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Changes in chemical properties and oxidative stability of refined vegetable oils during short-term deep-frying cycles

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Abstract

In this study, changes in chemical properties, oxidative stability, fatty acid composition (FAC) and Fourier transform infrared (FTIR) spectra of soybean (SBO), rice bran (RBO), canola (CNO), cottonseed (CSO), and sunflower (SFO) oils during short-term deep frying of chickpea splits were determined. Free fatty acid (FFA), peroxide value (PV), p-anisidine value (p-AV), and viscosity of refined vegetable oils increased while the oxidative stability index (OSI) decreased with increment in frying cycles (FCs). The minimum changes in FFA, PV, p-AV, viscosity, and OSI were observed in SBO till fifth FCs. Oil uptake was also low in SBO and high in CNO after 5th and 10th FCs. CNO, RBO, and CSO exhibited higher changes in FAC and FTIR spectra (peak intensities at 3,009, 2,925, 2,854, and 1,745 cm^{-1}) compared to SBO and SFO with increment in FCs. Thus, SBO with admissible $\omega 6/\ \omega 3$ ratio (9.42:1) is suitable for short-term deep frying of chickpea splits.

Practical applications

In the deep-frying process of chickpea splits, five vegetable oils behaved differently in quality and stability characteristics. Soybean oil exhibited minimum changes in FFA content, viscosity, and OSI with increment in frying cycles. Oil uptake was lowest in soybean oil and it contains a desirable FAC for health benefits. Thus, soybean oil is recommended for short-term deep-frying operations among the studied vegetable oils.

1 | INTRODUCTION

Deep frying is a popular method of food preparation that enhances the sensory attributes and gives crispy texture and unique flavor to food (Zribi et al., 2014). It is a method of cooking foodstuff in the presence of air and moisture by immersing in heated oil (Multari, Marsol-Vall, Heponiemi, Suomela, & Yang, 2019). Deep frying serves as a simultaneous heat and mass transfer medium that contributes to the desirable quality of deep-fat fried foods which is popular among consumers (Akil et al., 2015). The high-temperature processing of cooking oils during the production of fried foods leads to the oxidation and production of free radicals, anti-nutritional, and toxic substances in edible oils (Ramadan, 2015; Tavakoli, Naderi, Jafari, & Naeli, 2019). The use of cooking oils for multiple frying over prolonged and repeated periods of time is a common practice followed by street vendors and fast-food restaurants to reduce the frying costs (Nayak, Dash, Rayaguru, & Krishnan, 2016). The length of the deep-frying process at a high temperature can alter the chemical properties of vegetable oil (Giuffre, Capocasale, Zappia, & Poiana, 2017). The formation of phytosterols oxidation products during deep- and pan-frying in vegetable oils for long-duration could have adverse effects on the human health (Ramadan, 2015).

Abbreviations: AI, atherogenic index; CNO, canola oil; CSO, cottonseed oil; FAs, fatty acids; FAC, fatty acid composition; FCs, frying cycles; FFA, free fatty acid; FTIR, Fourier transform infrared spectroscopy; MUFAs, monosaturated fatty acids; PC, principal component; p-AV, p-anisidine value; PUFAs, polyunsaturated fatty acids; PV, peroxide value; RBO, rice bran oil; RVOs, refined vegetable oils; SBO, soybean oil; SFAs, saturated fatty acids; SFO, sunflower oil; TI, thrombogenic index; OSI, oxidative stability index; UFAs, unsaturated fatty acids.

During continuous and repeated exposure to air, moisture, and high temperature, the oils undergo complex series of chemical reactions (hydrolysis, oxidation, and polymerization) resulting in the thermal and oxidative decomposition (Dobarganes & Márquez-Ruiz, 2015; Nayak et al., 2016). Hydrolysis involves the breakage of triglycerides into free fatty acid (FFA), monoglycerides, glycerols, and diglycerides. Oxidation results into formation of primary and secondary oxidation products that impart unpalatable odor and flavor to the oil. The peroxides formed undergo a series of complex reactions like isomerization, polymerization, and cyclization to produce several secondary oxidation volatile and nonvolatile metabolites (Multari et al., 2019). Polymerization of UFAs forms dimers, oligomers, and polymers in frying oil (Nayak et al., 2016). Intermittent frying at high temperatures accelerates the process of oxidation and polymerization, thus degrade the oil quality (Choe & Min, 2007; Dobarganes & Márquez-Ruiz, 2015). Moreover, the adverse chemical reactions occurring in cooking oils can also put the consumer's health at serious risk (Tavakoli et al., 2019).

The volatile and nonvolatile compounds formed during deep frying changes the physical and chemical properties of frying oil (Choe & Min, 2007). The volatile compounds evaporate during deep frying while nonvolatile compounds accumulate in oil as frying continues. The chemical compounds formed during deep-frying changes viscosity, smoke point, and color of the frying oil (Hassanien & Sharoba, 2014; Nayak et al., 2016; Ramadan, Amer, & Sulieman, 2006). During repeated deep frying in oil, by-products such as FFA, alcohols, aldehydes, ketones, cyclic compounds, dimers, and polymers are formed (Nayak et al., 2016). The primary and secondary derivatives formed during deep frying in oils are absorbed by foodstuff, thus have an adverse effect on human health (Bansal et al., 2010; Nayak et al., 2016). The formation of free radicals and toxic substances in oil can contribute to aging and degenerative diseases (Soleimanifar, Niazmand, & Jafari, 2019). The consumption of repeatedly heated cooking oils causes genotoxicity (Dung, Wu, & Yen, 2006), carcinogenicity (Srivastava et al., 2010), hypertension (Soriguer et al., 2003), and cardiovascular diseases (Ng et al., 2014).

The susceptibility of the edible oils to oxidation depends largely on the fatty acid composition (FAC) as the oils with high UFAs are susceptible to oxidation. The study of quality characteristics and safety aspects of vegetable oils used in the frying process requires attention due to the growing demand of deep-fat fried snack foods (Multari et al., 2019; Tarmizi, Hishamuddin, & Razak, 2019). Determination of changes in oil during FCs is needed for the development of strategies to maintain the quality of oils. The heated oils should be tested for FFA, p-anisidine value (p-AV), peroxide value (PV), and fatty acid profile at regular intervals to monitor the quality of frying oil and fried food products. The study of changes in chemical properties is important to compare and find the most suitable oil for deep frying along with advantages for human health. Among the traditionally fried food products, chickpea splits (Figure 1) are commonly relished as anytime fried snack food by consumers of all age groups throughout India. The fried chickpeas are ready to eat, easy to carry, shelf-stable, and nutritious snack food (Bozdemir, Güneşer, & Yılmaz, 2015). Considering the literature review, there have been no previous reports on assessing the effect of FCs during short-term deep frying of chickpea splits on quality characteristics of refined vegetable oils (RVOs). The present study evaluated the changes in chemical properties, oxidative stability index (OSI), and FAC of five commonly consumed RVOs (sunflower, rice bran, soybean, cottonseed, and canola) during short term deep-frying cycles of chickpea splits. Moreover, this study also compared the suitability of different RVOs in short-term frying operations.

2 | MATERIAL AND METHODS

2.1 | Materials

Rice bran oil (RBO), cottonseed oil (CSO), sunflower oil (SFO), soybean oil (SBO), canola oil (CNO), and black chickpea (*Cicer arietinum*) splits were purchased from the market of Amritsar, India. All chemicals of analytical laboratory grade (AR) were used in this study.

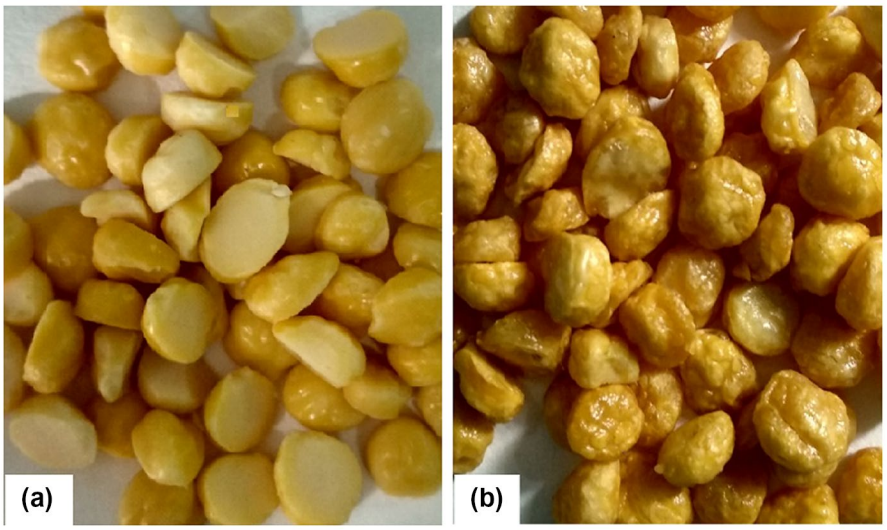

FIGURE 1 (a) Unfried and (b) fried chickpea splits

2.2 | Methods

2.2.1 | Frying process

Black chickpea splits (100 g) were washed and soaked in 500 ml filtered drinking water for 12 hr at room temperature (28°C ± 1°C) for hydration purposes. The soak water was drained off and chickpea splits were dried for 10 min at room temperature. The chickpea splits were then fried at 175°C for 2 min in RVO. A 2-L electrical fryer (Inalsa Professional 2, 1700 W, India) equipped with a thermostat, and had steel wire frying basket, was used for frying experiments. The frying pot was filled with 1 L of RVO and heated for 10 min at 175°C. About 100 g of hydrated chickpea splits deep-fried for 2 min and drained in the frying basket for 1 min constituted one FC. The chickpea splits were checked for moisture content before frying and for oil uptake after frying. The moisture content of hydrated chickpea splits was 49.80% ± 0.81%. After each FC, the electrical fryer was put off and RVO was allowed to cool down at room temperature. After the 1st, 5th, and 10th FCs, 100 ml of oil was collected from frying pot and filtered. The oil in frying pot was replenished to 1 L, followed by 10 min of reheating at 175°C before starting the next FC. RVOs (fresh and after the 1st, 5th, and 10th FCs) collected were stored in sealed dark glass bottles at −20°C till further analysis.

2.2.2 | FFA content

The FFA content of RVOs (fresh and after the 1st, 5th, and 10th FCs) was measured according to the official Ca. 5a-40 method of American Oil Chemists' Society (AOCS, 1997).

2.2.3 | Peroxide value (PV)

The PV of fresh RVOs and after FCs was measured according to the official Cd 8b-90 method of AOCS (1997).

2.2.4 | p-Anisidine value (p-AV)

The p-AV of fresh RVOs and after FCs was measured according to the official Cd 18-90 method of AOCS (2017).

2.2.5 | Oxidative stability index (OSI)

OSI of fresh RVOs and after FCs was studied using Rancimat (892 Professional, Metrohm, Switzerland) as described previously (Kaur, Singh, Kaur, & Singh, 2019). Briefly, the test was carried out at 120°C ± 1.6°C with an airflow of 10 L/hr using 3 g of oil sample. The OSI of RVOs was calculated in terms of the induction period (IP) in hours.

2.2.6 | Viscosity

The viscosity of fresh RVOs and after FCs was studied using the Rheometer (MCR 102 Modular Compact, Anton Paar, Austria) following the method described in a previous study (Kaur et al., 2019).

2.2.7 | Oil uptake in fried chickpea splits

The fried chickpea splits obtained from the 1st, 5th, and 10th FCs were cooled to room temperature and their oil contents were determined according to the official soxhlet extraction method of AOCS (1990). The oil uptake was obtained in terms of percent dry basis (% d.b.) using the following equation.

$$\text{Oil uptake } (\%) = \frac{\text{Of} - \text{Or}}{\text{Or}} \times 100.$$

where Or and Of is the oil content of raw and fried chickpeas splits, respectively.

2.2.8 | Fatty acid composition (FAC)

The FAC of fresh RVOs and after FCs were studied using a Gas chromatography (GC) system according to the official Ce-1h-05 method of AOCS (1997). Fatty acids (FAs) were converted to fatty acid methyl esters (FAMEs) by derivatization and determined by Agilent 7820A GC system (Agilent Technologies, USA) consisting of DB-WAX capillary column and flame ionization detector as described earlier (Suri, Singh, Kaur, Yadav, & Singh, 2019). The results of individual FAs were expressed in terms of relative percentage (g/100 g) of total FAs.

2.2.9 | Atherogenic (AI) and thrombogenic index (TI)

The AI and TI were calculated from the FAC of RVOs according to the following equations described in a previous study (Pereira et al., 2019).

$$\text{AI} = \frac{\text{LA} + 4 \times \text{MA} + \text{PA}}{\sum \text{MUFAs} + \sum \omega 6 + \sum \omega 3}$$

$$\text{TI} = \frac{\text{MA} + \text{PA} + \text{SA}}{(0.5 \times \sum \text{MUFAs}) + (0.5 \times \sum \omega 6) + (3 \times \sum \omega 3)}$$

where LA, MA, PA, SA, $\omega 3$, and $\omega 6$ are lauric, myristic, palmitic, stearic, linoleic, and linolenic acids, respectively.

2.2.10 | FTIR spectroscopy

Infrared (IR) spectra of fresh RVOs and after FCs were acquired using Fourier transform infrared (FTIR) Spectroscopy (Vertex-70, Brucker,

Germany) equipped with ATR assembly using the procedure described in earlier studies (Kaur et al., 2019; Suri, Singh, Kaur, Yadav, & Singh, 2019). IR spectra were recorded in the spectral region of 3500–500 cm^{-1} by accumulating 32 scans/sample at 4 cm^{-1} resolution.

2.3 | Statistical analysis

All the analytical tests and experiments were recorded in triplicate and data reported as mean ± standard deviation (*SD*) values. For statistical assessment, two-way analysis of variance (ANOVA), Principal component analysis (PCA), and Pearson correlation coefficient ([*r*], with significance levels at $p \leq .005$ and $p \leq .05$) were employed for determining the relationship between various parameters of RVOs after FCs. Minitab Software (version 14.12.0, Minitab) was used to apply these statistical tools.

3 | RESULTS AND DISCUSSION

3.1 | Changes in FFA content of RVOs with FCs

The FFA content of fresh RVOs and after FCs are given in Table 1. FFA content ranged between 0.11% and 0.20% in fresh RVOs, with the highest in RBO and the lowest in CNO. This might be due to variation in the oleic acid content of RVOs. CNO had the lowest FFA content due to the high proportion of oleic acid (Kaur et al., 2019). The results concur with the previous study on edible oils (Zribi et al., 2014). FFA content indicates the hydrolysis of triglycerides and decomposition of hydroperoxides in vegetable oils (Kaur et al., 2019). Changes in the level of FFA was observed in RVOs with increment in FCs. The FFA content remained unchanged in RVOs after the 1st FC, while a slight change was observed after the 5th FC, and a significant increase was observed after the 10th FC. The highest change in FFA content was observed in RBO (0.27%) and lowest in SBO (0.15%) after the 10th FC. Similar results of an increase in FFA content of RVOs with an increase in a successive pan and deep-frying sessions were reported in previous studies (Debnath, Rastogi, Krishna, & Lokesh, 2012; Zribi et al., 2014). Ramadan et al. (2006) also reported an increase in FFA content of two different vegetable oil blends by continuous frying of French fries for two consecutive days. The increase in FFA content indicates triacylglycerol hydrolysis and release of free FAs during the deep-frying process (Karimi, Wawire, & Mathooko, 2017). The *F* values of FFA content showed higher significant variation ($p \leq .005$) in RVOs than the FCs (Table 2). FFA content showed a significant positive correlation with FCs ($r = 0.418$, $p \leq .05$) (Table 3).

TABLE 1 Effect of frying cycles on chemical properties, oxidative stability, viscosity and oil uptake of refined vegetable oils

RVOs	FC	FFA (%)	PV (meqO_2/kg)	p-AV	OSI (h)	Viscosity (mPa s)	OU (%)
SBO	0	0.12 ± 0.01^{b}	1.80 ± 0.15^{a}	1.95 ± 0.02^{a}	4.32 ± 0.06^{c}	44.16 ± 0.36^{a}	–
	1	0.12 ± 0.01^{b}	8.67 ± 0.31^{c}	3.66 ± 0.08^{b}	4.11 ± 0.08^{c}	52.23 ± 0.42^{b}	13.5 ± 0.6^{a}
	5	0.13 ± 0.02^{c}	9.23 ± 0.19^{c}	4.17 ± 0.03^{b}	3.93 ± 0.01^{c}	55.31 ± 0.56^{b}	19.4 ± 0.1^{b}
	10	0.15 ± 0.04^{c}	15.33 ± 0.15^{d}	5.96 ± 0.05^{c}	3.03 ± 0.06^{b}	58.27 ± 0.27^{b}	23.5 ± 0.7^{b}
RBO	0	0.20 ± 0.06^{d}	4.82 ± 0.02^{b}	2.69 ± 0.03^{a}	7.10 ± 0.07^{d}	60.25 ± 0.24^{c}	–
	1	0.20 ± 0.06^{d}	8.52 ± 0.16^{c}	3.30 ± 0.03^{b}	5.45 ± 0.10^{c}	72.87 ± 0.39^{d}	31.3 ± 0.3^{c}
	5	0.21 ± 0.09^{e}	9.33 ± 0.15^{c}	4.54 ± 0.04^{b}	2.83 ± 0.09^{b}	73.10 ± 0.36^{d}	38.6 ± 0.5^{d}
	10	0.27 ± 0.10^{f}	16.67 ± 0.15^{d}	7.61 ± 0.09^{d}	2.64 ± 0.10^{b}	76.72 ± 0.20^{d}	46.3 ± 0.8^{e}
CNO	0	0.11 ± 0.05^{a}	4.63 ± 0.46^{b}	4.49 ± 0.07^{b}	5.31 ± 0.07^{c}	45.56 ± 0.46^{a}	–
	1	0.11 ± 0.06^{a}	8.06 ± 0.46^{c}	11.83 ± 0.06^{e}	3.33 ± 0.09^{b}	60.94 ± 0.30^{c}	37.5 ± 0.6^{d}
	5	0.12 ± 0.07^{a}	21.67 ± 0.16^{e}	16.28 ± 0.04^{f}	1.53 ± 0.02^{a}	64.25 ± 0.31^{c}	45.3 ± 0.8^{e}
	10	0.16 ± 0.09^{b}	43.48 ± 0.46^{f}	21.16 ± 0.05^{f}	1.34 ± 0.06^{a}	65.39 ± 0.21^{c}	65.9 ± 0.2^{f}
CSO	0	0.18 ± 0.04^{a}	3.23 ± 0.15^{a}	1.50 ± 0.06^{a}	3.81 ± 0.09^{b}	47.00 ± 0.25^{a}	–
	1	0.18 ± 0.05^{a}	4.67 ± 0.15^{b}	1.95 ± 0.02^{a}	3.14 ± 0.14^{b}	56.23 ± 0.25^{b}	25.8 ± 0.8^{b}
	5	0.19 ± 0.07^{b}	16.54 ± 0.17^{d}	6.01 ± 0.01^{c}	2.88 ± 0.04^{b}	65.12 ± 0.35^{c}	30.1 ± 0.5^{c}
	10	0.24 ± 0.09^{b}	22.45 ± 0.31^{e}	8.33 ± 0.07^{d}	2.83 ± 0.07^{b}	76.54 ± 0.26^{d}	34.5 ± 0.4^{c}
SFO	0	0.12 ± 0.01^{a}	2.44 ± 0.15^{a}	2.83 ± 0.01^{a}	6.39 ± 0.03^{d}	44.65 ± 0.21^{a}	–
	1	0.12 ± 0.01^{a}	8.14 ± 0.31^{c}	5.66 ± 0.05^{c}	4.47 ± 0.06^{c}	53.41 ± 0.92^{b}	19.2 ± 0.6^{b}
	5	0.14 ± 0.02^{a}	14.33 ± 0.15^{d}	8.83 ± 0.06^{d}	2.39 ± 0.07^{b}	58.24 ± 0.35^{b}	23.6 ± 0.9^{b}
	10	0.16 ± 0.04^{b}	25.12 ± 0.18^{e}	12.67 ± 0.07^{e}	2.23 ± 0.05^{b}	60.41 ± 0.35^{c}	32.1 ± 0.7^{c}

Note: Values within the column with different alphabets are significantly different ($p \leq .05$).

Abbreviations: CNO, canola oil; CSO, cottonseed oil; FC, frying cycle; FFA, free fatty acid; OSI, oxidative stability index; OU, oil uptake; PV, peroxide value; p-AV, p-Anisidine value; RBO, ricebran oil; RVOs, refined vegetable oils; SBO, soybean oil; SFO, sunflower oil.

TABLE 2 *F* values from ANOVA analysis of the data (RVOs vs FCs) in Tables 1 and 4

	DF	FFA	PV	p-AV	OSI	Viscosity	SFAs	MUFAs	PUFAs	UFAs	$\omega 6/\omega 3$	AI	TI	OU
RVOs	4	826.71**	276.43**	1,336.40**	1,852.50**	2,392.93**	211,826.67**	979,460.30**	537,055.52**	120,975.14**	16.53**	889.61**	9,732.87**	2,085.13**
FCs	3	11.51**	286.60**	6,788.69**	6,268.83**	7,487.31**	3,586.09**	9,028.24**	12,848.30**	19,771.20**	–	28.36**	380.91**	9,605.51**
Interaction	12	3.64**	25.45**	345.77**	221.54**	395.68**	669.02**	68.20**	3,128.70**	1,620.12**	2.54*	5.24**	72.72**	305.82**

Abbreviations: AI, atherogenic index; DF, degree of freedom; FCs, frying cycles; FFA, free fatty acid; MUFAs, monounsaturated fatty acids; OSI, oxidative stability index; OU, oil uptake; p-AV, p-Anisidine value; PUFAs, polyunsaturated fatty acids; PV, peroxide value; RVOs, refined vegetable oils; SFAs, saturated fatty acids; TI, thrombogenic index; UFAs, unsaturated fatty acids; $\omega 6/\omega 3$, linoleic/linolenic acid ratio.

*$p < .05$, **$p < .005$.

3.2 | Changes in PV of RVOs with FCs

The PV of fresh RVOs and after FCs are given in Table 1. PV indicates the formation of peroxides and hydroperoxides (primary oxidation products) due to the oxidative degradation of oils (Suri et al., 2019). PV ranged between 1.80 and 4.82 meqO_2/kg in fresh RVOs, with the highest in RBO and the lowest in SBO. All five fresh RVOs had PV within the recommended limits of less than 10 meqO_2/kg (FAO/WHO, 2009). PV changes with an increment in FCs. A minor change in PV was observed from the 1st to the 5th FC in SBO (8.67 to 9.23 meqO_2/kg) and RBO (8.52 to 9.33 meqO_2/kg) while the significant change was noticed in CNO (8.06 to 21.67 meqO_2/kg), CSO (4.67 to 16.54 meqO_2/kg), and SFO (8.14 to 14.33 meqO_2/kg). After the 10th FC, the highest change in PV was observed in CNO (43.48 meqO_2/kg) and lowest in SBO (15.33 meqO_2/kg). Similar results of an increase in PV of two vegetable oil blends during continuous frying of French fries for two consecutive days were previously reported (Ramadan et al., 2006). RVOs with a high proportion of UFAs were more sensitive to oxidation and showed more increase in PV with an increment in FCs. An increase in PV during the deep-frying process might be due to the free radical attack on UFAs and the accumulation of primary oxidation products in RVOs (Liu, Wang, Cao, & Liu, 2018). A similar trend in the PV of oils with an increase in deep-frying time was reported in earlier studies (Liu, Li, Cheng, & Liu, 2019). The *F* values of PV showed a higher significant variation ($p \leq .005$) with FCs than the oils (Table 2). PV showed a highly significant positive correlation with FCs and FFA content ($r = 0.903$ and 0.903, respectively, $p \leq .005$) (Table 3).

3.3 | Changes in p-AV of RVOs with FCs

The p-AV determines the formation of secondary oxidation products due to the degradation of hydroperoxides in oils (Zribi et al., 2014). The p-AV of fresh RVOs and after FCs are shown in Table 1. In fresh RVOs, p-AV ranged from 1.50 to 4.49, with the highest in CNO and lowest in CSO. The p-AV observed in fresh RVOs was significantly lower than the permissible limit of 10 for the edible oils (Casal, Malheiro, Sendas, Oliveira, & Pereira, 2010). The low p-AV indicates a low level of secondary oxidation products in fresh RVOs. Similar results have been reported in fresh and unheated edible oils (Casal et al., 2010; Liu et al., 2019; Zribi et al., 2014). The p-AV changes in RVOs with an increment in FCs. The primary oxidation products get decomposed into secondary oxidation products during the deep-frying process which increases the p-AV of RVOs. The highest increase in p-AV from the 1st to the 5th FC was observed in CSO (1.95 to 6.01), CNO (11.83 to 16.28) and SFO (5.66 to 8.83), while the lowest increase was observed in RBO (3.30 to 4.54) and SBO (3.66 to 4.17). After the 10th FC, the highest increase in p-AV was observed in CNO (21.16) and lowest in SBO (5.96) among RVOs. This might be due to the higher susceptibility of CNO to fatty acid oxidation during the deep-frying process. Recent studies had correlated the formation of unsaturated aldehydes during the deep-frying process

TABLE 3 Pearson correlation of various properties of refined vegetable oils after frying cycles

Parameters	FCs	FFA	PV	p-AV	OSI	Viscosity	SFAs	MUFAs	PUFAs
FFA	0.418*								
PV	0.903**	0.903**							
p-AV	0.737**	0.592**							
OSI	−0.768**		−0.494**	−0.426**					
Viscosity	0.535**	0.803**		0.579**	−0.434**				
SFAs		0.804**	−0.278*	0.321*					
MUFAs			0.334*	−0.304*			−0.797**		
PUFAs			−0.379**			−0.434**	0.453**	−0.855**	
UFAs	−0.366**	−0.785**		−0.486**		−0.375**	−0.878**	0.711**	−0.244*
$\omega 6/\omega 3$									0.252*
AI		0.807**				0.555**	0.962**		−0.606**
TI		0.812**				0.556*	0.968**		−0.633**
OU	0.702**		0.658**	0.706**	−0.753**	0.716**			−0.484**

Abbreviations: AI, atherogenic index; FCs, frying cycles; FFA, free fatty acid; MUFAs, monounsaturated fatty acids; OSI, oxidative stability index; OU, oil uptake; p-AV, p-Anisidine value; PUFAs, polyunsaturated fatty acids; PV, peroxide value; SFAs, saturated fatty acids; TI, thrombogenic index; UFAs, unsaturated fatty acids; $\omega 6/\ \omega 3$, linoleic/linolenic acid ratio.

*$p < .05$; **$p < .005$.

with the total amount of PUFAs present in edible oils (Multari et al., 2019; Zribi et al., 2014). The *F* values of p-AV showed a higher significant variation ($p \leq .005$) with FCs than the RVOs (Table 2). The p-AV of RVOs showed highly significant positive correlation with FCs and FFA ($r = 0.737$ and 0.592, respectively, $p \leq .005$) while highly significant negative correlation with OSI ($r = -0.426$, $p \leq .005$) and significant negative correlation with MUFAs ($r = -0.304$, $p \leq .05$) as shown in Table 3.

3.4 | Changes in OSI of RVOs with FCs

OSI determines the shelf life and overall quality of edible oils by measuring the formation of primary or secondary oxidation products in oils. The OSI of fresh RVOs and after FCs are given in Table 1. OSI ranged from 3.81 to 7.10 hr in fresh RVOs, with the highest in RBO and lowest in CSO. OSI depends on the position and number of double bonds in FAs and antioxidant compounds present in oil (Kaur et al., 2019; Li, Liu, Sun, & Liu, 2018; Pereira et al., 2019). The results of OSI for fresh RVOs are in agreement with earlier studies on edible oils (Farhoosh, Einafshar, & Sharayei, 2009; Kaur et al., 2019). OSI of RVOs decreases with an increase in FCs. No significant change in OSI was observed from the 1st to the 5th FC in SBO (4.11 to 3.93 hr) and CSO (3.14 to 2.88 hr). However, a significant decline in OSI was observed in RBO (5.45 to 2.83 hr), SFO (4.47 to 2.39 hr), and CNO (3.33 to 1.53 hr) from the 1st to the 5th FC. SBO showed the highest OSI and CNO showed the lowest OSI after the 10th FC. A similar trend of decrease in OSI with an increase in a successive pan and deep-frying sessions in olive oil, corn oil, SBO, and SFO have been reported in a previous study (Zribi et al., 2014). The susceptibility or resistance of oil to oxidation under deep-frying conditions depends on the FAC of oils (Liu et al., 2018; Multari et al., 2019). RVOs with low SFAs content (CNO and SFO) showed higher susceptibility to oxidative degradation during FCs (Table 4). The *F* values of OSI showed higher significant variation ($p \leq .005$) with FCs than the RVOs (Table 2). OSI showed a highly significant negative correlation with FCs, PV, and p-AV ($r = -0.768$, -0.494, -0.426, respectively, $p \leq .005$) (Table 3).

3.5 | Changes in viscosity of RVOs with FCs

The viscosity of RVOs (fresh and after FCs) are given in Table 1. The viscosity of fresh RVOs ranged between 44.16 and 60.25 mPa s, with the highest in RBO and the lowest in SBO. The fresh RBO contains a high proportion of long-chain SFAs (arachidic, behenic, and lignoceric acid) while fresh SBO contains high proportion of PUFAs (59.27%). The viscosities of RVOs were correlated with the proportion of long-chain FAs (Kaur et al., 2019). The previous earlier studies reported that oils rich in PUFAs had low viscosity due to their freely filled structure (Hassanien & Sharoba, 2014; Kaur et al., 2019; Kim, Kim, Lee, Yoo, & Lee, 2010). The PUFAs does not have a rigid and fixed structure, being loosely packed contribute greatly to the flow behavior of oils (Kim et al., 2010). The viscosity of RVOs increases with an increment in FCs. A slight change in viscosity of SBO (52.23 to 55.31 mPa s) and RBO (72.87 to 73.10 mPa s) was observed from the 1st to the 5th FC, while CSO (56.23 to 65.12 mPa s), SFO (53.41 to 58.24 mPa s), and CNO (60.94 to 64.25 mPa s) showed significant increase in viscosity. After the 10th FC, the higher increase in viscosity was observed in CSO and

TABLE 4 Effect of frying cycles on fatty acid composition of refined vegetable oils

RVOs	FC	C14:0 (Myristic)	C16:0 (Palmitic)	C18:0 (Stearic)	C20:0 (Arachidic)	C22:0 (Behnic)	C24:0 (Lignoceric)	C16:1 (Palmitoleic)	C18:1n9c (Oleic)	C20:1n11 (Gadoleic)	C18:2n6c (Linoleic)	C18:3n3c (Linolenic)	SFAs	MUFAs	PUFAs	UFAs	$\omega 6/\omega 3$	AI	TI
SBO	0	nd	10.88 ± 0.07^{b}	4.34 ± 0.08^{b}	0.44 ± 0.02^{b}	0.50 ± 0.01^{c}	0.18 ± 0.01^{a}	nd	22.93 ± 0.04^{b}	0.15 ± 0.04^{b}	53.58 ± 0.11^{d}	5.69 ± 0.08^{c}	16.34 ± 0.02^{c}	23.08 ± 0.07^{b}	59.27 ± 0.05^{e}	82.35 ± 0.02^{d}	9.42	0.13	0.27
	1	nd	11.10 ± 0.08^{b}	4.41 ± 0.05^{c}	0.45 ± 0.03^{b}	0.53 ± 0.02^{c}	0.21 ± 0.04^{b}	nd	20.44 ± 0.05^{b}	nd	53.16 ± 0.07^{d}	5.88 ± 0.07^{c}	16.70 ± 0.02^{c}	20.44 ± 0.05^{b}	59.04 ± 0.09^{e}	79.48 ± 0.03^{c}	9.04	0.14	0.28
	5	nd	11.60 ± 0.05^{b}	4.44 ± 0.04^{c}	0.45 ± 0.02^{b}	0.56 ± 0.02^{c}	0.23 ± 0.04^{b}	nd	20.31 ± 0.05^{b}	nd	52.39 ± 0.08^{d}	5.08 ± 0.07^{c}	17.28 ± 0.03^{c}	20.31 ± 0.08^{b}	57.47 ± 0.08^{e}	77.78 ± 0.02^{c}	10.31	0.15	0.31
	10	nd	11.64 ± 0.07^{b}	4.47 ± 0.61^{c}	0.47 ± 0.07^{b}	0.58 ± 0.04^{c}	0.27 ± 0.03^{b}	nd	20.12 ± 0.04^{b}	nd	52.11 ± 0.07^{d}	5.01 ± 0.06^{c}	17.43 ± 0.07^{c}	20.12 ± 0.03^{b}	57.12 ± 0.07^{e}	77.24 ± 0.04^{c}	10.40	0.15	0.31
RBO	0	0.35 ± 0.03^{c}	16.10 ± 0.04^{c}	2.52 ± 0.03^{a}	0.81 ± 0.01^{d}	0.43 ± 0.03^{b}	0.48 ± 0.06^{d}	0.18 ± 0.03^{c}	40.81 ± 0.06^{d}	0.44 ± 0.02^{c}	36.22 ± 0.07^{b}	0.49 ± 0.05^{b}	20.69 ± 0.05^{d}	41.43 ± 0.06^{d}	36.71 ± 0.03^{b}	78.14 ± 0.05^{c}	73.92	0.22	0.47
	1	0.39 ± 0.02^{c}	17.44 ± 0.05^{c}	2.25 ± 0.03^{a}	0.82 ± 0.09^{d}	0.46 ± 0.02^{b}	0.49 ± 0.05^{d}	0.14 ± 0.02^{b}	40.72 ± 0.07^{d}	0.38 ± 0.03^{c}	33.12 ± 0.05^{b}	0.23 ± 0.04^{b}	21.85 ± 0.09^{d}	41.24 ± 0.05^{d}	33.35 ± 0.02^{b}	74.59 ± 0.07^{b}	144.00	0.25	0.53
	5	0.41 ± 0.05^{c}	18.08 ± 0.06^{c}	2.07 ± 0.02^{a}	0.91 ± 0.05^{e}	0.49 ± 0.04^{b}	0.52 ± 0.04^{d}	nd	40.22 ± 0.06^{d}	0.37 ± 0.03^{c}	32.80 ± 0.05^{b}	0.17 ± 0.02^{a}	22.48 ± 0.08^{d}	40.59 ± 0.05^{d}	32.97 ± 0.03^{b}	73.56 ± 0.09^{a}	192.94	0.27	0.55
	10	0.43 ± 0.07^{c}	20.04 ± 0.06^{d}	2.02 ± 0.01^{a}	0.98 ± 0.04^{e}	0.53 ± 0.04^{c}	0.56 ± 0.05^{d}	nd	40.03 ± 0.08^{d}	0.32 ± 0.01^{c}	32.02 ± 0.06^{b}	nd	24.56 ± 0.02^{d}	40.35 ± 0.06^{d}	32.02 ± 0.03^{b}	72.37 ± 0.08^{a}	–	0.30	0.62
CNO	0	nd	4.82 ± 0.02^{a}	2.10 ± 0.02^{a}	0.63 ± 0.05^{c}	0.39 ± 0.01^{b}	0.17 ± 0.02^{a}	0.19 ± 0.05^{c}	59.70 ± 0.12^{e}	1.25 ± 0.06^{e}	23.17 ± 0.04^{a}	6.70 ± 0.09^{d}	8.11 ± 0.01^{a}	61.14 ± 0.04^{f}	29.87 ± 0.01^{a}	91.01 ± 0.01^{e}	3.46	0.05	0.11
	1	nd	4.96 ± 0.03^{a}	2.16 ± 0.03^{a}	0.69 ± 0.07^{c}	0.45 ± 0.01^{b}	0.19 ± 0.03^{a}	0.07 ± 0.03^{a}	57.67 ± 0.11^{e}	1.18 ± 0.03^{e}	21.56 ± 0.03^{a}	6.50 ± 0.09^{c}	8.45 ± 0.02^{a}	58.92 ± 0.01^{e}	28.06 ± 0.03^{a}	86.98 ± 0.02^{d}	3.32	0.06	0.12
	5	nd	5.06 ± 0.03^{a}	2.36 ± 0.02^{a}	0.79 ± 0.02^{d}	0.49 ± 0.04^{b}	0.24 ± 0.04^{b}	nd	57.49 ± 0.08^{e}	1.02 ± 0.05^{e}	20.04 ± 0.03^{a}	6.01 ± 0.08^{c}	8.94 ± 0.02^{a}	58.51 ± 0.02^{e}	26.05 ± 0.04^{a}	84.56 ± 0.02^{d}	3.33	0.06	0.13
	10	nd	5.68 ± 0.02^{a}	2.52 ± 0.04^{a}	0.83 ± 0.01^{d}	0.51 ± 0.03^{c}	0.27 ± 0.04^{b}	nd	57.15 ± 0.09^{e}	0.79 ± 0.04^{d}	19.81 ± 0.02^{a}	5.87 ± 0.07^{c}	9.81 ± 0.01^{a}	57.94 ± 0.04^{e}	25.68 ± 0.01^{a}	83.62 ± 0.03^{d}	3.37	0.07	0.15
CSO	0	0.72 ± 0.01^{d}	22.03 ± 0.06^{d}	3.06 ± 0.05^{b}	0.42 ± 0.04^{b}	0.25 ± 0.09^{a}	0.16 ± 0.01^{a}	nd	20.19 ± 0.02^{b}	0.16 ± 0.06^{b}	51.57 ± 0.09^{d}	nd	26.64 ± 0.05^{e}	20.35 ± 0.03^{b}	51.57 ± 0.08^{d}	71.92 ± 0.03^{a}	–	0.35	0.72
	1	0.91 ± 0.03^{e}	20.59 ± 0.07^{d}	3.12 ± 0.06^{b}	0.46 ± 0.07^{b}	0.27 ± 0.05^{a}	0.17 ± 0.04^{a}	nd	19.00 ± 0.03^{a}	0.17 ± 0.04^{b}	49.83 ± 0.06^{d}	nd	25.52 ± 0.04^{e}	19.17 ± 0.05^{a}	49.83 ± 0.07^{c}	69.00 ± 0.05^{a}	–	0.35	0.71
	5	0.97 ± 0.02^{e}	23.84 ± 0.09^{d}	3.44 ± 0.05^{b}	0.48 ± 0.06^{b}	0.31 ± 0.07^{a}	0.18 ± 0.05^{a}	nd	18.73 ± 0.01^{a}	0.17 ± 0.07^{b}	47.52 ± 0.05^{c}	nd	29.22 ± 0.06^{e}	18.90 ± 0.02^{a}	47.52 ± 0.06^{c}	66.42 ± 0.04^{a}	–	0.42	0.85
	10	0.98 ± 0.02^{e}	24.20 ± 0.11^{d}	3.64 ± 0.06^{b}	0.49 ± 0.04^{b}	0.95 ± 0.04^{d}	0.19 ± 0.04^{a}	nd	17.42 ± 0.02^{a}	0.19 ± 0.02^{b}	45.24 ± 0.04^{c}	nd	30.45 ± 0.05^{e}	17.61 ± 0.01^{a}	45.24 ± 0.05^{c}	62.85 ± 0.06^{a}	–	0.45	0.92
SFO	0	0.08 ± 0.06^{a}	6.01 ± 0.07^{a}	3.12 ± 0.06^{a}	0.27 ± 0.06^{a}	0.79 ± 0.03^{d}	0.35 ± 0.06^{c}	0.11 ± 0.04^{a}	37.15 ± 0.05^{c}	0.14 ± 0.01^{b}	51.23 ± 0.14^{c}	nd	10.62 ± 0.06^{b}	37.40 ± 0.04^{c}	51.23 ± 0.04^{d}	88.63 ± 0.03^{d}	–	0.07	0.21
	1	0.12 ± 0.05^{a}	6.25 ± 0.04^{a}	3.18 ± 0.07^{b}	0.27 ± 0.02^{a}	0.81 ± 0.04^{d}	0.35 ± 0.02^{c}	nd	37.09 ± 0.06^{c}	0.06 ± 0.03^{a}	50.77 ± 0.09^{c}	nd	10.98 ± 0.07^{b}	37.15 ± 0.07^{c}	50.77 ± 0.03^{d}	87.92 ± 0.01^{d}	–	0.08	0.22
	5	0.15 ± 0.06^{b}	6.38 ± 0.05^{a}	3.41 ± 0.05^{b}	0.29 ± 0.03^{a}	0.94 ± 0.07^{d}	0.44 ± 0.03^{c}	nd	36.87 ± 0.07^{c}	0.04 ± 0.05^{a}	49.26 ± 0.08^{c}	nd	11.61 ± 0.06^{b}	36.91 ± 0.06^{c}	49.26 ± 0.06^{c}	86.17 ± 0.02^{d}	–	0.08	0.23
	10	0.19 ± 0.04^{b}	6.76 ± 0.04^{a}	3.55 ± 0.07^{b}	0.31 ± 0.01^{a}	0.96 ± 0.08^{d}	0.47 ± 0.04^{c}	nd	36.20 ± 0.05^{c}	nd	48.46 ± 0.07^{c}	nd	12.24 ± 0.04^{b}	36.20 ± 0.04^{c}	48.46 ± 0.05^{c}	84.66 ± 0.01^{d}	–	0.09	0.25

Note: Values within the column with different alphabets are significantly different ($p \leq .05$).

Abbreviations: AI, atherogenic index; FC, frying cycle; MUFAs, monounsaturated fatty acids; PUFAs, polyunsaturated fatty acids; RVOs, refined vegetable oils; SFAs, saturated fatty acids; UFAs, unsaturated fatty acids; TI, thrombogenic index; $\omega 6/\omega 3$, linoleic/linolenic acid ratio; nd, not detected.

lower in RBO. Similar results of an increase in viscosity of two different vegetable oil blends during the intermittent frying of French fries for two consecutive days was reported in the previous study (Ramadan et al., 2006). With an increment in FCs, the level of SFAs increased while PUFAs decreased in RVOs as shown in Table 4. Viscosity is correlated with changes in the level of UFAs in oils during the deep- frying process (Kim et al., 2010). The changes in viscosity of oil during frying has been correlated with the formation of undesirable compounds in oxidation and polymerization reactions (Hassanien & Sharoba, 2014). Santos, Santos, and Souza (2005) also reported a rapid increase in viscosity of edible oils with an increment in frying time due to an increase in SFAs and the formation of degradation products. The *F* values of viscosity showed higher significant variation ($p \leq .005$) with FCs than the RVOs (Table 2). The viscosity of RVOs showed a highly significant positive correlation with FCs ($r = 0.535$, $p \leq .005$) and negative correlation with PUFAs ($r = -0.434$, $p \leq .005$) (Table 3).

3.6 | Changes in oil uptake with FCs

The changes in oil uptake by the chickpea splits after the 1st, 5th, and 10th FCs are given in Table 1. Oil uptake was significantly increased with an increment in FCs. The oil uptake after the 1st and the 5th FCs ranged from 13.5% to 37.5% and from 19.4% to 45.3%, respectively, with the lowest in SBO and highest in CNO. After the 10th FC, the highest oil uptake was observed in CNO (65.9%) and lowest in SBO (23.5%) compared to other RVOs. It is generally recognized that an increase in the viscosity of frying oils increases the oil uptake in fried food products. The high viscosity allows more accumulation of oils on the surface of fried foods, which penetrate inside the fried food products during the cooling process (Dana & Saguy, 2006; Kim et al., 2010). Kim et al. (2010) correlated viscosity and FAC of vegetable oils with oil uptake in fried products. The *F* values of oil uptake showed higher significant variation ($p \leq .005$) with FCs than the RVOs (Table 2). Oil uptake showed highly significant positive correlation with FCs and viscosity ($r = 0.702$, $r = 0.716$, respectively, $p \leq .005$) while highly significant negative correlation with PUFAs ($r = -0.484$, $p \leq .005$) as shown in Table 3.

3.7 | Changes in FAC of RVOs with FCs

The FAC of the fresh RVOs and after FCs are given in Table 4. The seven FAs detected in fresh RVOs were palmitic (4.82%–22.03%), stearic (2.10%–4.34%), arachidic (0.27%–0.81%), behenic (0.25%–0.79%), lignoceric (0.16%–0.48%), oleic (20.19%–59.70%), and linoleic (23.17%–53.58%) acids. The other FAs detected in fresh RVOs include myristic (0.08%–0.72% in RBO, CSO, and SFO), palmitoleic (0.11%–0.19% in SFO, RBO, and CNO) and linolenic (0.49%–6.70% in RBO, SBO, and CNO) acids. The main differences in FAC of RVOs were associated with a difference in the level of linoleic and linolenic acids (Kaur et al., 2019). The level of linoleic acid in CNO, RBO, SFO, CSO, and SBO was 23.17%, 36.22%, 51.23%, 51.57%, and 53.58%, respectively. The linolenic acid was not detected in CSO and SFO while in RBO, SBO, and CNO its level was 0.49%, 5.69%, 6.70%, respectively. Our results concur with the FAC of RVOs reported in previous studies (Dorni, Sharma, Saikia, & Longvah, 2018; Kaur et al., 2019). The sum of SFAs, MUFAs, and PUFAs were high in CSO (26.64%), CNO (61.14%), and SBO (59.27%) while lower in CNO (8.11%), CSO (20.35%), and CNO (29.87%), respectively. The linoleic (ω-6) and linolenic (ω-3) acids are the essential FAs that must be supplied in the human diet (Kaur et al., 2019; Saini & Keum, 2018). PUFAs was high in fresh SBO (59.27%) with a significant amount of ω-6 and ω-3 FAs ($\omega 6$:$\omega 3$ ratio of 9.42:1) compared to other RVOs. The RVOs with high PUFAs content and a significant amount of $\omega 6$ and $\omega 3$ FAs (with acceptable limits of $\omega 6$:$\omega 3$ ratio of 5–10:1) play important role in human health (Kaur et al., 2019; Saini & Keum, 2018).

Changes in the level of FAs of RVOs were observed with increment in FCs. The level of MUFAs and PUFAs were slightly decreased while SFAs were slightly increased in RVOs with an increment in FCs. After the 1st and the 5th FCs, a minor increase in SFAs and decrease in UFAs were observed. However, after the 10th FC significant increase in SFAs and decrease in UFAs were observed in all RVOs. Our results are in close agreement with those of Ramadan et al. (2006) who reported a decrease in the level of linoleic acid and an increase in the level of palmitic and stearic acid across two consecutive days of frying in two vegetable oil blends. The changes in the level of FAs might be due to the degradation of UFAs in the RVOs during deep frying. The FAs with double (π) bonds are more labile to oxidative and thermal degradation during repeated FCs (Debnath et al., 2012; Hassanien & Sharoba, 2014). As shown in Table 4, there was a marginal change in FAC of SBO after the 1st, 5th, and 10th FCs compared to other RVOs. After the 10th FC, CSO showed higher variation in the level of SFAs, MUFAs, and PUFAs, while SFO, CNO, and RBO showed slight variation. Hassanien and Sharoba (2014) also observed higher variation in FAC of CSO (decrease in linoleic and increase in palmitic and stearic acids levels) compared to SFO and palm olein oil during deep frying of French fries for two consecutive days. Similar results of an increase in SFAs and a decrease in PUFAs and MUFAs in oils with an increase in successive frying sessions or FCs have been reported in previous studies (Debnath et al., 2012; Multari et al., 2019; Zribi et al., 2014). Myristic acid (short-chain SFA) was slightly increased in CSO, RBO, and SFO with an increase in FCs. Siri-Tarino, Sun, Hu, and Krauss (2010) reported that short-chain SFAs can increase the risk of cardiovascular problems due to their LDL cholesterol-raising effect. The *F* values of SFAs, MUFAs, and PUFAs showed higher significant variation ($p \leq .005$) in RVOs than the FCs (Table 2). The UFAs showed a highly significant negative correlation with FCs, FFA, p-AV, viscosity, and SFAs ($r = -0.366, -0.785, -0.486, -0.375, -0.878$, respectively, $p \leq .005$) (Table 3).

3.8 | Changes in AI and TI of RVOs with FCs

The AI and TI of the fresh RVOs and after FCs are given in Table 4. AI and TI are the two dietary indices proposed to know the effects of SFAs, MUFAs, and PUFAs in the development of coronary heart diseases (Mohanty et al., 2012; Pereira et al., 2019). Some FAs have a greater role in atherosclerosis, while others have a greater role in thrombogenesis. Myristic, lauric, and palmitic acids have a cholesterol-raising effect and are thus considered as atherogenic SFAs. Myristic, stearic, and palmitic acids influence clot formation in blood vessels and are known as thromogenic SFAs (Mohanty et al., 2012). MUFAs and PUFAs have a role in minimizing the atherogenicity and thrombogenicity of edible oils. AI and TI ranged from 0.05 to 0.35 and 0.11 to 0.72, respectively, in fresh RVOs. The highest values for both indices were observed for CSO and lowest for CNO. The changes in indices varied among RVOs due to differences in their SFAs contents. No significant change in AI and TI was observed after 1st FC in RVOs. However, a slight increase in these indices was observed in CSO and RBO after the 5th FC. An increase in AI (0.45) and TI (0.92) was observed for CSO after the 10th FC, while both indices of SBO after the 5th and 10th FCs remained unchanged. CSO contains high myristic acid content than the other RVOs. The myristic acid is considered as four times more atherogenic than palmitic acid. PUFAs had a more prominent role in minimizing atherosclerosis and thrombogenesis than MUFAs (Ulbricht & Southgate, 1991). The *F* values of AI and TI showed higher significant variation ($p \leq .005$) in RVOs than the FCs (Table 2). AI and TI showed a highly positive significant correlation with FFA content ($r = 0.807$ and 0.812, respectively, $p \leq .005$) and highly significant negative correlation with PUFAs ($r = -0.606$, -0.633, respectively, $p \leq .005$) as shown in Table 3.

3.9 | Changes in FTIR spectra of RVOs with FCs

The FTIR spectral information of SBO as a descriptive example is shown in Figure 2a and of RVOs (fresh and after the 1st, 5th, and 10th FCs) are given in Figure 2b–f. The visible peaks at 3,009, 2,955, 2,925, 2,854, 1,745, 1,650, 1,417, 1,377, 1,238, 1,157, and 725 cm^{-1} were observed in FTIR spectral studies of RVOs (Kaur et al., 2019). The spectral information observed in 3,100–2,800 cm^{-1} and 1,800–700 cm^{-1} regions confirms specific functional groups in RVOs (Kaur et al., 2019). The small peak detected at 3,009 cm^{-1} (attributed to C–H stretching symmetric vibration of cis-olefinic double-bonds attributed to UFAs), small shoulder at 2,955 cm^{-1}, sharp peak detected at 2,925 cm^{-1} (represents stretching vibration of C–H of aliphatic CH_3 and CH_2 groups), and shoulder peak detected at 2,854 cm^{-1} (associated with symmetric stretching vibration of C–H of aliphatic CH_2 groups) representing the region of hydrogen bond stretching of the functional groups of triglycerides in RVOs as described previously (Kaur et al., 2019). The sharp peak detected at 1,745 cm^{-1} (represents stretching vibration of C–O ester groups), very weak peak detected at 1,650 cm^{-1} (attributed to C=C stretching vibration of *cis*-disubstituted olefins), small peak detected at 1,417 cm^{-1} (represents rocking vibrations of C–H bonds of *cis*-disubstituted olefins), a very small peak detected at 1,377 cm^{-1} (assigned to bending symmetric vibration of C–H bonds of CH_2 groups), shoulder peak at 1,238 and strong peak at 1,157 cm^{-1} (stretching vibration of ester carbonyl groups), and medium peak at 725 cm^{-1} (attributed to out-of-plane vibration of *cis* –HC=CH– group of disubstituted olefins and overlapping of CH_2 rocking vibration) were observed in RVOs as described previously (Kaur et al., 2019; Suri et al., 2019).

FTIR spectra were used to evaluate the quality of RVOs after FCs. Visual observation of FTIR spectra does not show any difference in peaks of fresh RVOs and of those collected after the 1st, 5th, and 10th FCs. The overall signal patterns looked similar to each other. However, upon closer observation, the intensities of some peaks varied slightly indicating changes in FAC and quality characteristics of RVOs with increment in FCs. The small peak at 3,009 cm^{-1} in RVOs showed a decline in intensity with an increment in FCs. The peak intensity at 3,009 cm^{-1} decreased more in CNO and CSO as compared to other RVOs after the 10th FC. This might be due to higher degradation of *cis*-olefinic C–H double bonds in UFAs of CNO and CSO with increment in FCs. The level of UFAs showed a significant decline in CNO (91.01% to 83.92%) and CSO (71.92% to 62.85%) after the 10th FC when compared with fresh oils (Table 4). The sharp peak at 2,925 cm^{-1} and shoulder peaks at 2,955 and 2,854 cm^{-1} showed an increase in intensities after the 5th and 10th FCs compared to 1st FC (Figure 2b–f). The higher increase in intensities of these peaks was observed in CSO, RBO and SFO compared to CNO and SBO after the 5th and 10th FCs. This might be related to an increase in the level of SFAs in CSO, RBO, and SFO with increment in FCs (Table 4). The peak at 3,009 cm^{-1} is associated with the level of UFAs while peaks at 2,925, 2,955, and 2,854 cm^{-1} are linked with the level of SFAs in RVOs. The intensities of these peaks showed variation due to the difference in the degree of unsaturation in the FAC of RVOs (Kaur et al., 2019). The level of SFAs increased while PUFAs and MUFAs were decreased more in RBO, CSO, and SFO after the 10th FC (as shown in Table 4) compared to CNO and SBO. Thus, the differences in peaks intensities at 3,009, 2,925, 2,955, and 2,854 cm^{-1} reflect the changes in FAC of RVOs after FCs. The previous studies had also correlated the levels of SFAs and UFAs with peaks at 3,009, 2,925, and 2,854 cm^{-1} of edible oils (Kaur et al., 2019; Sim & Ting, 2012; Suri et al., 2019).

The peaks intensities at 1,157 and 1,745 cm^{-1} of RVOs increased with increment in FCs. The higher increase in peak intensities at 1,157 and 1,745 cm^{-1} were in RBO, SFO, and CSO while in CNO and SBO minor increase was noticed with an increment in FCs (Figure 2b–f). The changes in intensities of peaks at 1,157 and 1,745 cm^{-1} could be associated with the hydrolysis of carbonyl ester functional groups into ketones and the formation of secondary oxidation products from peroxides and hydroperoxides during the deep-frying process. Srivastava and Semwal (2015) correlated the

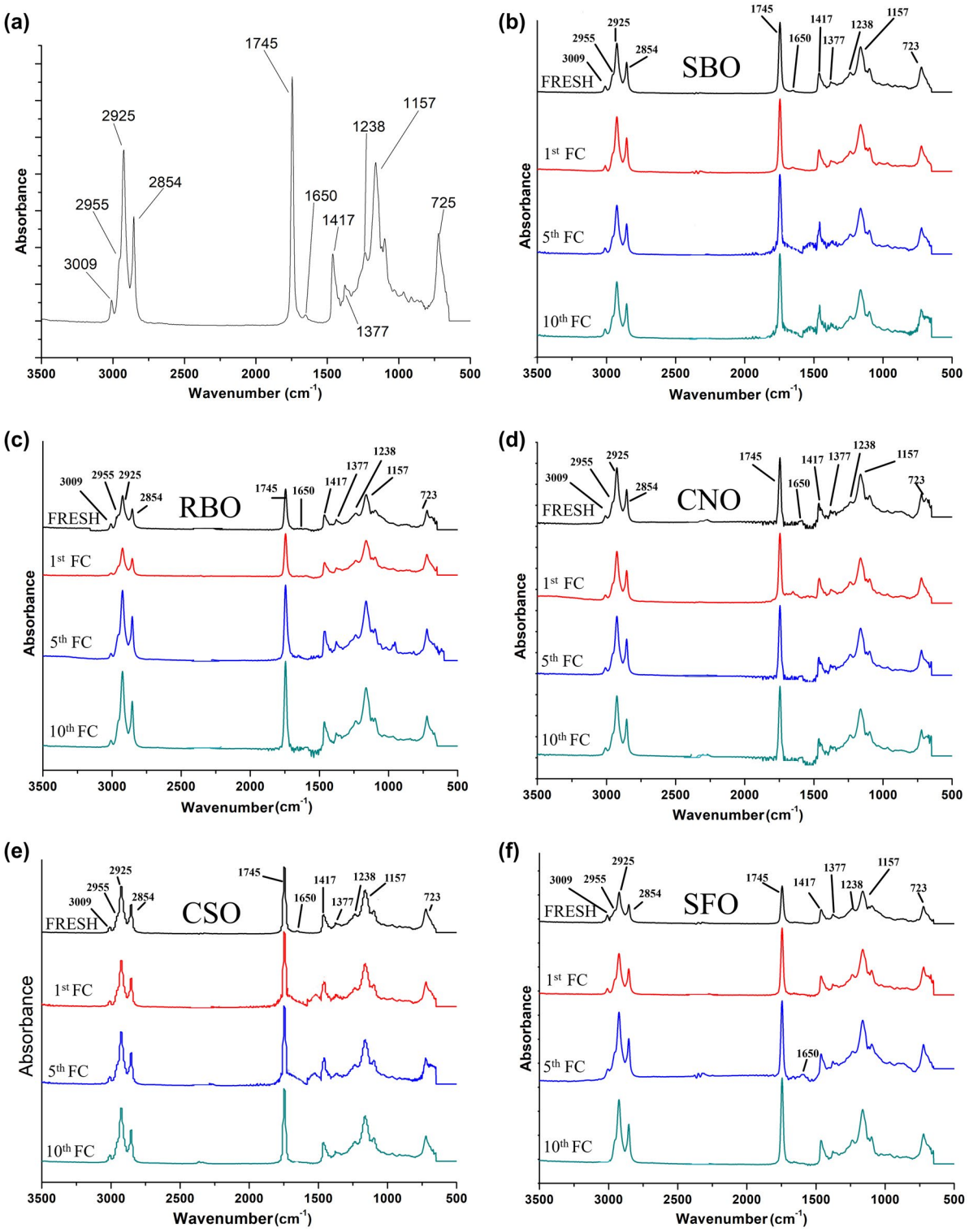

FIGURE 2 (a). A descriptive FTIR spectrum of SBO. (b). FTIR spectra of SBO (fresh and after the 1st, 5th, and 10th FCs). (c). FTIR spectra of RBO (fresh and after the 1st, 5th, and 10th FCs). (d). FTIR spectra of CNO (fresh and after the 1st, 5th, and 10th FCs). (e). FTIR spectra of CSO (fresh and after the 1st, 5th, and 10th FCs). (f). FTIR spectra of SFO (fresh and after the 1st, 5th, and 10th FCs)

change in peak intensity at 1,745 cm^{-1} with the degradation of hydroperoxides in virgin coconut oil during the prolonged deep-frying process. After the 5th and 10th FCs, the intensity of a small peak at 1,417 cm^{-1} showed a slight decline in CNO, CSO, RBO, and SFO while minor variation was observed in SBO. The intensities of the peaks at 1,650, 1,377, 1,238, and 725 cm^{-1} showed minor variation with increment in FCs when compared with fresh RVOs. After the 5th and 10th FCs, all these peaks were observed at designated wavenumbers. The differences in peak intensities observed at certain wavenumbers in the FTIR spectra correlate with the changes in FAC

and formation of oxidation products in RVOs with increment in FCs. Earlier studies had also related changes in peak intensities of FTIR spectra with oxidative changes in edible oils during the deep-frying process (Srivastava & Semwal, 2015; Talpur et al., 2014).

4 | PRINCIPAL COMPONENT ANALYSIS (PCA)

Multivariate analysis was carried out to correlate the results of chemical properties, viscosity, FAC, and FTIR spectra of fresh RVOs and after FCs. The PCA was performed to evaluate the effect of FCs on the quality characteristics of RVOs. The eigenvalues greater than one was considered to evaluate the relative contribution of the principal components (PCs) in the overall total data variability. The first five PCs were significant and accounted for 93% variability in the data set (Table 5). The first principal component (PC1) accounted for 36% of the total variability. The variables that correlated with the PC1 were FFA (−0.335), viscosity (0.283), SFAs (0.343), UFAs (−0.355), AI (0.342), and TI (0.347). While, PV (0.370), p-AV (0.381), OSI (−0.320), and oil uptake (0.353) mainly contributed to the PC2, with a total variability contribution of 29%. The PC3 accounted for 15% of the total variability and was contributed by MUFAs (0.402), PUFAs (−0.530), and $\omega 6/\omega 3$ (0.431). The PC4 and PC5 accounted for 8% and 5% of the total variability, respectively.

TABLE 5 Principal components for illustrating the interpretation in Figure 2

Variable	PC1	PC2	PC3	PC4	PC5
FFA	0.335	0.019	0.227	0.001	−0.020
PV	0.071	0.370	−0.205	−0.141	−0.066
p-AV	−0.062	0.381	−0.011	−0.084	0.306
OSI	−0.102	−0.320	0.198	−0.240	−0.085
Viscosity	0.283	0.200	0.270	0.012	−0.058
SFAs	0.343	−0.154	0.037	0.176	−0.009
MUFAs	−0.215	0.210	0.402	−0.096	0.167
PUFAs	0.036	−0.218	−0.530	−0.034	−0.220
UFAs	−0.355	0.075	−0.020	−0.232	−0.008
$\omega 6/\omega 3$	0.086	−0.035	0.431	−0.146	−0.603
AI	0.342	−0.138	0.075	0.191	0.126
TI	0.347	−0.142	0.059	0.138	0.137
OU	0.128	0.353	0.105	0.132	0.009
1,157	−0.162	0.264	0.037	0.181	−0.549
1,417	−0.255	−0.078	0.310	0.212	0.306
1,650	−0.258	−0.111	0.014	0.434	−0.109
1,745	−0.038	0.257	−0.097	0.546	−0.041
3,009	−0.205	−0.148	0.050	0.401	−0.062

Note: PC1, PC2, PC3, PC4 and PC5 are the first five principal components.

From the loading plot (Figure 3a), it is evident that AI and TI had a close relation with SFAs. OSI had opposite relation with FCs while oil uptake and PV were closely related to FCs. FTIR peaks at 3,009, 1,417, and 1,650 cm^{-1} showed a positive relation with UFAs and negative relation with viscosity. While the FTIR peak at 1,745 cm^{-1} represents the secondary oxidation products like carbonyl compounds showed close relation with p-AV and opposite relation with PUFAs. Figure 3b shows the sore plot between the first two PCs, where three distinct groups based upon FCs and RVOs were observed. With the increment in FCs, the OSI of the RVOs decreased, while PV, p-AV, and oil uptake increased. This was evident from the alignment of the colored ligands. Further, after the 5th FC, a rapid increment in oil uptake and deterioration of OSI occurred, which was highest in CNO followed by SFO and the lowest in SBO and CSO. Moreover, CSO grouped with RBO (group III) was marked with high SFAs, FFA, viscosity, AI, and TI indicating that these oils not only possessed poor FA profile (unacceptable $\omega 6/\omega$ three ratios) but also exhibited high deterioration with increment in FCs. Further, in comparison to RVOs in group III, group I & II had high UFAs and low AI and TI. SBO had the most favorable $\omega 6/\omega 3$ ratio and exhibited higher OSI, lower FFA, PV, and p-AV than SFO and CNO. Thus, SBO was found ideal RVO for deep frying.

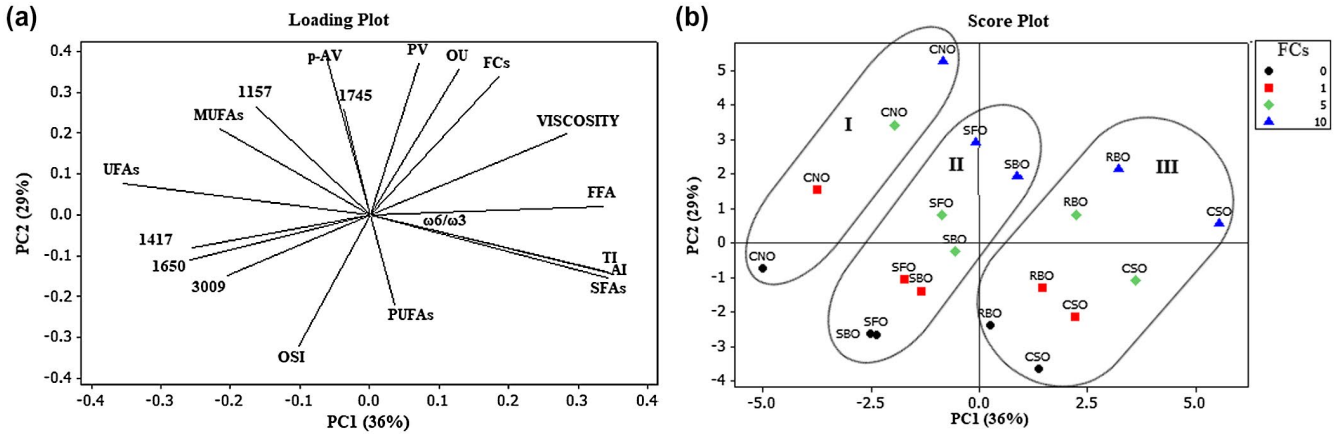

FIGURE 3 (a) Principal component analysis (PCA) loading plot describing relationship among different properties of fresh RVOs and after FCs. (b) PCA and score plot describing relationship among different properties of fresh RVOs and after FCs

5 | CONCLUSIONS

Changes in chemical properties, oxidative stability, and FAC of RVOs were observed with increment in FCs. RVOs behaved differently with increment in FCs for various quality and stability parameters. RBO and CSO showed a higher change in FFA content, PV, p-AV, viscosity, SFAs, AI, and TI than other RVOs with increment in FCs. Oil uptake was observed highest in CNO and lowest in SBO. The minimum changes in FFA content, viscosity, and OSI were observed in SBO and SFO with increment in FCs. The change in FTIR peak intensities at 3,009, 2,925, 2,854, and 1,745 cm^{-1} was minimum for SBO indicating its stability during short-term deep-frying cycles. SBO contains desirable FAC with admissible limits of $\omega 6$:$\omega 3$ ratio for health benefits. It exhibited high OSI and the lowest increase in viscosity with increment in FCs. Thus, SBO was found to be ideal for short-term deep frying as compared to other RVOs.

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CONFLICT OF INTEREST

The authors have declared no conflicts of interest for this article.

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