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Development of spicy flavored virgin coconut oil by incorporating a mixture of spices oleoresins

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Abstract – Lipid oxidation is a major cause of deterioration in the quality of cooking oils. Degradation of unsaturated fatty acids in oils directly leads to changes in nutritional value, flavor, and storage properties. Oleoresins of ginger, garlic, nutmeg, pepper, cloves, and cinnamon were extracted and incorporated into virgin coconut oil (VCO) to overcome adverse effects of lipid oxidation and changes occurrence on physicochemical properties, thermal stability, shelf life, antioxidant activity, total phenolics and sensory evaluation were conducted against same properties of VCO. Lipid oxidation was assessed in terms of free fatty acid (FFA) value and peroxide value (PV). For the comparison purpose, another oil sample was prepared by incorporating vitamin E too. Results revealed that both PV and FFA of VCO and spicy flavored oil (SFO) samples after one-week storage period were 1.93 ± 0.01 and 1.62 ± 0.02 mEq/kg and 0.36 ± 0.01 and 0.24 ± 0.01 (%) respectively. Saponification value (SV), iodine value (IV), smoke point, flashpoint, viscosity, and the specific gravity of SFO were increased and there was no significant difference in moisture content and insoluble impurities in SFO and VCO. The highest phenolic content and DPPH free radical scavenging activity were found in SFO. The thermal stability of SFO was better than VCO. Both oleoresins and vitamin E incorporated samples showed the same pattern of increment of FFA and PV during storage; however, those increments were slower than VCO. SFO was highly accepted by the sensory panelists in terms of color, aroma, taste, texture, and overall acceptability.

Keywords: virgin coconut oil / spices / free fatty acid / peroxide value / phenolic content

Résumé – Développement d'une huile de coco vierge aromatisée et épicée en incorporant un mélange d'oléorésines d'épices. L'oxydation des lipides est une cause majeure de détérioration de la qualité des huiles de cuisson. La dégradation des acides gras insaturés de ces huiles entraîne une modification des propriétés nutritionnelles, organoleptiques et de stockage. Des oléorésines de gingembre, d'ail, de noix de muscade, de poivre, de clous de girofle et de cannelle ont été extraites et incorporées dans de l'huile de noix de coco vierge (VCO) pour contrer les effets délétères de l'oxydation des lipides ; l'évaluation sensorielle ainsi que les changements intervenus au niveau des propriétés physico-chimiques, de la stabilité thermique, de la durée de conservation, de l'activité anti-oxydante et des composés phénoliques totaux ont été mesurés par rapport aux mêmes propriétés de l'huile vierge (VCO). L'oxydation des lipides a été évaluée en termes de teneur en acides gras libres (FFA) et d'indice de peroxydes (PV). À des fins de la comparaison, un autre échantillon d'huile a été préparé en y incorporant également de la vitamine E. Les résultats ont révélé qu'après une semaine de stockage, les valeurs de PV et FFA des échantillons de VCO et d'huile aromatisée épicée (SFO) étaient respectivement de 1,93 ± 0,01 et 1,62 ± 0,02 mEq/kg et de 0,36 ± 0,01 et 0,24 ± 0,01 (%). L'indice de saponification (SV), l'indice d'iode (IV), le point de fumée, le point d'éclair, la viscosité et la densité relative de la SFO ont augmenté et il n'y a pas eu de différence significative entre SFO et VCO au niveau des teneurs en humidité et en impuretés insolubles. Les valeurs les plus élevées de teneur en composés phénoliques totaux et d'activité antiradicalaire (DPPH) ont été constatées pour la SFO. La stabilité thermique de la SFO s'est avérée supérieure à celle de la VCO. Les échantillons incorporant des oléorésines et de la vitamine E ont montré le même comportement avec l'augmentation des teneurs en FFA et PV pendant le stockage ; cependant, ces augmentations étaient plus

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lentes que celles observées pour la VCO. L'huile épicée SFO a été très bien accepté par les membres du panel sensoriel en termes de couleur, d'arôme, de goût, de texture et d'acceptabilité générale.

Mots clés : huile de noix de coco vierge / épices / acides gras libres / indice de peroxyde / teneur en composés phénoliques

1 Introduction

Coconut oil is one of the most extensively used cooking oil in Asian countries as well as a rich source of dietary fat (Krishna *et al.*, 2010). Commercially, coconut oil can be obtained from copra and based on the extraction method of oil from coconut meat, it can be categorized into two groups: refined, bleached, and deodorized coconut oil (RBD) and virgin coconut oil (VCO) (Narayanankutty *et al.*, 2018). RBD oil process involves high temperature treatments, between 204 °C and 245 °C, which degrades the essential amino acids, tocopherols and other beneficial compounds present in coconut oil (Mulyadi *et al.*, 2018). VCO is extracted naturally from cold extraction, hot extraction, wet extraction, fermentation, and enzymatic extraction techniques without using high temperatures or chemical treatments (Agarwal and Bosco, 2017).

The fatty acid composition is found to be similar in RBD coconut oil and VCO (Narayanankutty *et al.*, 2018). Coconut oil contains a high level of low molecular weight saturated fatty acids. Among these, lauric acid (C12:0) is the major fatty acid (45–52%), others are myristic acid (15–19%) and palmitic acid (10–11%) (Dayrit, 2014). VCO has become more popular because of its beneficial effects than RBD oil. Several studies have investigated the medicinal properties of VCO including anti-inflammatory, analgesic, antipyretic, antioxidant, anti-stress, and antimicrobial properties. Nevin and Rajamohan (2004) reported that VCO reduces triglycerides, phospholipids, low-density lipoprotein (LDL) cholesterol and increases high-density lipoprotein (HDL) cholesterol compared to the RBD oil. Furthermore, VCO is capable of reducing lipid peroxidation as it contains high amounts of antioxidants (Marina *et al.*, 2009). Rancidification is one of the major deterioration processes involve in cooking oils. As a result, organoleptic properties and keeping quality of cooking oils and oil-related products are altered. Synthetic antioxidants are effective in improving the stability; recently found that it can be carcinogenic and some are banned in many countries (Madhujith and Sivakanthan, 2018). Therefore, natural antioxidants from plant sources have attracted a lot of attention in controlling lipid oxidation.

Apart from that, Sri Lankan and Mediterranean gastronomies also used cooking oils along with different herbs and spices as a traditional practice to enhance the taste and aroma (Caporaso *et al.*, 2013). Due to the presence of natural antioxidants and an attractive flavour profile, it was able to make products with better stability and shelf life. Under these circumstances, this study was designed to develop spicy flavored virgin coconut oil using oleoresins of spice mixture and analyzing physicochemical properties, thermal stability, shelf life, total phenolics, antioxidant activity and sensory evaluation of this oil against the same of VCO (without incorporation of oleoresins).

2 Materials and methods

A commercial VCO supplied by the Marina Oil Company in Sri Lanka (SL) was used for the experiments. Oleoresins of ginger, garlic, nutmeg, cloves, pepper, and cinnamon were purchased from a reputed internationally registered supplier in SL (Lakessence, Homagama, Sri Lanka).

2.1 Preparation of spicy flavored oil

An oleoresins mixture was prepared by adding ginger: garlic:cinnamon:nutmeg:cloves:black pepper into 1:1:1:1:1:1 ratio (0.001 g each) and the mixture was added into 12 g of VCO oil samples. The mixture was blended thoroughly, and it was kept in dark brown colored bottles and stored under ambient condition (27 °C) for the subsequent use of the study.

2.2 Determination of physicochemical properties of oils

Chemical properties such as PV (AOCS Cd 8b-90), FFA (AOCS Cd 3d-63), IV (AOCS Cd 1b-87), SV (AOCS Cd 3-25) and physical properties such as moisture (AOAC 925.10), insoluble impurities percentage (IUPAC 2.604), smoke point (AOCS Cc 9a-48), flash point (AOCS Cc 9a-48), specific gravity (AOAC 920.212) and viscosity (AOAC 22.00) were measured according to the protocols mentioned herewith.

2.3 Determination of thermal stability of spicy flavored oil

Ten grams of oil was taken and it was heated. The stability of oils during frying was analyzed at 170° for 2 hours by taking oil samples for every 30 minutes and collected samples were subjected to analyze PV and FFA value.

2.4 Storage stability of spicy flavored oil

Storage stability of oils was assessed by resorting rapid aging test. Therein, SFO, as well as VCO samples, were taken, and each oil was filled into 36 glass bottles and stored them in a hot air oven at 60° for twelve weeks. Positive control of VCO in 36 bottles with vitamin E (without oleoresins) was also kept under the same condition for comparison purposes. Samples were withdrawn at seven days of intervals for twelve weeks and storage stability was analyzed in terms of PV and FFA level.

2.5 Total phenolic content

The total phenolic content was analyzed according to the method of Redondo-Cuevas *et al.* (2019) with slight

modifications. Therein, 100 μL aliquot of each sample was transferred to a test tube and it was mixed with 5.80 mL of distilled water, 500 μL Folin–Ciocalteu reagent, and 1500 μL of sodium carbonate. The mixture thereafter was vortexed for 30 seconds and incubated at 40° for 30 minutes. The absorbance of the mixture at 760 nm wavelength was measured using Uv-Vis spectrophotometer. Gallic acid was used as the standard to generate a calibration curve. Total phenolic content was expressed as a gallic acid equivalent using a linear equation based on the calibration curve.

2.6 Determination of DPPH radical scavenging activity

Antioxidant activity of the oil was analyzed using, the method described by Pradhananga and Manandhar (2018) with slight modifications. Aliquots of 500 μL taken from each concentration were mixed with 2500 μL of DPPH working solution in a screw-capped 4 mL micro-test tubes covered with Al foils. Thereafter, the mixtures were vortexed for 30 seconds and left to react for 30 minutes in the dark. Finally, the absorbance of oil was measured at 517 nm wavelength using a spectrophotometer.

2.7 Sensory evaluation

The acceptance of the developed SFO was evaluated for its sensory characteristics. The SFO samples were prepared one week before to the sensory evaluation and were stored in sealed glass bottles for consistent mixing and stabilization of the samples. Potato chips were fried with SFO, VCO and RBD coconut oil for the evaluation of taste and texture of products after frying with those oils. Sensory evaluation was carried out by 30 semi-trained panelists, who were nonsmokers and with an age range of 25–35. The panelists were provided with oil samples in numbered closed vessels and potato chips fried with SFO, VCO and RBD oil. The product was evaluated for its sensory characteristics for color, aroma, taste, texture and overall acceptability using a 5-point hedonic scale (like extremely-5, like moderately-4, neither like nor dislike-3, dislike moderately-2, dislike extremely-1). The mean sensory scores for various attributes of the SFO were calculated.

2.8 GC-MS analysis for spicy flavored cooking oil

Sodium methyl ester of oil samples was prepared according to AOAC 969.33 (preparation of methyl esters by sodium methoxide method) (Nielsen 2017), and transfer into the GC valves. For the GC analysis HP − 5 ms nonpolar column was used and the temperature was programmed from 80 °C to 200 °C with the increment rate of temperature 5 °C/min during analysis. Helium was used as the carrier gas and internal pressure was maintained at 100 kPa. The injector temperature was 250 °C.

2.9 SPME – GCMS analysis for volatile compounds

Two grams of the SFO sample was placed into 20 mL GC vial, tightly capped with polytetrafluorethylene (PTFE) septum, and left for 3 hours at 30° to allow for the equilibration of the volatiles in the headspace. After equilibration, the septum covering each vial was pierced with SPME needle, and the fiber was exposed to the headspace for 40 minutes. The volatiles adsorbed by the fiber were thermally desorbed in the hot injection port of a gas chromatograph. Helium was used as the carrier gas, at a flow rate of 0.9 mL/min and the oven temperature was maintained at 40 °C for 10 minutes and the final temperature of 200 °C at 3 °C/min. Identification of isolated volatile compounds was investigated by comparing obtained mass spectra of unknown peaks with standard reference library ICUSJ (Instrument center, University of Sri Jayewardenepura, Sri Lanka).

2.10 Statistical analysis

Statistical analysis was performed with Minitab statistical package (version 17). All data reported are the means ± SE of three repetitions. Significance of differences between samples was calculated by the ANOVA procedure, using a significance level of $P \leq 0.05$. The means were separated using Fisher's Least Significance Difference (LSD) with a 95% confidence level.

3 Results and discussion

3.1 Physiochemical properties of oil

SFO and VCO samples were analyzed in relation to the following physicochemical properties and results are given in Table 1.

According to the results given in Table 1, the changes in physicochemical properties that occurred in two oil samples after one week of storage could be explained. PV and FFA of SFO were reported as 1.62 ± 0.02 and 0.24 ± 0.01% respectively and values for the same parameters of VCO were 1.93 ± 0.01 and 0.36 ± 0.01%, respectively. The statistical results revealed that *P*-value of both samples was less than 0.05 ($P < 0.05$). Because of the natural antioxidants, which present in spices oleoresins PV and FFA level of SFO was significantly decreased compared to the VCO. Thus, these natural extracts may prevent lipid oxidation in cooking oils (Taghvaei and Jafari, 2015). Several studies have been investigated to analyze PV and FFA levels of cooking oils after the incorporation of different plant extracts. Shahid *et al.* (2018) studied the changes in PV and FFA of palm oil after the inclusion of cinnamon extracts. According to their results, PV and FFA levels were significantly decreased in treat oil with cinnamon extract. Pradhananga and Manandhar (2018) reported that PV and FFA levels of sunflower oil were decreased after the incorporation of spices oleoresins.

Saponification value (SV) of VCO and SFO were reported as 258 ± 1 and 262 ± 1, respectively (Tab. 1). SV indicates the average fatty acid chain length; long-chain fatty acids have a low SV because they have a relatively fewer number of carboxylic functional groups per unit mass (Guillaume *et al.*, 2018). Statistical results revealed that p value of both samples was less than 0.05 ($P < 0.05$). Therefore, SFO has significantly high SV compared to VCO. Similar results were found in the study of spices with palm oil used for culinary practices in Nigeria (Osu and Ogoko, 2019).

Table 1. Physicochemical properties of VCO and SFO samples after one-week of storage.

Parameter			VCO	SFO		
Peroxide value (mEq/kg)			1.93 ± 0.01	1.62 ± 0.02		
FFA value (%) as lauric acid			0.36 ± 0.01	0.24 ± 0.01		
Saponification value			258 ± 1	262 ± 1		
Iodine value			8.08 ± 0.04	8.26 ± 0.06		
Moisture %			0.1639 ± 0.05	0.1702 ± 0.01		
Insoluble impurities %			0.05 ± 0.01	0.05 ± 0.01		
Smoke point (°)			173 °C ± 2	179 °C ± 3		
Flashpoint (°)			186 °C ± 1	192 °C ± 2		
Viscosity (mPa.s)			47.965 ± 0.1	50.812 ± 0.1		
Specific gravity			0.9121 ± 0.001	0.9711 ± 0.002		
Color	L*	a*	b*	L*	a*	b*
	65 ± 2	4 ± 1	60 ± 2	63 ± 2	12 ± 1	48 ± 2

Cooking oils are made of different kinds of fatty acid chains; depending on the number of double bonds it may be saturated or unsaturated. The degree of unsaturation is measured by the iodine value of the oil (Imoisi *et al.*, 2018). According to the data given in Table 1, IV of both SFO and VCO were 8.26 ± 0.06 and 8.08 ± 0.04, respectively. Since the *P*-value of both samples was less than 0.05 ($P < 0.05$), IV of SFO is significantly higher than VCO. Al-Dalain *et al.* (2011) illustrated that the addition of antioxidants (BHT and essential oils) on IV of the sunflower oil. Both antioxidants effectively reduced the oxidation rate in the oil, as detected by increases in iodine values as compared with control samples.

The maximum allowed moisture content in edible oil is 0.2% when moisture content ranges from 0.05 to 0.3, it indicates that rancidity likely to happen (Negash *et al.*, 2019). The moisture contents of all SFO and VCO samples of this study were within the range of 0.1–0.2%. According to this study, the moisture content of SFO samples was increased after one week of storage. However, this increment is not significant because the calculated *P*-value of both samples was greater than 0.05 ($P > 0.05$). The reason for this occurrence was based on the ethanol base oleoresins extraction as it did not contain free water (Abdullah *et al.*, 2018). Insoluble impurities of SFO as the same as VCO samples (0.05 ± 0.01%) and the p value of all samples were higher than 0.05 ($P > 0.05$) after one week of storage. Therefore, there is no significant difference between insoluble impurities in SFO and VCO samples.

The smoke point is a temperature at which oil begins to liberate smoke which may contain toxic fumes and harmful free radicals. Unrefined oils will have a high smoke point than refined oils because they contain impurities and free fatty acids (Katragadda *et al.*, 2010). Oils with a high smoke point are suitable for cooking at high temperatures (Eyres, 2015). The smoke point and flashpoint of SFO subjected to this study were 179° ± 3, 192° ± 2 respectively while these values for the VCO in the same order were 173° ± 2 and 186° ± 1, respectively. According to the results, the smoke point and the flashpoint of all the SFO samples were increased, and the p value of all samples were less than 0.05 ($P < 0.05$). Therefore, the smoke point and the flashpoint of SFO samples were significantly higher than that of VCO. Yen *et al.* (1997) evaluated the effects of synthetic antioxidants on the smoke point of soybean oil and lard. Their results revealed that the smoke point of soybean oil and lard was increased by adding synthetic antioxidants such as butylated hydroxyanisole (BHA), butylated hydroxytoluene (BHT) or tert-butylhydroquinone (TBHQ). At the same time, the high smoke point of lard was noticed with the combination of antioxidants. This might be due to a decrease in free fatty acids of oils. The results of this study were also revealed that antioxidants having a high capability of increasing the smoke point of oils.

Oil viscosity depends on oil type, TGs present in, frying temperature, and oil quality. The viscosity changed due to the different arrangement of the fatty acids on the glycerol backbone of the triglyceride molecule (Zahir *et al.*, 2017). The viscosity of SFO and VCO samples of this study after one week of storage were 50.812 ± 0.1 and 47.965 ± 0.1 mPa.s at 35° respectively. According to the results given in Table 1, the viscosity of SFO was increased after the incorporation of oleoresins. Since the p value of all samples was less than 0.05 ($P < 0.05$), there is a significant difference between the viscosity of SFO and VCO samples. This phenomenon may be addressed to the content of antioxidants having the capacity of controlling oxidation which consequently preventing oil from breakdown and thickening (increasing viscosity) (Soleimani *et al.*, 2018).

The specific gravity of SFO and VCO samples were 0.9711 ± 0.002 and 0.9121 ± 0.001, respectively. According to the results, the specific gravity of SFO samples was increased in comparison to the VCO samples and the *P*-value of all oil samples was less than 0.05 ($P < 0.05$). Therefore, there is a significant difference in the specific gravity of SFO and VCO samples. The reason for the increment of specific gravity of SFO samples was the addition of oleoresins, which are usually compact and dense comparatively pure oils (Parthasarathy *et al.*, 2008).

The color of SFO was increased from yellow to pale brown while decreasing *L*-value from 65 ± 2 to 63 ± 2. Hence, the darkness of SFO has been increased considerably. Further, redness (a*) of SFO has been increased from 4 ± 1 to 12 ± 1

while decreasing yellowness (b*) from 60 ± 2 to 48 ± 2. Therefore, while increasing the redness of SFO, the yellowness reciprocally coming down. Since the p-value of all oil samples was less than 0.05 ($P < 0.05$), there is a significant difference between the color of SFO and VCO samples. The reason for color change of SFO was adding the oleoresins mixture which was dark in color. Besides heating also produce many decomposed products that are also dark in color. Similar findings have been reported in color change of oils subjected to heating due to the accumulation of non-volatile decomposed products such as oxidized triacylglycerol and FFA that can lead to color changes which indicate the oil deterioration (Chandran *et al.*, 2017).

3.2 Thermal stability of spicy flavored oil compared to the pure oil

3.2.1 Effect of heating on FFA and PV of flavored and pure oil samples

High-temperature processing of cooking oils leads to the thermal degradation of fatty acids as well as consequently causes to lipid oxidation problem in edible oils. During frying, oils react with oxygen and followed by an autoxidation mechanism. Oxidation occurs more rapidly at high temperatures than the reaction which proceeds at lower temperatures (Xu *et al.*, 2015). Thermal degradation of edible oils generates many volatile organic compounds, including aldehydes, ketones, epoxides, hydroxy compounds, carboxylic acids, hydrocarbons, esters, lactones, and aromatic compounds (Majchrzak *et al.*, 2017). Acrolein and aldehydes olefin are the major volatile aldehydes, which are hazardous to human health. Other products also may be carcinogenic, cause diabetes, atherosclerosis, Alzheimer's and Parkinson's diseases, coronary heart disease, sudden cardiac death, and systemic vasculitis (Martínez-Yusta *et al.*, 2015). Nevertheless, prolonged heating also changes the organoleptic and nutritive quality of oils. Thus, in this study, the thermal stability of the SFO at 170 °C, was evaluated in terms of FFA and PV.

Figure 1 illustrates the FFA level changes with frying time. According to the results, the FFA level found to be increasing with the increment of heating time of all samples subjected to this study. VCO samples exhibited the highest FFA increment (when the temperature was maintained at 170 °C for 0, 30, 60, 90 and 120 minutes) when the temperature was maintained at 170 °C for 120 minutes and it was 0.72 ± 0.01. However, for the SFO, it was 0.48 ± 0.01 under the same conditions. Further, when the high temperature of hot oil is maintained for a long time, the increment of the FFA level has also occurred correspondently; however, the rate of increment of SFO was lower. In this study, since the calculated p value of all oil samples was less than 0.05 ($P < 0.05$), there was a significant difference between the FFA of SFO and VCO samples at all temperatures.

PV of oil samples was also increased with the increment of heating time and a high PV was observed in VCO samples comparatively SFO samples (Fig. 2). Conversely, the rate of increment of PV of SFO samples was lower than that of VCO samples. Further, as the calculated p value of all oil samples was less than 0.05 ($P < 0.05$), there was a significant difference

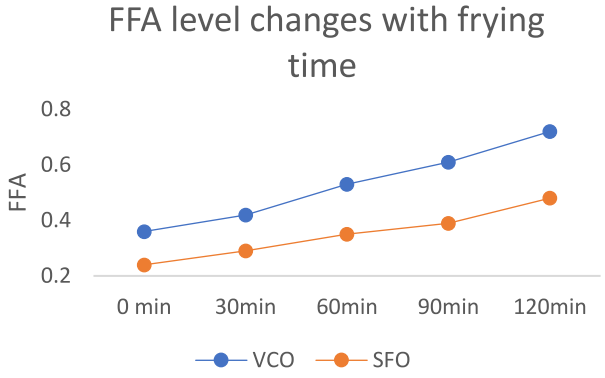

Fig. 1. FFA level changes with frying time.

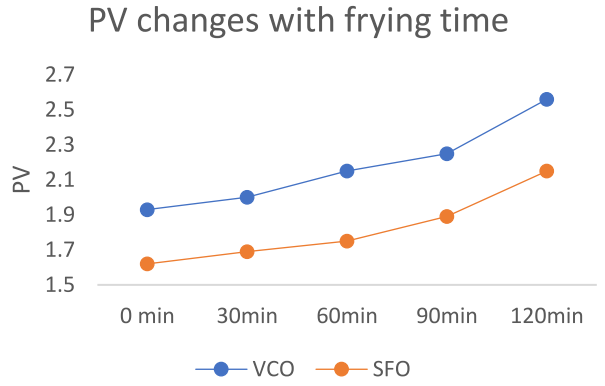

Fig. 2. PV changes with frying time.

between the PV of SFO and VCO samples. VCO samples were exhibited the highest PV (when the temperature was maintained at 170 °C for 0, 30, 60, 90, and 120 minutes) when the temperature was maintained at 170 °C for 120 minutes and it was 2.56 ± 0.01. However, for the SFO it was 2.15 ± 0.01 under the same conditions.

The low PV and FFA level of SFO may be due to the presence of antioxidants in the essential oils of pepper, garlic, cinnamon, nutmeg, and ginger that can quench the initiation and propagation steps of auto-oxidation reactions (Dapkevicius, 2002).

Srivastava and Semwal (2015) investigated that FFA level and PV of VCO during prolonged deep-frying at 180 °C for 8 h. Their results were indicated that the FFA level and PV increased significantly with respect to the frying time. Chandran *et al.* (2017) evaluated the thermal stability of coconut oil by incorporating essential oils from black pepper and ginger. Their results revealed that FFA content and PV increment were significantly low in spices incorporated oil samples compared to that pure oil samples. Similar results were found that Pradhananga and Manandhar (2018) for sunflower oil. According to their results, FFA and PV of sunflower oil were found to be increasing with the heating time

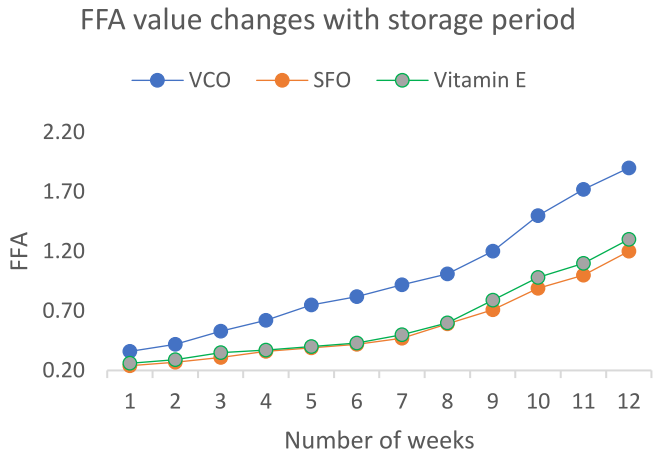

Fig. 3. FFA level changes with storage period.

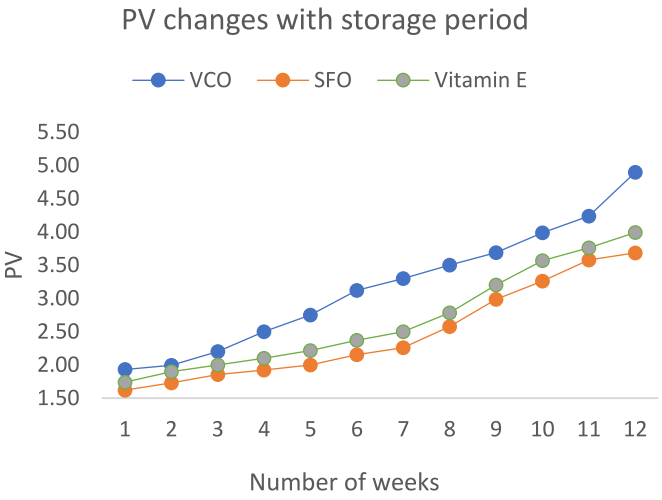

Fig. 4. PV changes with storage period.

and reported that the FFA level and PV can be decreased by adding TBHQ. Furthermore, Trivedi *et al.* (2017) reported that frying the performance of palm olein oil can be improved by incorporating synthetic antioxidants namely TBHQ and NaturFORT (combination consisting of rosemary and green tea extracts) solution for frying industries that get extra frying cycles for their products.

3.3 Shelf stability of SFO against VCO

The shelf stability of VCO and SFO samples were measured in terms of PV and FFA for 12 weeks by storing them at 60 °C. The degradation of oil is considerably high when stored in the elevated temperature while exposing it to light rather than storing under in-house conditions (Choe and Min, 2006; Ghanbari *et al.*, 2018). Inadequate processing conditions, lipase activity, or other hydrolytic actions may be led to a high FFA level. According to Figure 3, it is evident that the initial FFA level of VCO was slightly higher than that of the SFO. On accelerated storage at 60 °C for 12 weeks, a slow and steady increase in FFA was noted in all the oil samples. VCO samples were exhibited at the highest FFA level during the whole 12-week period of storage and it was highest (1.90 ± 0.01) in the 12th week (Fig. 3). In the case of SFO and vitamin E added VCO samples, FFA levels at the 12th week of storage were 1.21 ± 01 and 1.30 ± 0.01 respectively. The FFA level of all oil samples was increased with the storage time, but the rate of increment in SFO and vitamin E added oil was rather lower than the VCO samples (Fig. 3). Since the p-value of VCO and SFO samples was less than 0.05 ($P < 0.05$), there was a significant difference between FFA levels of SFO and VCO samples. However, there was no significant difference between FFA levels of SFO and vitamin E added samples because the p value was higher than 0.05 ($P > 0.05$).

A similar pattern was revealed in the formation of FFA by Iqbal *et al.* (2005). They had used sunflower oil and garlic extract in stabilizing the oil at accelerated oxidation condition and they have found that the development of FFA in blank treatment was faster than that of garlic incorporated oil. Chandran *et al.* (2017) also found a similar pattern of FFA level for coconut oil by incorporating essential oils from black pepper and ginger.

Oxidative stability of vegetable oils can be assessed in terms of PV; the amount of peroxides reveals the degree of primary oxidation and therefore is related to the rancidity (Chandran *et al.*, 2017). The steady increase in peroxide indicates the formation of hydroperoxides during the oxidation of fat. It can be observed in Figure 4. This study also illustrated that PV was increasing with the increment of the storage period of all oil samples (Fig. 4). However, VCO samples exhibited the highest PV from 1st week to 12th week and in the 12th week, it was 4.90 ± 0.01. In the case of SFO and vitamin E added samples, PV was 3.68 ± 0.01 and 3.99 ± 0.01 respectively (at the 12th week period of storage). Further, the peroxide value was increased in all oil samples against the period of storage, however, in SFO and vitamin E added oil samples, it was increased at a lower rate than the VCO (Fig. 4). Since, the *P*-value of VCO and SFO samples of this study was less than 0.05 ($P < 0.05$), there was a significant difference between the PV of SFO and VCO samples. However since, the *P*-value of SFO and vitamin E added samples were higher than 0.05 ($P > 0.05$), there were no significant differences between PV of SFO and Vitamin E added oil samples. This may be due to the radical scavenging activity of spices lead to decreased peroxide formation (Palma *et al.*, 2014).

Ramadan and Wahdan (2012) have also reported that the addition of spice extracts into vegetable oil reduced the peroxide value during storage. Similar results of increments of FFA and PV were obtained by Almeida *et al.* (2019) for crude palm oil and refined palm oil during storage. Furthermore, Shahid *et al.* (2018) were also performed a stability test for palm oil weekly to assess the effectiveness of cinnamon extract against oxidation damage or rancidity. Their results revealed that a significant increase in PV with storage time and increment was slower in treated samples than the control sample.

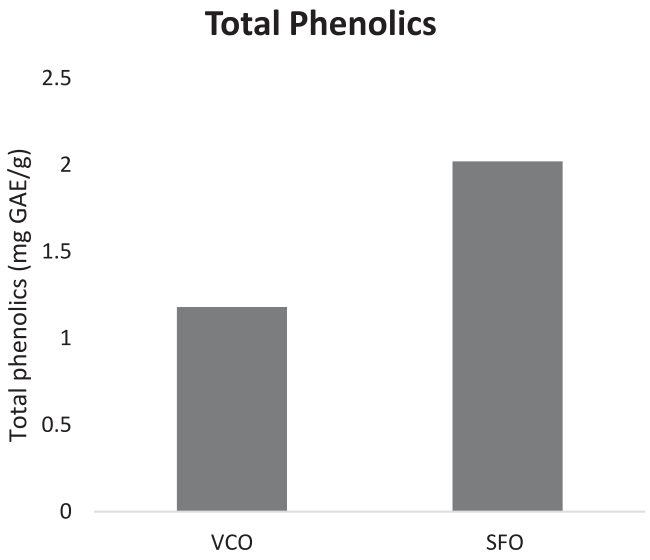

Fig. 5. Total phenolic content of VCO and SFO.

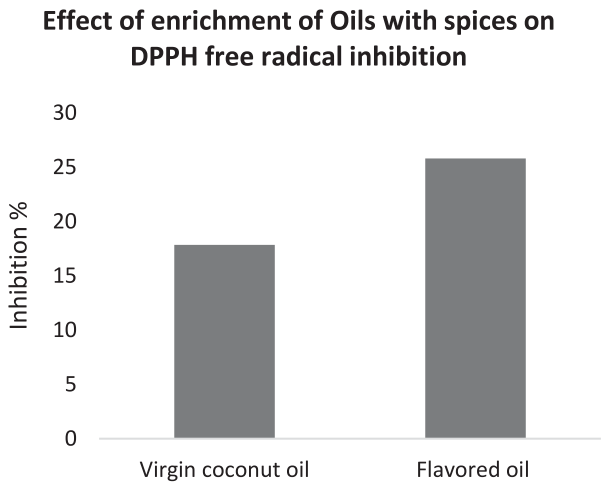

Fig. 6. Effect of DPPH free radical inhibition of VCO and SFO.

3.4 Total phenolic content of spicy flavored oil and VCO oils

Total phenolic content is a crucial parameter to evaluate the benefits over certain antioxidant activity, which gives a preliminary insight into whether the extraction of oleoresins is worthwhile for the oil industry. The main phenolic compounds found in VCO were determined by Marina *et al.* (2009). According to their findings protocatechuic, vanillic, caffeic, syringic, ferulic, and p-coumaric derivatives, are the major compounds that strongly contribute to the antioxidant activity of the VCO. The amount of phenolic acid presence in VCO is significantly higher than that of the RBD Coconut oil because some phenolic compounds were degraded during the refining process (Seneviratne *et al.*, 2009). The results obtained from this study were expressed as gallic acid equivalents ranging from 1.18 mgGAE/g to 2.02 mgGAE/g. According to the results given in Figure 5, SFO contained a significantly higher amount of phenolic compounds than the VCO; because *P*-value for all oleoresins incorporated oil samples was less than 0.05 ($P < 0.05$). The reason for this phenomenon is spices having a notable level of phenolic compounds. Hence, the total phenolic content in SFO samples was higher than that of VCO. The change in the phenolic contents in SFO was most probably due to the added oleoresins as illustrated by Figure 5.

Results of a similar study conducted by Ghani *et al.* (2018) were somewhat compatible with the results in this study. Furthermore, increments in phenolics in SFO could be explained based on the interactions taking place between the oils and the flavoring agents during the mixing phase which is being responsible for the formation of bonds between phenolics and components of spices, herbs, and fruits (Clodoveo *et al.*, 2015). McManus *et al.* (1985) have studied the binding behavior of polyphenolics and polysaccharides of the plant cell walls and found that the molecular size of polyphenols and their conformational flexibility are important to the binding. They also noticed that small changes in the structure of either the polyphenol or the polysaccharide resulted in noticeable changes in their affinity for each other. Since phenolics are the most important naturally occurring antioxidants, the direct relation of phenolics with antioxidant capacity has been reported by Robards *et al.* (1999) and according to Amarowicz and Pegg, (2019) phenolics act as a dietary antioxidant, antimutagen, antiproliferative and anti-carcinogenic agents.

3.5 DPPH antioxidant activity of spicy flavored oil and VCO

Antioxidant activity of the studied samples associated significantly with the total phenolic content in oil. Thus, phenolic content might be attributed to the high antioxidant activity in VCO. According to Figure 6, the scavenging effects of SFO on DPPH radicals were greater than that of VCO. As the *P*-value of all oil samples was less than 0.05 ($P < 0.05$), there is a significant difference between the antioxidant activity of SFO and VCO samples. The reason for this phenomenon is the methanolic extract of spices as it contains a considerable number of phytochemicals including polyphenols that are reported to have considerably high free radical scavenging and peroxide inhibition activity (Hamad *et al.*, 2015; Okhli *et al.*, 2020). Active components in spices, the concentration of the constituents, and extraction procedure are some of the important factors that affect the strength of the extract (Dua *et al.*, 2013). Thus, the improved oxidative stability of the treated oils can be attributed to the presence of extracted lipophilic phenolic compounds (Žanetić *et al.*, 2013). Taha *et al.* (2014) found that only lipophilic components of the herbal materials were dissolved in the oil while leaving polar phenolic compounds

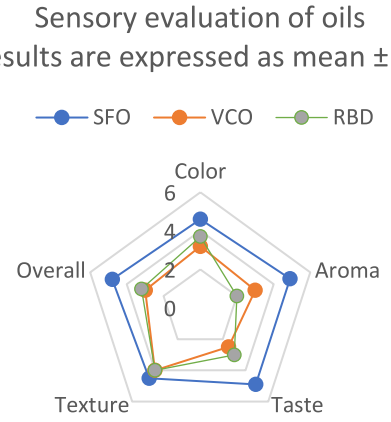

Fig. 7. Sensory evaluation of oils.

undissolved because they are not fat-soluble. Hence lipophilic components in spices are merely responsible for the antioxidant activity of the oil.

A research was conducted by Wang *et al.* (2008) employed 45 oils essential to determine the antioxidant activity via total phenolics content and DPPH free radical scavenging activity. They found out that the best antioxidant activity was having with the cinnamon leaf oil and clove bud oil and have mentioned that both natural and synthetic antioxidants can inhibit or postpone the process of fat oxidation. Moreover, Shahid *et al.* (2018) also described that antioxidants can play an important role in the prevention of oxidation of fat and oil by acting as a reducing agent, free radical scavenger, chelator, and singlet oxygen scavenger.

3.6 Sensory analysis

Sensory analysis of the developed SFO was carried out and compared with VCO and RBD coconut oil. On sensory evaluation, it was found that SFO was highly preferred by the sensory panelists in terms of color, aroma, taste, texture, and overall acceptability (Fig. 7).

3.7 GC-MS analysis for oils

Gas chromatography (GC) techniques can be widely applied for the analysis of major FA and minor constituents in edible oils. In this study, oleoresins extracted from selected spices were incorporated into VCO and magnitude of augmentation of bioactive compounds from oleoresins were measured in terms of GC-MS method. According to the analysis of VCO samples as per fatty acid methyl esters, were found out that eugenol, cinnamaldehyde, piperine, and other flavor compounds (Fig. 8) in SFO samples and none of them have presented in VCO samples (Fig. 9).

3.8 SPME – GCMS analyze for volatile compounds in SFO

Solid phase microextraction (SPME) is a micro-sampling technique which has found wide application in flavor and fragrance research. It is a solvent-free method that is used to trap flavors and fragrances either from aqueous samples (immersion SPME) or from the vapor space above a liquid or a solid sample (headspace SPME).

According to the SPME analysis of SFO samples of this study (Fig. 10), was found out that following bioactive compounds namely, cinnamaldehyde, eugenol, piperine, caryophyllene which are natural bicyclic sesquiterpenes of many essential oils, especially found in clove oil (Ravindran and Divakaran, 2012) and 2-methoxy-4-(2-propenyl) phenol which is a constituent of clove oil, nutmeg, cinnamon, and bay leaf (Srivastava *et al.*, 2005).

4 Conclusions

This study was conducted to develop a SFO along with high thermal stability as well as extended shelf life. VCO was selected for this study because; it is widely used for cooking purposes and other food-related commercial applications. To prepare the SFO oil ginger, garlic, nutmeg, black pepper, cinnamon, and cloves oleoresins were incorporated into oil samples. SFO and VCO samples of this study showed different physiochemical properties. FFA and PV were decreased, and SV, IV, viscosity, and specific gravity were increased of SFO relative to the same parameters of VCO. Moisture content and insoluble impurities of SFO and VCO samples were remained almost the same during the period of study. The smoke point and flash point of SFO were increased. Since, the spices containing antioxidants, the thermal stability of SFO samples were better than that of VCO. Increment of FFA level and peroxide value of SFO samples occurred at a slower rate than that of VCO samples during the study period. The shelf life of SFO samples was analyzed according to the FFA and peroxide value. In SFO oil, FFA and peroxide value were increased at a slower rate than the VCO samples. Shelf-life testing further revealed that oleoresins and vitamin E incorporated samples having the same rate of increment of FFA and PV. Total phenol content and antioxidant activity of SFO samples were increased considerably against VCO samples. Therefore, SFO samples were less susceptible to oxidation, as they are rich in antioxidants of spices. Hence, SFO oil is a better option for the future oil-related food industry.

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Conflicts of interest. The authors declare that they have no conflicts of interest in relation to this article.

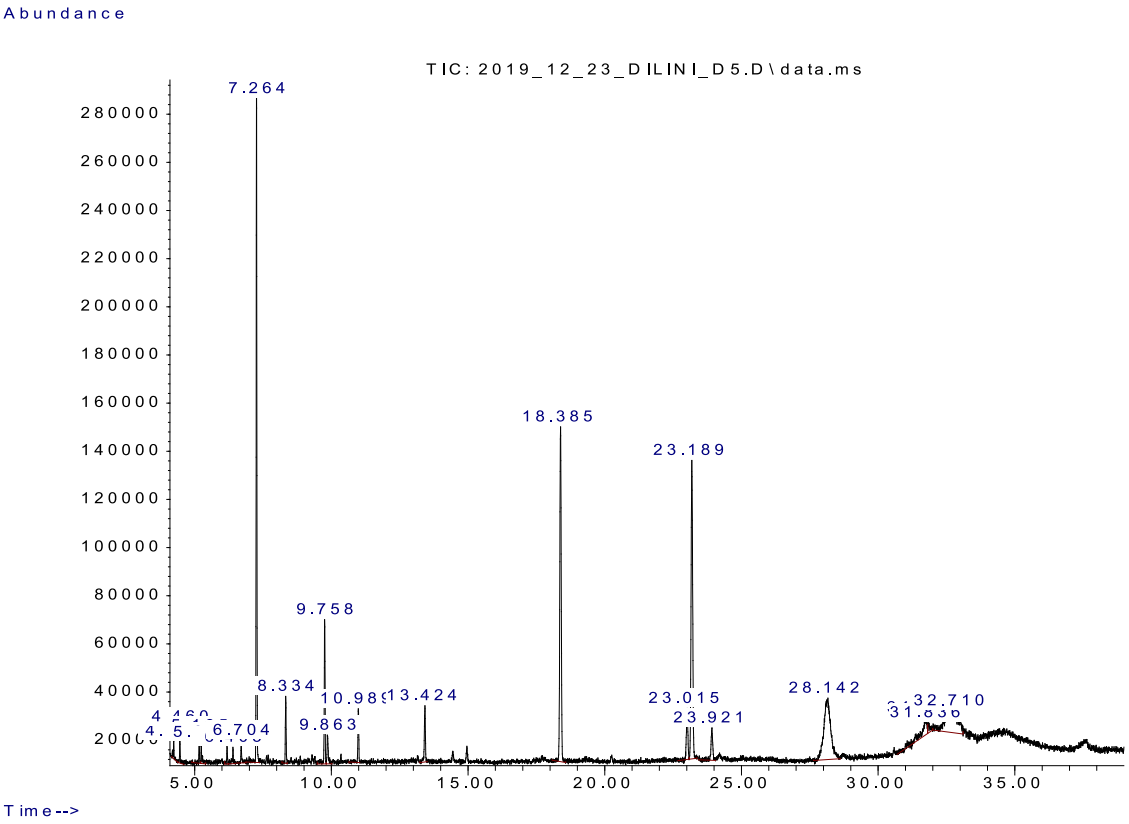

Fig. 8. Major fatty acids and flavor compounds in flavored oil (7.264 = eugenol, 9.758 = lauric acid, 13.424 = cinnamaldehyde, 18.385 = palmitic acid, 23.189 = myristic acid, 28.142 = piperine).

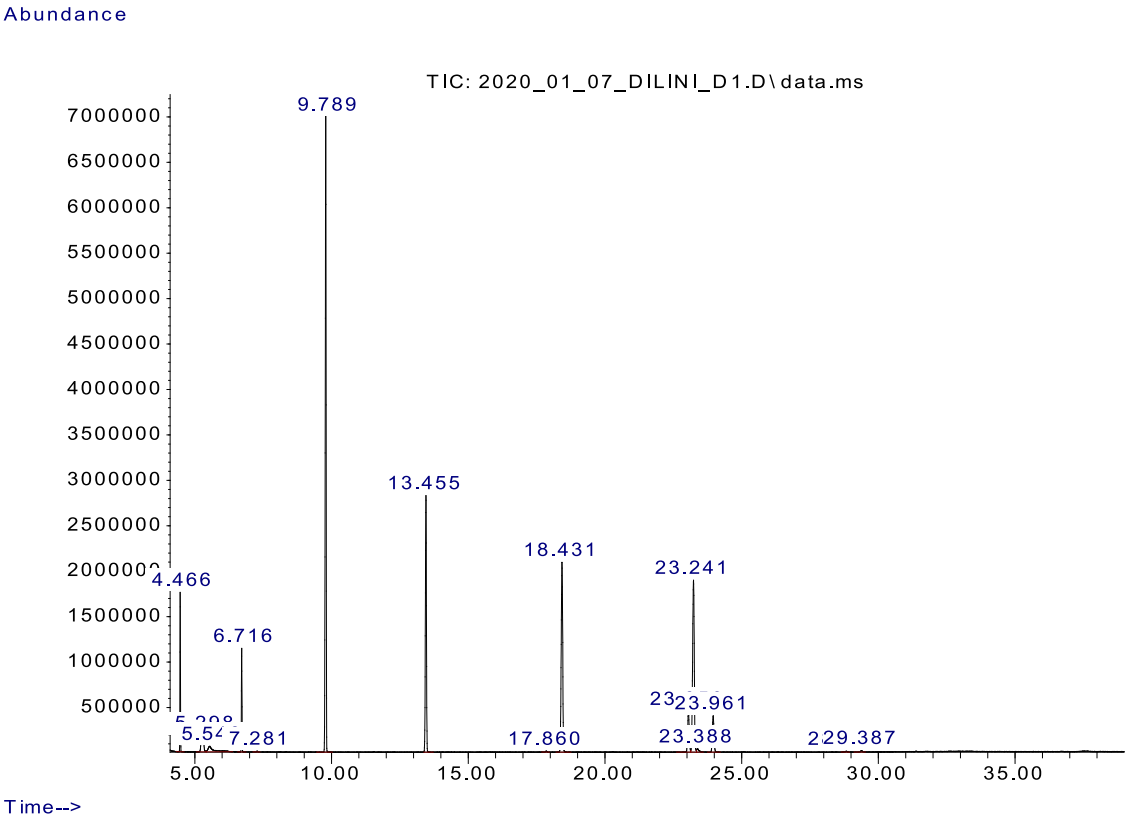

Fig. 9. Major fatty acids pure coconut oil (9.758 = lauric acid, 13.455 = capric acid, 18.431 = palmitic acid, 23.241 = myristic acid).

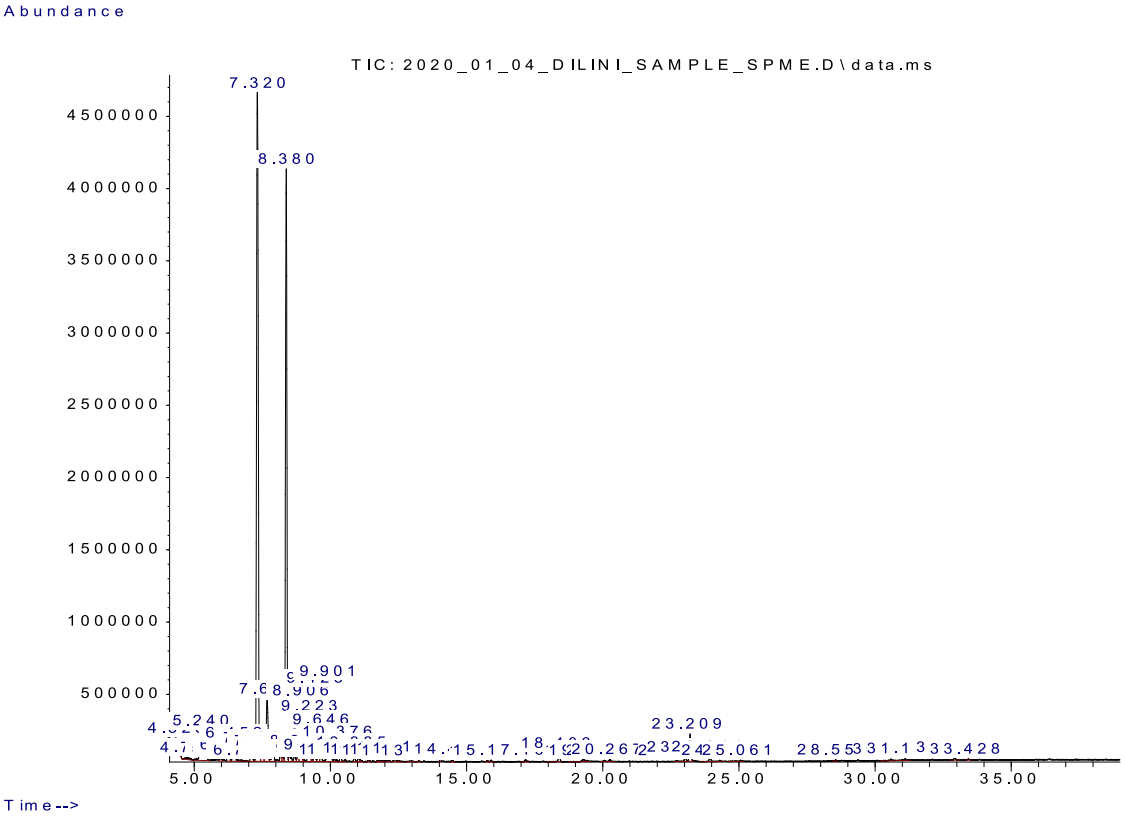

Fig. 10. SPME results for SFO (7.320 = phenol,2-methoxy-3-(2-propynyl), 8.380 = caryophyllene, 7.684 = eugenol, 10.424 = cinnamaldehyde, 28.532 = piperine).

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