comparatively more stable to oxidation. During the oxidation of unsaturated fatty acids, CDs are produced to attain a more stable radical. During oxidation, linoleic acid produces stable conjugated diene (Choe and Min, 2007). A high amount of CDs is produced when the amount of polyunsaturated fatty acids in the oil is high (Sayyad, 2017). Accordingly, in this study, sunflower oil showed a significant increase (p<0.05) in CD value indicating the susceptibility of sunflower oil to oxidation than the other two oils which is mainly attributed to the high amount of linoleic acid. All oils showed less significant changes in CT values with frying hours. Comparatively very low CT value of coconut oil indicates that coconut oil is more stable to oxidation.

Coconut oil had the lowest iodine value followed by palm oil and sunflower oil. The iodine value of coconut oil and palm oil did not change significantly (p>0.05) throughout the frying, whereas sunflower oil showed a significant reduction (p<0.05) in iodine value (rate of reduction was 1.15/ hour of frying) (Table 1). Iodine value is a measure of the degree of saturation of the oils and the differences in the changes in iodine value of the oils could be explained by the differences in the fatty acid composition of the oils and the susceptibility of the fatty acids to oxidation. The saturated fatty acids are highly stable to oxidation. Comparatively, monounsaturated fatty acids are more stable to oxidation than polyunsaturated fatty acids (Santos *et al.,* 2019). Coconut oil is a highly saturated oil (>90% of saturated fatty acids) and was more stability to oxidation reflected by non-significant changes in the iodine value during frying. Sunflower oil contains a higher amount of polyunsaturated fatty acids and palm oil contain a higher amount of oleic acid. Thus the highly significant changes in the iodine value in sunflower oil are due to the comparatively high amount of linoleic acid, which can be readily oxidized (Yun and Surh, 2012). Similarly, the iodine value of refined olive pomace oil decreased from 87.4 to 78.8 (Ben Hammouda *et al*., 2018). Further, the changes in the iodine value of all three oils is in agreement with the changes in the fatty acid composition of the oils observed during frying as shown in Tables 3, 4 and 5.

The percentage of total polar compounds in frying oils has been widely used as a reliable indicator to monitor the deterioration of frying oil (Li *et al*., 2019). Total polar compounds measure the total amount of newly formed compounds with high polarity and oxidized fatty acids (Alavijeh *et al.*, 2015). The total polar content of all three oils increased significantly (p<0.05) throughout frying (Table 1). Thus indicates that the rate of generation of polar compounds had increased with frying hours. The initial total polar content of palm oil was significantly higher (p<0.05) than the other two oils. It could be due to the poor initial quality of palm oil. The rate of increase in the total polar content of sunflower oil, coconut oil and palm oil were 1.90, 0.715 and 1.10 % /hour of frying, respectively with coconut oil exhibiting the least rate of increase in polar compounds. It further indicates that coconut oil showed better quality during frying than others. The total polar content of sunflower oil showed a high rate of increase compared to palm oil., It is noteworthy that until twelve hours of frying, the coconut oil did not reach the maximum acceptable limit of polar compounds (24-27%) (Firestone, 2007). An abrupt increase (6.13% / hour of frying) in the total polar content of sunflower oil from ten hours to

Table 2: Correlation analysis of the chemical parameters of sunflower oil, coconut oil and palm oil during deep frying.

Variable	AV	PV	*p*-AV	TOTOX	CD	CT	TPC
Sunflower oil							
AV							
PV	0.30						
p-AV	0.55	0.93					
TOTOX	0.54	0.94	1.00				
CD	0.42	0.95	0.99	0.99			
CT	0.38	0.73	0.84	0.84	0.86		
TPC	0.93	0.56	0.79	0.78	0.69	0.65	
IV	-0.71	-0.83	-0.90	-0.90	-0.84	-0.56	-0.84
Coconut oil							
AV							
PV	-0.72						
p-AV	-0.97	0.78					
TOTOX	-0.97	0.80	1.00				
CD	-0.69	0.57	0.63	0.64			
CT	-0.92	0.83	0.91	0.92	0.65		
TPC	-0.95	0.70	0.88	0.87	0.62	0.88	
IV	-0.46	0.19	0.57	0.56	0.06	0.48	0.27
Palm oil							
AV							
PV	0.71						
p-AV	0.99	0.74					
TOTOX	0.99	0.76	1.00				
CD	0.09	-0.07	0.06	0.05			
CT	0.47	0.56	0.45	0.46	0.04		
TPC	0.94	0.66	0.94	0.94	0.15	0.36	
IV	-0.09	-0.08	-0.06	-0.06	-0.12	0.23	-0.36

(Values represent the Spearman's correlation coefficient (r) for the linear analysis, Abbreviations: AV - Acid value, PV - Peroxide value, p-AV - p-Anisidine value, TOTOX - Total oxidation, CD - Conjugated dienes, CT - Conjugated trienes, TPC - Total polar compounds, IV - Iodine value)

twelve hours of frying indicates that the oil has deteriorated and reached the maximum acceptable limit of total polar compounds during ten hours of frying. Palm oil reached the maximum acceptable limit for polar compounds after four hours of frying earlier than sunflower oil. This could be due to the fact that the initial amount of polar compounds was significantly higher ($p<0.05$). Despite the initial difference in total polar content, at the end of twelve hours of continuous frying, both sunflower oil and palm oil reached the values which were not significant ($p>0.05$). That is, the degree of deterioration was higher in sunflower oil than in palm oil. Fresh refined olive pomace oil had 7.3% of total polar compounds which increased up to 30.6% after frying for 60 cycles (Hammouda *et al*., 2018).

The smoke point is related to the buildup of decomposition products during heating and the formation of smoke indicates the degradation of the quality of the frying oil. A low smoke point is generally related to low thermal stability of the oil and vice-versa (Khor *et al.,* 2019). In the present study, sunflower oil had the highest initial smoke point followed by palm oil and coconut oil (Table 1). Chain length is the important factor that affect the smoke point of the oils. Oils with a high amount of long chain fatty acids have higher smoke points than the oils containing high amount of short chain fatty acids (Katragadda *et al*., 2010). Among the three oils used in this study, sunflower oil has a high proportion of long chain fatty acids. Accordingly, in this study, sunflower oil showed the highest smoke point whereas coconut oil showed the lowest. Smoke point of all three oils significantly reduced ($p<0.05$) with frying hours. The reduction in smoke point was consistent with the increase in the free fatty acid content of the oil.

As shown in Table 2, all chemical parameters determined for sunflower oil and palm oil, during frying, except iodine value, showed positive correlations among them. Iodine value had negative relationships with other parameters. While the iodine value decreased the other parameters increased with increasing oxidation. The acid value of coconut oil showed a negative relationship with other parameters. This is due to the reduction in the acid

Table 3: Changes in the fatty acid composition of sunflower oil during continuous deep frying.

Fatty acid/ Fatty acidgroup	Frying duration (hours)						
	0	2	4	6	8	10	12
C4-C14	1.95±0.91^{a}	0.94±0.05^{a}	4.31±0.25^{a}	1.41±0.32^{a}	1.95±0.26^{a}	3.32±1.75^{a}	2.20±0.02^{a}
C16:0	6.80±0.20^{c}	7.18±0.11^{ab}	6.88±0.02^{bc}	7.12±0.02^{abc}	7.13±0.15^{abc}	7.39±0.16^{a}	7.40±0.20^{a}
C18:0	3.84±0.14^{b}	4.03±0.05^{ab}	3.86±0.01^{b}	3.98±0.00^{ab}	4.06±0.14^{ab}	4.12±0.02^{a}	4.18±0.10^{a}
C18:1	23.44±0.1^{bcd}	23.27±0.22^{cd}	22.55±0.07^{d}	24.14±0.06^{abc}	24.38±0.53^{ab}	24.58±0.39^{a}	24.87±0.69^{a}
C18:2 t	0.61±0.08^{c}	0.71±0.01^{b}	0.690±0.01^{b}	0.68±0.01^{b}	0.66±0.04^{b}	0.71±0.00^{a}	0.72±0.03^{a}
C18:2	61.04±0.37^{ab}	61.76±0.61^{a}	59.71±0.14^{abc}	60.57±0.06^{ab}	59.22±1.16^{bcd}	57.43±1.05^{d}	58.20±1.54^{cd}
C20:0	0.26±0.0^{a}	0.29±0.0^{a}	0.27±0.00^{a}	0.31±0.04^{a}	0.31±0.04^{a}	0.29±0.01^{a}	0.29±0.01^{a}
C20:1	0.08±0.01^{a}	0.09±0.01^{a}	0.07±0.02^{b}	0.10±0.01^{a}	0.10±0.01^{a}	0.120.03±^{a}	0.11±0.04^{a}
C18:3	0.16±0.02^{a}	0.15±0.04^{a}	0.15±0.10^{a}	0.16±0.04^{a}	0.16±0.21^{a}	0.17±0.01^{a}	0.67±0.01^{a}
Others	1.83±0.31^{ab}	1.57±0.02^{b}	1.53±0.00^{b}	1.54±0.12^{b}	2.03±0.27^{a}	1.84±0.11^{ab}	1.85±0.09^{ab}

C16:0-Palmitic acid, C18:0-Stearic acid, C18:1-Oleic acid, C18:2t- Trans isomers of linoleic acid (trans 9,12 octadecadienoic acid, trans 9, cis12 octadecadienoic acid, cis 9, trans 12 octadecadienoic acid), C18:2-Linoleic acid, C20:0-Arachidic acid, C20:1-Paullinic acid, C18:3-α-Linolenic acid. Values are means ± standard deviation for triplicate analyses. Means in each row followed by different superscripts letters (a-g) are significantly different (p< 0.05).

Table 4: Changes in the fatty acid composition of coconut oil during continuous deep frying.

Fatty acid/ Fatty acid group	Frying duration (hours)						
	0	2	4	6	8	10	12
C6:0	0.74±0.01^{a}	0.68±0.01^{bc}	0.68±0.00^{bc}	0.69±0.01^{b}	0.68±0.00^{bc}	0.69±0.00^{b}	0.67±0.01^{c}
C8:0	8.41±0.04^{a}	7.77±0.02^{d}	7.81±0.01^{cd}	7.92±0.01^{b}	7.90±0.07^{b}	7.89±0.04^{bc}	7.66±0.01^{e}
C10:0	5.65±0.03^{a}	5.29±0.01^{d}	5.30±0.01^{cd}	5.38±0.00^{bc}	5.32±0.03^{cd}	5.43±0.07^{b}	5.19±0.00^{e}
C12:0	41.72±1.12^{a}	41.08±1.08^{a}	40.40±1.80^{a}	40.78±1.82^{a}	41.23±0.47^{a}	41.26±1.20^{a}	39.42±2.61^{a}
C14:0	18.69±1.07^{a}	17.84±2.06^{a}	17.99±1.51^{a}	18.16±0.80^{a}	18.09±0.74^{a}	18.22±0.03^{a}	17.34±1.80^{a}
C16:0	10.16±0.47^{a}	9.79±0.51^{a}	9.82±0.75^{a}	9.92±0.80^{a}	9.91±0.62^{a}	10.04±0.60^{a}	9.73±1.14^{a}
C18:0	3.12±0.04^{a}	3.07±0.06^{a}	3.11±0.10^{a}	3.10±0.00^{a}	3.15±0.13^{a}	3.16±0.10^{a}	3.02±0.12^{a}
C18:1	7.03±0.14^{a}	6.96±0.23^{a}	7.02±0.20^{a}	7.13±0.41^{a}	7.20±0.25^{a}	7.23±0.11^{a}	6.90±0.40^{a}
C18:2 t	0.11±0.03^{a}	0.10±0.00^{a}	0.09±0.00^{a}	0.09±0.00^{a}	0.11±0.01^{a}	0.10±0.00^{a}	0.11±0.00^{a}
C18:2	1.50±0.11^{a}	1.56±0.20^{a}	1.53±0.10^{a}	1.52±0.20^{a}	1.53±0.25^{a}	1.52±0.28^{a}	1.48±1.06^{a}
Others	2.89±0.42^{g}	6.86±0.07^{b}	6.24±0.00^{c}	5.40±0.00^{d}	4.76±0.09^{e}	4.32±0.03^{f}	8.44±0.05^{a}

C6:0-Caproic acid, C8:0-Caprylic acid, C10:0-Capric acid, C12:0-Lauric acid, C14:0-Myristic acid, C16:0-Palmitic acid, C18:0-Stearic acid, C18:1-Oleic acid, C18:2t- Trans isomers of linoleic acid (trans 9,12 octadecadienoic acid, trans 9, cis12 octadecadienoic acid, cis 9, trans 12 octadecadienoic acid), C18:2-Linoleic acid. Values are means ± standard deviation for triplicate analyses. Means in each row followed by different superscripts letters (a-g) are significantly different (p< 0.05).

Table 5: Changes in the fatty acid composition of palm oil during continuous deep frying.

Fatty acid/ Fatty acid group	Frying duration (hours)						
	0	2	4	6	8	10	12
C4:0-C14:0	3.22±1.14^{ab}	4.14±0.03^{a}	2.38±0.01^{b}	2.63±0.02^{b}	2.60±0.00^{b}	2.60±0.01^{b}	2.46±0.01^{b}
C16:0	36.61±1.40^{a}	35.49±1.20^{a}	37.12±1.04^{a}	37.17±1.00^{a}	37.29±0.16^{a}	37.49±0.15^{a}	37.91±1.12^{a}
C18:0	3.94±0.15^{a}	3.91±0.12^{a}	4.06±0.41^{a}	4.04±0.10^{a}	4.07±0.07^{a}	4.14±0.32^{a}	4.21±0.40^{a}
C18:1	42.42±1.50^{a}	42.89±1.07^{a}	43.24±0.54^{a}	42.92±1.10^{a}	43.23±0.90^{a}	43.16±0.80^{a}	43.10±1.00^{a}
C18:2 t	1.14±0.01^{b}	1.13±0.00^{b}	1.10±0.01^{b}	1.20±0.00^{a}	1.19±0.01^{a}	1.20±0.00^{a}	1.21±0.00^{a}
C18:2	11.68±0.14^{a}	11.39±0.03^{b}	11.08±0.02^{c}	11.08±0.01^{c}	10.61±0.20^{d}	10.06±0.40^{e}	9.72±0.60^{f}
C20:0	0.37±0.00^{a}	0.36±0.01^{a}	0.39±0.0^{a}	0.37±0.01^{a}	0.38±0.00^{a}	0.39±0.00^{a}	0.38±0.00^{a}
C20:1	0.19±0.00^{a}	0.17±0.01^{a}	0.16±0.03^{a}	0.17±0.00^{a}	0.15±0.00^{b}	0.15±0.00^{b}	0.13±0.00^{b}
C18:3	0.16±0.00^{a}	0.15±0.00^{b}	0.15±0.01^{b}	0.15±0.00^{a}	0.15±0.00^{a}	0.14±0.00^{c}	0.14±0.00^{c}
Others	0.27±0.00^{e}	0.35±0.00^{c}	0.31±0.00^{d}	0.26±0.00^{e}	0.30±0.00^{d}	0.72±0.00^{b}	0.83±0.02^{a}

C16:0-Palmitic acid, C18:0-Stearic acid, C18:1-Oleic acid, C18:2t- Trans isomers of linoleic acid (trans 9,12 octadecadienoic acid, trans 9, cis12 octadecadienoic acid, cis 9, trans 12 octadecadienoic acid), C18:2-Linoleic acid, C20:0-Arachidic acid, C20:1-Eicosenoic acid, C18:3-α-Linolenic acid. Values are means ± standard deviation for triplicate analyses. Means in each row followed by different superscripts letters (a-g) are significantly different (p< 0.05)

value of coconut oil with increasing frying with the possible reason that the coconut oil contains a higher proportion of shorter chain fatty acids that could be volatilized during frying.

The changes in the fatty acid composition of sunflower oil, coconut oil and palm oil during deep frying are shown in Tables 3, 4 and 5 respectively. During frying, each oil showed different behaviour in the changes in the fatty acid composition. Fewer changes in the fatty acid composition were observed in coconut oil followed by palm oil and more changes were observed in sunflower oil. Significant reductions (p<0.05) in linoleic acid content were observed with frying hours in sunflower oil and palm oil, whereas, changes in the oleic acid content were non-significant (p>0.05). These results were in agreement with previous studies. For instance, in a study to determine the stability of different types of edible oil (refined coconut oil, sunflower oil, olive oil and vegetable shortening) during frying of potato chips, unsaturated fatty acids content decreased significantly after 80 cycles of frying (Yu *et al*., 2017). This pattern of change could be attributed to the higher stability of oleic acid compared to polyunsaturated fatty acids (Asadi and Farahmandfar, 2020). The reduction in linoleic acid content indicates that the unsaturation of sunflower oil and palm oil was decreased during frying and this was evident by the pattern of changes in iodine values of these oils.

The reductions in the linoleic acid content were in line with the increase in the *trans* fat content of both oils. All three oils contained small amounts of *trans* fat which could have formed during the extraction and processing of the oil using high temperature. Another study (Sajitha, 2016) also reported the presence of a small quantity of *trans* fat in coconut oil. The *Trans* fat content of coconut oil was not significantly different (p>0.05) with frying hours indicating that *trans* fats are not formed in the coconut oil during frying. The rate of formation of *trans* fat in the palm oil (1.11% /hour) was higher than that of sunflower oil (0.66% /hour). Considering the composition of monounsaturated and polyunsaturated fatty acids composition of both oils, theoretically, the rate of formation of *trans* fat in the sunflower oil should be higher than that in palm oil. It is reported that a high increase of the *trans* isomers of polyunsaturated fatty acids is detected in refined sunflower oil which contains high amount of polyunsaturated fatty acids and a very small increase of these compounds is reported in refined olive oil which contain high amount of monounsaturated fatty acids during repeated pan and deep frying (Zribi *et al*., 2016). The higher rate of *trans* fat formation in the palm oil compared to sunflower oil in the present study could be due to the higher initial level of *trans* fat in the palm oil (1.14%) than sunflower oil (0.61%), that is the palm oil has undergone isomerization already. Further, a higher percentage of initial polar compounds was observed in the palm oil than in sunflower oil (Table 1) indicates that the palm oil used in this study has already undergone chemical alterations.

The formation of *trans* fats in frying oils depends on the temperature of frying as well as the use time of the oil (Oteng and Kersten, 2020). The pattern of formation of *trans* fats in frying oils reported in this study is in agreement with Ben Hammouda *et al.* (2018) who evaluated the refined olive pomace oil pure and its blend coconut oil and reported that at the end of frying at 180 °C, an increase in the formation of *trans* fats was observed in both pure refined olive pomace and its blend coconut oil samples. Based on these results, it can be concluded that coconut oil remained more stable during frying compared to the other two oils. Palm oil and sunflower oil underwent the oxidation of polyunsaturated fatty acids and *trans* fats are also generated. Major *trans* fatty acids identified in all three oils were a mixture of *trans* isomers of linoleic acid (*trans*-9, *trans*-12-Octadecadienoic acid, *cis*-9, *trans*-12-Octadecadienoic acid, *trans*-9, *cis*-12-Octadecadienoic acid and *cis*-9, *cis*-12-Octadecadienoic acid).

CONCLUSION

This study demonstrated that all three oils showed significant differences in their stability during frying. During continuous deep frying, coconut oil showed fewer alterations in the parameters examined than the sunflower oil and palm oil, that is, coconut oil is more stable than the other two oils during continuous deep frying. According to the permitted level of polar compounds in frying oil, coconut oil showed acceptable quality until twelve hours of frying, palm oil showed until four hours of frying and sunflower oil showed until ten hours of frying. Even though sunflower oil showed stability up to 10 hours of frying in terms of polar compounds, the degree of oxidation and formation of *trans* fats during frying indicate that sunflower oil is not suitable for continuous deep frying. Further studies are needed to be carried out to study more market samples of the studied oil types to draw a strong conclusion and recommendation on the acceptable usage times of frying.

DECLARATION OF CONFLICT OF INTEREST

The authors declare no conflict of interest.

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