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Physico-Chemical Alterations In Vegetable Oils On Deep Fat Frying: A Review.

Rita Awasthi[1]*. and Anushri Pandey[2].

[1]Professor, Department of Chemistry, Brahmanand College, Kanpur 208004, Uttar Pradesh, India.
[2]M. Sc Sem-3, Department of Chemistry, Brahmanand College, Kanpur 208004, Uttar Pradesh, India.

ABSTRACT

In food industries, deep-fat frying is widely used because of oil's low cost, demand and nutritional value. The process of deep frying is based on the oil-food interaction at high temperatures, which cooks and dehydrates the food, leading to physical and chemical changes. Some food and oil compounds are lost during the frying process, and potentially toxic compounds are developed in the oxidized oil. The purpose of this review is to understand the changes in food caused by the deep fat frying process over the years.
Keywords: Deep fat frying, Composition of food, oxidative degradation, fatty acid composition, frying oil quality.

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**Corresponding author*

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INTRODUCTION

Deep-Frying or deep fat frying has emerged out to be most popular technique of cooking out of various techniques practiced since ages. It is so because people nowadays love to have crunch in every bite. Deep-frying is becoming increasingly popular in industrial and home-cooked foods. It has been reported that almost half of the lunch and dinner food orders in commercial restaurants contain one or more fried products [1-4]. Deep frying basically is a method where a piece of food is fully submerged in a pan full of hot oil at temperature ranging from 350°F (177°C) to 375°F (191°C)) resulting in quick and even cooking and a crispy outer layer is developed. Sometimes deep frying oil can reach temperature of over 400°F (205°C). Usually a deep fryer, chip pan, pressure fryer or vacuum fryer maybe used but mostly it is done in a pot full of oil also known as hot fat cooking method. It was around nineteenth century when concept of deep-frying gained popularity before that cookbooks claim that European countries where the first one to adopt it and gradually it spread to other parts of America. In the United States, the Chicago tribune notes that "you can deep-fry almost anything". deep-fried food has become very popular in recent years and is also described as a staple of almost all state cuisines everywhere around the world [5]. On daily basis billions of people consume fried items as they believe that life is too short to think and eat as a result there has been a tremendous spike in number of heart-attack and cardiac arrest cases worldwide. Not only this but fried-food has also become a potential reason behind increasing number of cancer patients. Some scientists believe that compounds formed during the process of frying like 'acrylamide' (chemical formed when food rich in carbohydrates are cooked at very high temperature) can damage the genes potentially causing genetic mutations. But on the contrary bunch of people also believe that it is high time that people should start consuming mindfully as they realize that counting calories is just not enough, the sizzling oil in the pot may incorporate chemical compounds like acrylamide, acrolein, heterocyclic aromatic amines (HAAs), polycyclic aromatic hydrocarbons (PAHs) etc. in the food which are carcinogenic in nature. On daily basis billions of people consume fried items as they believe that life is too short to think and eat as a result there has been a tremendous spike in number of heart-attack and cardiac arrest cases worldwide. Not only this but fried-food has also become a potential reason behind increasing number of cancer patients. Some scientists believe that compounds formed during the process of frying like 'acrylamide' (chemical formed when food rich in carbohydrates are cooked at very high temperature) can damage the genes potentially causing genetic mutations. But on the contrary bunch of people also believe that it is high time that people should start consuming mindfully as they realize that counting calories is just not enough, the sizzling oil in the pot may incorporate chemical compounds like acrylamide, acrolein, heterocyclic aromatic amines (HAAs), polycyclic aromatic hydrocarbons (PAHs) etc. in the food which are carcinogenic in nature.

Chemistry Of Vegetable Oils

Oils and fats are glyceryl ester of glycerides of higher fatty acids [6-8]. Those which are liquids at ordinary temperatures are called oils. The largest source of the oil seeds produced by the annual plants in our country are linseed oil, soybean oil, cotton seed oil, groundnut oil, sunflower oil, safflower oil, mustard oil, castor oil, and sesame oil etc. Hence, oil seeds play an important role in planning the development of the country. The trend of consumption of oils and fats is shifting from saturated fats to fatty oil rich in polyunsaturated fatty acid (PUFAs) because of more nutritional value they are receiving increasing attention. It is becoming important to formulate new vegetable oil composition of improve stability and nutritional value. The nutritive value of edible value of edible oil depends on the fatty acid profile, degree of unsaturation, arrangement of fatty acid in triglycerides structure. According to WHO, a healthy oil should have three characteristics:

- The ratio of saturated, mono and polyunsaturated should be 1:1.5:1
- Ratio of essential fatty acid linoleic acid: linolenic should be 5:10:1.
- Presence of oxidants

Vegetable oils are used worldwide for the purpose of frying and the main reason behind extra flavorful fried foods. Vegetable oils can be of different botanical origin and composition depending upon the region. some of the common known vegetable oils are sunflower oil, peanut oil, corn oil, palm oil, rapeseed oil, soyabean oil, rice bran oil, etc. these frying oils largely effect the food quality, texture, taste, aroma, etc. These oils are rich in monosaturated fatty acyl chains (MUFA) while oils like palm and coconut oil are rich in saturated fatty acyl chains (SFA) the type of oil used in frying mainly depends upon its availability in the region, economic feasibility, traditions etc. the major component of these oils are triglycerides (TG) approximately 95-98% of the total lipid content whereas minor components include terpenic lipids like vitamin E isoforms (mainly tocopherols and tocotrienols) , carotenoids, phenolic compounds, chlorophylls, phospholipids, sterols etc. The fatty acids of the oils esterified with the glycerol backbone of the TG called as fatty acyl chains (FA) are of two types namely unsaturated (UFA) or saturated (SFA). Further former is again classified into monosaturated (MUFA) or polysaturated (PUFA) depending upon the number of single or multiple double bond present in the structure. MUFA rich oils are prioritized over SFA rich oils due to nutritional reasons. Vegetable oils rich in MUFA are olive oil, peanut oil high oleic sunflower oil, rapeseed oil etc. some vegetable oils rich in PUFA are soyabean oil, corn oil, maize oil whereas palm kernel oil rich in rich in SFA.

Mechanism Of Deep- Fat Frying

Deep-frying is a complex process resulting in multiple reactions along with chemical and physicochemical changes occurring throughout the process. Frying oil not only adds texture and flavors to the fried food but also transfers mass and heat from the point when food is immersed in the hot oil and a contact between food, oil and air is developed. The oil subjected to deep fat frying in the presence of air and moisture undergoes series of chemical reactions – oxidation, hydrolysis, polymerization and thermal decomposition, that further results in the formation of oil and food degrading carcinogenic chemicals, which cam be non-volatile or volatile in nature. The latter evaporates along with steam in the environment or absorbed by the fried food whereas former remains in the oil and stands responsible for the changes in the chemical and physical properties of the oil. For example, it deteriorates the unsaturated fatty acids (UFA) of oil and increases color, specific heat, free fatty acids, density, viscosity and polarity of the material [9].

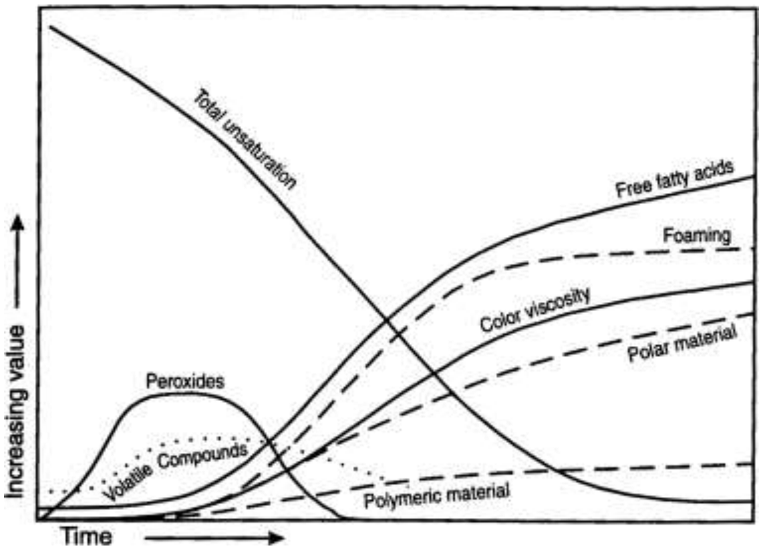

Figure 1 : Physical and chemical changes during deep-frying [9].

Physicochemical Changes During Deep-Frying

Oxidation

Oxidation reaction occurs mainly in oils rich in PUFAs and MUFAs it is a primary degradation process oxygen reacts with double bonds present in unsaturated fatty acids to form lipid hydroperoxides which are very unstable at frying temperatures and breakdown into secondary products like aldehydes, ketones, epoxides, alcohols, and dicarboxylic acids.

$$RH + O_2 \rightarrow ROOH \rightarrow R'CHO + R''COOH$$

These products are toxic, carcinogenic and mutagenic in nature. This thermal oxidation involves three steps: initiation, propagation and termination [10, 11].

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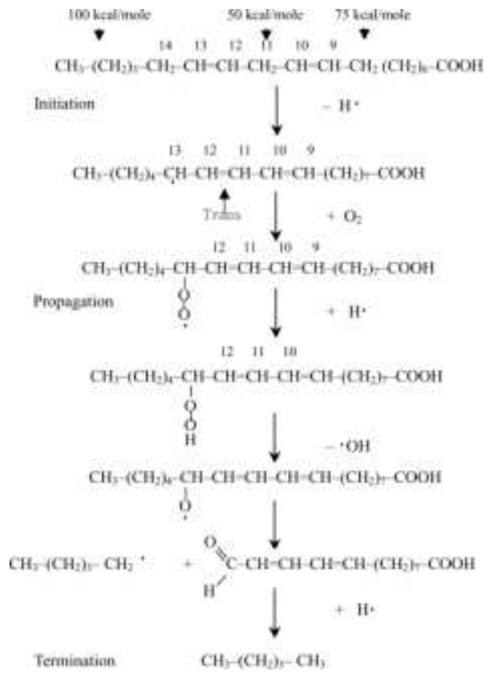

Figure 2: Initiation, propagation and termination process.

Hydrolysis Of Oil

During the process of frying, the moisture content of the food being fried forms steam and evaporate [12], resulting in thermal hydrolysis. It is a reversible processes where water reacts with triglycerides (TGs) by breaking ester bonds. During thermal hydrolysis the moisture in the oil acts as weak nucleophile attacks the ester linkage of TAGs, to produce diacylglycerols, glycerol's and FFA (free fatty acids). Measure of these products determine the quality of oil [13].

$$\text{triglyceride} + \text{H2O} \rightarrow \text{diglyceride} + (\text{FFA})$$

Polymerization

Polymerization results in the formation of foam on the oil surface and oil becomes more viscous, sticky and darker in appearance. It takes place when free radicals from oxidation come together form high molecular weight polymers. The conjugated dienes and free radicals form high molecular weight polymers. This results in increase in the total polar compounds (TPC).

$$\text{R}\cdot + \text{R}\cdot \rightarrow \text{R} - \text{R (dimer)}$$

$$\text{ROO}\cdot + \text{ROO}\cdot \rightarrow \text{polymeric peroxide}$$

Thermal Decomposition

During decomposition cleavage of carbon-carbon bond takes place resulting in formation of short chain hydrocarbons. It takes place due to very high temperature or prolonged frying often leads to release of volatile organic compounds and loss of nutritional values.

$$\text{R} - \text{CH} = \text{CH} - \text{CH}_2 - \text{COOH} \rightarrow \underset{\text{(acetic acid)}}{\text{CH}_3 - \text{COOH}} + \underset{\text{(propene)}}{\text{CH}_3 - \text{CH} = \text{CH}_2}$$

The oil seeds or the source of vegetable oil play a very important role in deciding the quality, availability and economic scenario of the country. In India the largest source of oil seeds produced by annual plants are sunflower oil, castor oil, mustard oil, sesame oil, peanut oil, cotton seed oil, linseed oil, soyabean oil etc [14].

Degradation Chemistry Of Various Oils

Sunflower Oil

Sunflower oils are PUFA rich containing approximately [60-65%] of linoleic acid. Due to presence of multiple double bond, it shows high reactivity with O_2 at high temperatures. It undergoes auto-oxidation of linoleic acid ($-CH = CH - CH_2 - CH = CH -$) to form lipid hydroperoxides (*ROOH)* which further decomposes into aldehydes (malondialdehyde, 4-hydroxynonenal) and ketones. It can also undergo polymerization during which cross -linking forms sticky polymers as a result increase the viscosity of the oil and increased oil absorption by the fried food. Such circumstances lead to loss of vitamin E (tocopherols).

Soyabean Oil

Soyabean oils are PUFA rich containing linoleic and linolenic acid. Its high PUFA content makes it more vulnerable hence very unstable and shows rapid hydroperoxide formation. Hexanal formed gives grassy smell. Shows rancidity hence very unsuitable for deep-frying.

Rice Bran Oil

It consists of approximately 40% MUFA and 35% moderate PUFA and oryzanol antioxidants. They are rich in natural antioxidants such as tocopherols. During their degradation although hydroperoxides form from PUFA content but γ-oryzanol delays breakdown. It also shows less polymerization as compared to sunflower oil hence stable for multiple frying cycles and can be considered as healthiest frying oil according to Indian context.

Groundnut Oil

It is MUFA rich oil containing ~50-60% of oleic acid. Due to high percentage of MUFA content it is more stable. Hydroperoxide forms but at slower rate. It promises better frying stability, less rancid smell and preserves flavour of the snack oleic acid *(-CH=CH-)* at C9 oxidises slower than linoleic acid.

Palmolein Oil

This oil is the combination of ~40% SFA (palmitic), ~40% oleic acid (MUFA) its PUFA content is low hence it is very stable under frying conditions. Palmitic acid present is heat stable, shows less PUFA oxidation and hence low aldehyde production but on overheating produces acrolein. Although its stable frying oil but high palmitic acid content can help elevate the LDL cholesterol level.

Safflower Oil

Safflower is grown on both irrigated and semiarid lands in the southwestern United States. Safflower oil has a high linoleic acid content (73%) and a low linoleic acid content (<1%), and for these reasons it commands a premium price. A high-oleic safflower oil variety is also available, which contains 78% oleic and 12% linoleic acids.

Olive Oil

Olive oil is a rich source of MUFAs, with oleic acid being the most abundant. Olive oil also contains PUFAs, including omega-6 and omega-3 fatty acids. omega-6 to omega-3 ratio in olive oil is relatively high, around 20:1. Olive oil also contains a small amount of saturated fatty.

Literature Review

Various scientists and research scholars have worked on studies to analyse the changes that occur in different vegetable oils caused by deep fat frying. Various spectroscopic methods can be employed to study these changes like FTIR, NMR, GC-MS etc. peroxide value, acid value, iodine value are also used. A brief review of that is mentioned below.

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At National level

S. Ganguly [15], studied regarding various edible oils obtained from plants and the presence of carboxylic acid group *(COOH)*. They also demonstrated the comparative study of different types of frying oils, keeping focus on their composition and physicochemical properties also talks about major edible oils used in India. MA. Shad [16], presented a detailed study on antioxidant potential along with physical properties of different oils like sunflower, canola, rapeseed etc.

P K. Nayak et. Al [17], studied the reactions like oxidation, hydrolysis, polymerisation, and also about the by-products such as FFA, alcohol, cyclic compound, dimers and polymers obtained.

Krishna and Swamy [18] investigated the formation of carcinogenic compounds such as malondialdehyde and polycyclic aromatic hydrocarbons when oils are used repeatedly along with their chemical transformation, discussed the policy loopholes and the FSSAI regulations that order FBO_S to discard oil after four times of use or when TPC levels reaches 25. Diseases like hypertension and Alzheimer's caused by consumption. SFA, PUFA, MUFA and how double bonds are more prone to oxidation. Mentions about TAG_S usually > 95% along with diacylglycerols <5% and tocopherols up to 900 $mgKg^{-1}$.

P V Bhavadharani, G. Parameswaran and A. Sakkaravarthy [19] conducted a demonstration based on different types of oils commonly available and used in the market. α-tocopherols, retinol, calciferol to fortify the oils and heated at 220°C and their stability was governed. Acid value (mgNaOh\g) of deep-fried oils were noted peroxide values (meq of active O_2/kg) of coconut oil, groundnut oil, sesame oil, rice bran oil, palm olein oil and refined sunflower oil at same temp came out to be 1.25, 2.6, 3.2, 1.5, 6.98 and 2.25.

Yadav et. Al [20], studied about the heat transfer, oil uptake and moisture loss of the fried food changes that occur in the oil during degradation process a nd how after certain time of usage oil becomes unfit for consumption and about oil quality parameters like peroxide value (PV), acid value (AV) and free fatty acids.

S Asokapandian , G J, Swamy and H Hajjul et. Al [21], investigated detailed procedure of deep fat frying and about the pre-frying preparations, characteristics and quality of frying oils. Advantages and disadvantages of the whole process were also studied by the authors.

M A Ali et. al. [22], studied microwave heating and its effect on blend of canola and palm oil as results of other heating processes do similar harm as in other cooking techniques. In their study they found that Palmolein oil (PO) is more oxidative stable than canola oil (CO) which is heat sensitive. A blend was prepared in the volume ratio of 40:60 (PO:CO) and tested it for different time intervals by employing different instrumental and spectroscopic techniques. Result demonstrated that blending resulted in deacceleration of rate of deterioration of CO.

M. Choudhary and K. Grover [23], observed the physicochemical properties of rice bran oil (RBO) and its blends upon deep fat frying. All the RBO blends were repeatedly fried and different characteristics were observed. It was found that high oxidative property of RBO blends make it more stable, a 100 ml mixture of RBO and other vegetable oil was used for each blend. S. Debnath et al [24], studied the effect of frying cycles and heat transfer on RBO the frying of poori an Indian dish, was carried out in five bathes, each for 3 minutes in a day and for 6 frying cycles. The convective heat transfer coefficient decreased due to increase in kinematic viscosity of RBO. RBO shows best oxidative resistance. The frying oil is discarded when the percentage of FFA reaches 1.0% there was no significant difference (P >0.05) in moisture content of fried poori.

P.K. Ghosh, D. Chatterjee and P. Bhattacharjee [25], observed the antioxidant stability in soyabean oil. It has a PUFA rich profile and low stability, shows high trans and aldehyde formation. Titrimetric analysis of oils was carried out. Iodine value was calculated by AOAC method. TOTOX value was calculated by [TOTOX = 2 PV + p-AV] where TOTOX is total oxidation value . absorption spectra were recorded.

R. Ismail [26], presented a study on palm and palmolein oils and their industrial production of pre-fried frozen French fries, potato chips and other fast foods. Palmolein has good frying life suitable for Indian snack frying and methods.

At International level

B. Yilmaz, T.O. Sahin and D. Agagunduz [27], studied oxidative changes in ten vegetable oils due to deep frying processes of Solanum tuberosum. They analyzed the thermos -oxidative alterations with 10 commonly used culinary oils. One liter of each oil was used under constant conditions. Malondialdehyde and peroxide value of oil like vegetable ghee came out to be [0.669 nmol/gm]and [21.0mEqO_2/KG], higher than oils like hazelnut oil, olive oil, palm oil etc.

E. Yuki and Y. Ishikawa [28] observed that saturated oils like palm and coconut possess higher initial active oxygen method values as compared to unsaturated oils but, as a result of deep frying there was a sharp decrease in the values of saturated oils.

W.H. Morison III, J.A. Robertson and D. Burdick [29], studied the effect of deep fat frying on sunflower oil, their oxidizing capability after every heating was recorded also active oxygen method value were determined and a graph between log of active oxygen method values v/s time was plotted that came out to be a straight line. Its slope gives the oxidiazibility of the oil.

E. Choe and D.B. Min [30] investigated the chemistry behind deep-fat frying oils, hydrolysis, oxidation and polymerization. Other factors responsible for these reactions such as quality of food and oil, frying conditions, types of fryers used, antioxidants, no. of frying's, temperature, content of FFA etc. They studied how oxidation produces hydroperoxide and aldehydes, ketones and alkanes and alkenes, hydrolysis increases concentration of FFA, Diel's alder reaction results in the formation of dimer and polymer.

S Multari et al. [31], observed different vegetable oils like rapeseed oil, soy and sunflower oil for short term (60 minutes) of deep-frying and noted the changes in the FFA composition, oxidation, soy oil produced highest level of because of thermal oxidation and rapeseed oil aroma had Sulphur content. Sunflower oil showed a significant decrease ($P>0.05$) in total terpenes and aldehyde rise. Providing a table of comparison of these oils and their suitability.

C S P Santos et al. [32], investigated the monounsaturated rich vegetable oils for impact of deep frying. Oils like peanut oil (PO), canola (CO) and extra virgin olive oil were employed. PO showed conjugated dienes.

S Casal et al. [33], studied stability of olive oil during deep-frying to check its suitability for domestic use. It was observed after every 3 hours of frying and peroxide and p-anisidine, oxidative stability, fatty acids, vitamin E and acidity values were collected up till the top\tal polar compounds TPC values reach the maximum level of 25%.

L.M. Garcia et al. [34], observed the PUFA rich vegetable oils to demonstrate the changes in the toxic volatile values during deep-frying. PO, CO AND EVOO were studied due to their high MUFA:PUFA ratios. Mass spectrometry, chromatography and solid phase micro extraction techniques were employed for the purpose. Initial fatty acid composition largely defined the composition of aldehyde formed after deep frying. Formation of unsaturated aldehyde were lower in EVOO whereas CO produced less volatile.

L Bruhl [30] investigated the alterations in oils upon frying. Fats usually degrade during heating and double bonds leads to the formation of trans fatty acids. B-scission can also occur at carbonyl or alkyl side of the oxygen bearing C- atom in the FA chain followed by the formation of aldehydic keto and hydroxyl acids, short chain fatty acids appear along with volatile compounds.

W.P. Quek et al. [35], studied different vegetable oils and their performance during deep-frying from the various studied that have been conducted and their data was used for comparison on the basis of rate of change of parameters palm oil as compared to other soft oils was found to be higher in stability. The quality parameters of palm oil, soyabean oil, sunflower oil and canola oil were examined.

D. Dodoo et al. [37], observed and evaluated sunflower oil, coconut oil and soyabean oil for their qualities on repeated heating. Found that oils having low unsaturated fatty acid were thermally stable. Repeated heating resulted in catastrophic thermal changes. It was concluded that highly saturated oils are less susceptible to thermal changes during deep frying.

C. Gertz, S. Klostermann and S.P. Kochhar [38], developed a method where refined and non-refined oils were heated at 170°C and addition of silica gel containing water and oil stability at different frying temperatures were measured.

A C Godwill et al. [39], studied the values of peroxide and refractive index of sunflower, palm olein and sesame oils. The value of refractive index (RI) for sunflower oil increased from 1.4722 to 1.4724 whereas palm olein oil has lower R.I due to higher saturated fat. Lower peroxide value indicates freshness of oil, higher value indicates rancidity.

F. Sciano et al. [40] talks about generation of toxic aldehydes in vegetable oils subjected to high-temperature cooking processes, such as frying, poses significant health risks due to their high reactivity and potential to form carcinogenic and mutagenic compounds. This review discusses the mechanisms of aldehydes formation in vegetable oils, focusing on key factors such as oil composition, cooking temperature, and heating time. The major toxic aldehydes identified include acrolein, acetaldehyde, formaldehyde, *t,t*-2,4-decadienal (*t,t*-2,4-DDE), 4-hydroxy-2-hexenal (4-HHE), and 4-hydroxynonenal (4-HNE), which have been associated with adverse health effects ranging from respiratory irritation to carcinogenicity.

B. Yilmaz, T.O. Sahin and D. Agagunduz [27], analysed the thermo-oxidative alterations that occur during deep frying with 10 commonly used culinary oils. Deep frying processes (180 ± 5°C) were carried out with a total of one liter of each type of oil and 100-g potatoes at every turn (sliced into 1 cm∗1 cm∗6 cm pieces). The process was carried out keeping the conditions constant for all oils and was repeated 10 times consecutively for all oils. The malondialdehyde and peroxide values of vegetable ghee (VG) were found to be 0.669 nmol/g and 21.0 mEqO2/kg, respectively, and were higher than those of other oils. J. Kim et al[42] studied different techniques to improve fried food quality and reduce oil absorption. H. Yun et al.[43] developed an improved test kit for detecting polar compounds in used frying oils and to compare the distribution of polar compounds across different types of cooking oils. The polar compound distribution was analysed in six types of oils: palm oil, coconut oil, rice bran oil, sunflower oil, canola oil, and soybean oil. Coconut oil was found to be the least suitable for frying due to the rapid formation of polar compounds.

CONCLUSION

Various experimental work and studies conducted worldwide and the literature reviews mentioned in this paper clearly highlights the degradation in composition of different vegetable oils during deep-fat frying. The paper also presents the interplay of physicochemical reactions such as oxidation, hydrolysis, polymerization and isomerization, that results in the active degradation of oil quality and its nutritional value. The literature review highlights various studies conducted on the changes occurring in pure oils during deep fat frying. Researchers have investigated the physicochemical changes, fatty acid composition, oxidative stability, and nutritional evaluation of different oil blends to understand their properties and potential benefits. Antioxidants including TBHQ, BHT, and BHA have been proven to extend the shelf life of oils while storage and frying at high temperatures. These antioxidants provide protection and aid in the stabilization of oils by lowering peroxide values (PV), free fatty acids (FFA), and color index. Synthetic antioxidants have shown potential in preserving oil quality.

For many years, fried foods and frying oils have become frequently addressed topics in the literature well as among consumers and the food industry. Researchers especially tried determining the thermo-oxidative alterations that occur in oils and to determine the oil types that best affect the sensory properties of fried foods. Although there are some of the common parameters witnessing the degradation in different types of vegetable oils commonly used for deep-fat frying worldwide, the extent of these changes majorly depends upon the degree of unsaturation, antioxidant content, smoke point and fatty-acid composition of the oils. The temperature maintained during deep-frying also stands responsible for the quality of food resulting from the deep-frying process temperature below 180°C and not more than 200°C or above is usually recommended to reduce the possibility of formation of trans fatty acyl chains.

The literature review highlights various studies conducted on the changes occurring in pure oils during deep fat frying. Researchers [40] have investigated the physicochemical changes, fatty acid composition, oxidative stability, and nutritional evaluation of different oil blends to understand their properties and potential benefits. Antioxidants including TBHQ, BHT, and BHA have been proven to extend the shelf life of oils while storage and frying at high temperatures. These antioxidants provide protection and aid in the stabilization of oils by lowering peroxide values (PV), free fatty acids (FFA), and color index. Synthetic antioxidants have shown potential in preserving oil quality.

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