Chapter 6:

Understanding Lipid Oxidation Influences in Halalan Toyyiban Products

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Food is a fundamental human necessity, providing essential energy and supporting body growth through various macro-components such as carbohydrates, proteins, lipids, and minerals. Among them, lipids play a crucial role as a concentrated form of energy and carriers of fat-soluble vitamins in the diet. Besides their nutritional value, lipids enhance the organoleptic properties of food formulations. Frying with lipids has been a common practice to improve the overall flavor, aroma, and textural properties of foods. However, high-temperature processing conditions can significantly influence the nutritional value and quality of lipids as well as fried food products. A critical factor in this regard is lipids oxidation, which directly affects the palatability and shelf life of foods. This chapter aims to discuss the mechanisms of lipid oxidation, the influence of both intrinsic and extrinsic factors on lipid oxidation, the effect of frying lipid oxidation, and strategies to mitigate or delay oxidation in lipids.

1 Introduction

Lipid is one of the most important subclasses of the human diet; they are a concentrated form of energy and carriers of fat-soluble vitamins in our diet. Apart from their nutritional role in the human diet, they are utilized for delivering better organoleptic properties that include flavor, aroma, desirable texture and mouth feel in food formulations (Marikkar & Yanty, 2018). Lipids are also used as fat ingredients in bakery product formulations. Shortening and margarine are frequently employed as ingredients in bakery products to achieve desirable baking outcomes (Yanty *et al.*, 2017). For a long time, lipids have been used as a frying medium to improve the taste, texture, and mouthfeel of foods. According to recorded history, frying as a method of food preparation was introduced by the ancient Egyptians as early as 1600 BC. With the invention of the frying pan, deep-fat frying became commonplace in Asia, Africa, and Europe. The development of the continuous fryer marked the beginning of frying on an industrial scale. Thus, fried foods like potato chips became popular among the masses across the globe. Nevertheless, it has been known that the nutritional value and quality attributes of lipids and fat-based foods are significantly affected by extreme processing conditions. Lipid oxidation is the key issue in this contest, as it is directly affects the quality of foods in terms of palatability and shelf life of food products (Wang et al., 2023). This chapter aims to discuss the origin and mechanisms of lipid oxidation, the influences of both intrinsic and extrinsic factors in this process, the impact of frying on lipid oxidation, and strategies adopted to control lipid oxidation.

2 Lipid oxidation

When unsaturated lipid molecules undergo a peroxidation reaction, a lipid hydroperoxide is formed as a primary oxidation product. As shown in Fig 6.1, lipid radical (L•) rapidly reacts with molecular oxygen, forming a lipid peroxy radical (LOO•). This radical then abstracts a hydrogen from another lipid molecule, generating a new lipid radical (L•) that continues the chain reaction and lipid hydroperoxide (LOOH), thus continuing the chain reaction. The process of lipid oxidation might be categorized into several subgroups, which include autoxidation, photo-oxidation, and enzyme-catalyzed oxidation, based on the mechanism of the reaction and causative factors (Abeyrathne et al., 2021; Ahmed et al., 2016; Wang et al., 2023). Among these different mechanisms, autoxidation is the most common phenomena proceeding via three distinct

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stages, namely initiation, propagation and termination. Generally, oils with high degree of unsaturation are particularly susceptible to lipid oxidation. Oils with high linoleic acid have been reported to be poor as frying oils since they rapidly deteriorate when subject to frying (Warner and Knowlton, 1997)

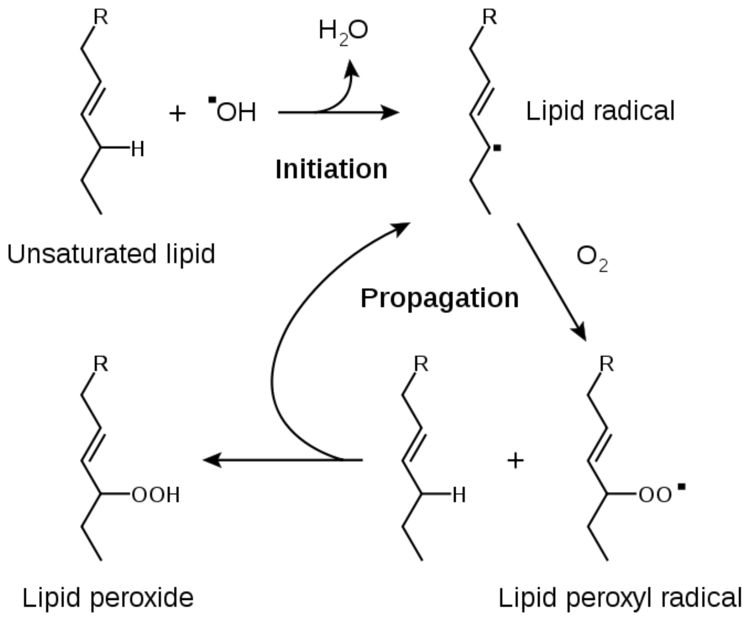

Figure 6.1: Mechanism of lipid peroxide formation

Lipid oxidation leads formation of a wide range of oxidation products through breakdown of lipid (Abeyrathne et al., 2021; Wang et al., 2023). The photooxidation mechanism produces a wide range of aliphatic and aromatic oxidized compounds, including fatty acids, aldehydes, ketones, carboxylic acids, alcohols esters, epoxies, phenols, sulfoxides, sulfones, anhydrides and quinones (Ahmed et al., 2016; Wang et al., 2023). In the enzyme-catalyzed oxidation, the process of oxidation proceeds through the involvement of enzymes, namely lipoxygenase and hydro-peroxidase (Wang et al., 2023).

The nature of the lipid oxidation is governed by several intrinsic and extrinsic factors. The triacylglycerol composition and minor constituent present in oils are the main intrinsic factors. Generally, food lipids fall under two major categories namely, saturated and unsaturated fats based on chemical compositions. Under the saturated fats, there are different sub-classes such as lauric oils (coconut kernel oil, virgin coconut oil, palm kernel oil, coconut testa oil), palmitic oils and stearic oils. Oils and fats under this category have a reduced tendency to undergo lipid oxidation. On the other hand, unsaturated oils are highly susceptible for lipid oxidation process (Ahmed et al., 2016; Jackson & Penumetcha, 2019). As shown in Table 6.1, there are two main sub-classes known as monounsaturated and polyunsaturated oils under this category. Generally, oleic oils such as olive oil, safflower oil, canola oil, peanut oil, lard fall under the monounsaturated oils while omega-3 and omega-6 fatty acid rich oils fall under the subclass of polyunsaturated oils. Walnut oil, sunflower oil, flaxseed oil and soybean oil are some of the well-known examples of omega-3 oils. Apart from these, oils that originate from fish and marine vertebrates might also fall under the category of polyunsaturated oils. Fish oils are generally known to possess little amounts of eicosapentaenoic (EPA) and docosahexaenoic (DHA) acids (Abeyrathne et al., 2021; Eshak et al., 2018; Secci & Parisi, 2016: Nizar et al., 2013).

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3 Effect of frying on lipid oxidation

Frying is one of the oldest food preparation techniques which might have considerable influence on the sensory attributes of foods (Boskou, 1988; Domínguez et al., 2014; Vieira et al., 2018). Many factors including moisture, atmospheric oxygen, storage temperature, light and food processing operations might influence lipid oxidation. The onset of lipid oxidation might start even at the beginning of the production and refining stages due to factors including thermal agitation. Food processing operations such as frying, roasting, and cooking are some of the major extrinsic factors that influence the lipid oxidation (Abeyrathne et al., 2021; Wang et al., 2023). According to the mechanistic explanation, oils act as the heat-transferring medium in the process of frying foods (Choe & Min, 2007). In frying, the type of frying oil, frying temperature and time, type of fryer, type of food, and antioxidants are determinant factors in lipid oxidation. The presence of free fatty acids in frying oil and their degree of unsaturation can significantly enhance the thermo-oxidative degradation of these oils (Choe & Min, 2007; Marikkar et al., 2007). The high frying temperature and intermittent heating and cooling might accelerate the deterioration of frying oil. Moreover, metal fryers especially those made of iron and/or copper can catalyze or accelerate the oxidation process. Interestingly, fryers with an optimum surface-to-volume ratio could decrease the rate of oxidation of frying oils (Boskou, 1988; Choe & Min, 2007). Antioxidants present in oil and frying medium might decrease or slow the oxidation process. However, the high temperature of the cooking medium can deplete the effectiveness of antioxidants (Choe & Min, 2007). During the deep-frying process, the formation of hydro-peroxides due to autoxidation of oils and fats is common. Peroxide value, which measures the concentration of hydro-peroxides in oils is used as a quality parameter to detect the preliminary stage of lipid oxidation (Marikkar et al., 2007). Other notable changes that occur in a frying oil are increases in viscosity and FFA content, decrease in iodine value, changes in refractive index, etc. Prolonged deep-frying, may also lead to the polymerization of fatty acids, which is generally known to increase the oil viscosity leading to the greasiness of fried products.

Table 6.1: *Composition of different oil types on lipid oxidation*

Oil type	Finding related to lipid oxidation	References
Coconut oil	As coconut oil is rich in SFA, heating period-dependent levels of aldehydes formed are lower.	Grootveld *et al*. (2020)
	Peroxide value increased significantly when oil was heated for 8 h (PV= 10.33), indicating hydro-peroxide formation during frying	Marikkar *et al*. (2007)
Virgin coconut oil	Peroxide value gets significantly increased when oil heated for 8 h (PV= 5.20), indicating hydro-peroxides formation during frying	Marikkar *et al*. (2007)
Olive oil	The concentration of acrolein formed during frying was 72.3 - 491.2 μg/kg	Vieira *et al*. (2017)
Canola oil	When heated at 180 °C for 24 h, the concentration of crotonaldehyde formed was 12.3 mg/kg	Vieira *et al*. (2017)
	As canola oil is rich in MUFA, heating period-dependent levels of aldehydes formed are intermediate	Grootveld *et al*. (2020)
Corn oil	As corn oil is rich in PUFA, heating period-dependent levels of aldehydes formed are high	Grootveld *et al*. (2020)

	Peroxide value gets significantly increased when the oil was heated for 8 h (PV= 10.98), indicating hydro-peroxide formation during frying	Marikkar *et al*. (2007)
Sunflower oil	As sunflower oil is rich in PUFA, heating period-dependent levels of aldehydes formed are high	Grootveld *et al*. (2020)
Palm oil	Highly stable during frying due to the presence of natural antioxidants, such as β-carotene and tocotrienol. The useful life as a frying medium is to be 12 days of continuous frying	Mba *et al*. (2015)
Soybean oil	Accelerate the oxidation process by the higher content of polyunsaturated fatty acids (PUFAs) present in soybean oil	Abdillah *et al*. (2020)

4 Nutritional implications of lipid oxidation

The double bonds in polyunsaturated and monounsaturated oils are known to get oxidized due to the oxidation process. As a result of lipid oxidation, nutritional value, sensory quality and safety of food are affected. While peroxides are produced as primary oxidation products, aldehydes, ketones, hydroxyl compounds, epoxides, oligomers, and polymers are formed as secondary oxidation products of lipid oxidation (Marikkar et al., 2007; Wang et al., 2023). The mechanism of formation of these products via photosensitized oxidation of lipids was reviewed recently by Bacellar and Baptista (2019). These products make the oils lose their quality attributes organoleptically due to rancidity development. Apart from the quality deterioration, these products lead to the risk of non-communicable diseases including cancers and cardiovascular diseases (Domínguez et al., 2014; Ahmed et al., 2016; Grootveld et al., 2020). When exposed to high-temperature frying practices at ca. 180° C, or stored at ambient temperatures for a prolonged period of time, polyunsaturated fatty acids (PUFAs) of corn, sunflower or soybean oils might tend to produce hydroperoxides. Owing to the effect of continuous thermal agitation, these hydroperoxides might undergo cleavage to produce lipid oxidation products (LOP) (Fig 6.2). The deleterious effect of lipid oxidation products (LOP) on human health has recently received much attention from various research groups (Grootvelt *et al.*, 2019). When absorbed from the gastrointestinal (GI) system into the systemic circulation, such LOPs may significantly contribute to enhanced chronic non-communicable disease (NCDs) risks. In recent times, Grootvelt *et al.* (2019) reviewed public health threats posed by the dietary ingestion of LOPs through fried foods.

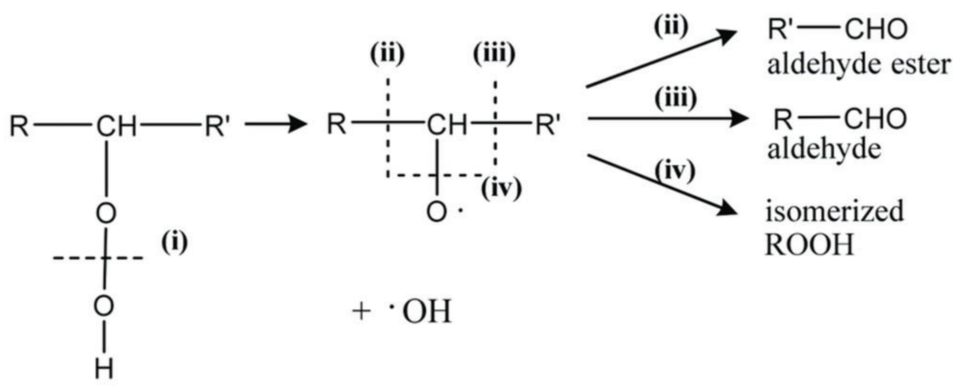

Figure 6.2: Cleavage of lipid hydroperoxide

Chiang et al. (2011) investigated the impact of consuming foods rich in oxidized frying oils on the development of type 2 diabetes. The study results showed that those foods are capable of depleting the secretion of insulin and causing glucose intolerance. This study attributed that the situation is caused by an oxidative damage-mediated alteration of glucose metabolism. Chemical changes in specific amino acids within the low-density lipoprotein (LDL), caused by various aldehydes, promote the uptake of LDL by macrophages, leading to the formation of foam cells. These foam cells contribute to the development of artery-blocking fatty streaks. A previous study by Staprans et al. (1996) showed that feeding rabbits with an oxidized lipid-rich diet resulted in a significant increase in fatty streak lesions in their aortas, along with elevated cholesterol levels in the pulmonary artery. Modified proteins resulting from these aldehydes have been detected in atherosclerotic lesions, and they are also associated with various diseases related to atherosclerosis, including cardiovascular disease and complications of diabetes (Grootveld et al., 2020). Reactive aldehydes have been shown to negatively affect various aspects of heart function, including mitochondrial respiration, ion-channel activity, and muscle contraction. Acrolein, in particular, has been found to worsen heart damage during ischemia and reduce the nitric oxide-induced protective effects in mice. Long-term exposure to acrolein at levels comparable to human intake of unsaturated aldehydes has also been linked to a dilated cardiomyopathy-like condition in mice, suggesting that exposure to acrolein and similar aldehydes through diet could potentially contribute to the development of heart failure in humans (Wang et al., 2008; Ishmail et al., 2011).

Srivastava *et al.* (2010) investigated the genotoxic and carcinogenic risks linked to the ingestion of repeatedly-boiled sunflower oil. This study found that its oral administration to Wistar rats resulted in a dose-dependent induction of aberrant cells and micronuclei. Based on both animal models and epidemiologic studies, Woutersen et al. (1999) concluded that the increasing intake of dietary fat exerted a significant influence on prostaglandin and leukotriene biosynthetic routes. Peroxidation of lipids can disturb the assembly of the membrane, causing changes in fluidity and permeability, alterations of ion transport and inhibition of metabolic processes. Products of lipid oxidation such as aldehydes act as potent toxins as they are extremely chemically reactive (Bacellar & Baptista, 2019; Grootveld et al., 2020). Higher concentrations of this reactive aldehyde are effective in potently suppressing a wide range of cellular processes, which leads to indiscriminate cellular damage and ultimately apoptosis (Grootveld et al., 2020). All these suggested that improving oxidative stability is crucial to preventing adverse effects of lipid oxidation.

Several studies have examined the relationship between fried food consumption and cancer risk. According to a study by Stott-Miller et al., (2013), males who consumed deep-fried foods frequently, such as fried chicken, French fries, fried fish, and doughnuts had a higher chance of prostate cancer. A meta-analysis of the published data revealed that greater intake of fried foods could increase the risk of prostate cancer by about 35%. These findings provide evidence for a link between fried food consumption and an elevated risk of prostate cancer. Knecht et al. (1994) found a relationship between fried meat consumption and female hormone-related cancers such as breast, endometrium and ovarian cancers in women. The studies by Bosetti et al. (2002) also found that a high consumption of fried potatoes, meat, fish and egg significantly would increase the risk of laryngeal cancers.

5 Control over lipid oxidation

A great deal of work has been dedicated to find out ways and means of controlling lipid oxidation. As shown in Table 6.2, some of the most effective ways of minimizing lipid oxidation would be to reducing the exposure to oxygen, decreasing the degree of unsaturation, using free radical scavenging antioxidants, incorporating singlet oxygen quenchers, blocking the light exposure, reducing storage temperature, and adding suitable metal chelators (Johnson & Decker, 2015). Among these strategies, application of antioxidants is one of the most effective methods to maintain control over oxidation. In addition to these, different physical methods are adopted by food industries to delay the onset of auto-oxidation of lipids during storage. This may include blocking light exposure, modified atmosphere packaging, vacuum packaging, nitrogen flushing etc. (Johnson & Decker, 2015).

Sunlight is said to accelerate lipid oxidation, leading to degradation of antioxidants and pigments present in oils and fats. An amber glass bottle is the best container to protect oils such as unsaturated oil as it is capable of blocking sunlight, particularly ultraviolet (UV) light. Amber glass is generally known to have the ability to block out UV rays by absorbing light wavelengths shorter than 450 nm. Singlet oxygen quenchers deactivate singlet oxygen by absorbing its excess energy and converting it to a stable form of oxygen.

Table 6.2: *Role of different factors influencing lipid oxidation*

Extrinsic/ intrinsic factor	Control measures	Reference
Presence of high proportions of unsaturated fatty acids	Adjust the fatty acid content (E.g.: Partial hydrogenation)	Choe & Min (2007); Abdillah et al. (2020)
	Genetic modification of plants to reduce the synthesis of PUFAs	Choe & Min (2007); Abdillah et al. (2020)
	Lipid interesterification to modifying the physical and chemical properties of oil by redistributing triacylglycerol (TAG) intermolecular and intramolecular fatty acids	Abdillah et al. (2020)
Exposure to atmospheric oxygen, moisture and light	Use of appropriate packaging E.g.: Encapsulation/microencapsulation	Shahidi & Zhong (2010)
Presence of trace metals	Use of metal chelators E.g.: citric acid, phosphoric acid	Shahidi & Zhong (2010)
Presence of oxidation promotors. E.g.: reactive oxygen species, oxidation catalytic enzymes	Use of antioxidants (either natural or synthetic)	Shahidi & Zhong (2010)
High storage temperature	Decrease the storage temperature	Johnson & Decker (2015); Liu et al. (2019)
Food processing operations. E.g.: Frying / deep frying	Use of fryer with a small surface-to-volume ratio	Choe & Min (2007)
	Avoid intermittent heating and cooling during frying	Choe & Min (2007)

This process involves both physical quenching, where the energy is released as heat, and chemical quenching, where singlet oxygen reacts with the quencher molecules to form hydroperoxides. The ability of these quenchers to quench singlet oxygen depends on the number of double bonds in the molecule. For instance, carotenoids, like b-carotene found in fruits and vegetables, with 11 double bonds can deactivate up to 1000 molecules of singlet oxygen (Fereidoon & Ying, 2010).

Controlling storage temperature is one of the most effective methods of preventing lipid oxidation in food products. Generally, storing lipids and lipid-derived products at lower temperatures reduce the rate of lipid oxidation. The shelf life of foods can be significantly extended under refrigeration and freezing conditions when compared to ambient temperature condition. Liu et al. (2019) studied the effect of storage temperature on lipid oxidation in peanuts and found that the degree of oxidation due to the heat accelerated and gets significantly increased with the storage temperature and time. In another instance, Jakobsen &

Bertelsen (2000) discovered that temperature is the most crucial factor for preserving red oxymyoglobin color and the degree of lipid oxidation of fresh beef meat. It is also found that the low temperature, below 4^o C almost prevents lipid oxidation, irrespective of oxygen concentration.

Antioxidants might deplete or prevent lipid oxidation via several mechanisms, including radical scavenging, singlet oxygen quenching, metal ion chelating, and inhibiting the per-oxidases (Wang et al., 2023). Metal ions such as iron and copper are known to act as catalysts of lipid oxidation reactions and generate free radicals via transferring electrons. The pro-oxidant effect of these metal ions can be reduced by the metal chelators, such as phosphoric acid, citric acid and ethylenediaminetetraacetic acid (EDTA). These metal chelators form stable complexes with the metal ions, thereby reduce their redox potential and deplete the pro-oxidant effect of metal ions (Fereidoon & Ying, 2010).

6 Toyyiban perspective

The term 'toyyiban' refers to the aspects of nutrition, cleanliness and safety of the food and drinks we consume. The implementation of 'toyyiban' concept in the food service or processing sector is meant to eliminate all harmful things against the human welfare so that they fall in compliance with the goals of the Islamic law. From a secular point of view, the toyyiban concept is in line with one of the 17 sustainable goals of development proclaimed by the United Nations Organization. Hence, establishing proper food safety protocols is one of the prerequisites for countries to fulfill the goals and compliances related to human welfare. As discussed previously, the formation of carcinogens in frying oils is an important food quality issue, which has created high concern among the producers and consumers. Particularly, a variety of physicochemical changes might occur during deep-frying in the frying oil leading to formation of various decomposition products. The oxidative deterioration of polyunsaturated fatty acids in particular is said to be responsible for major chemical changes, especially at elevated temperature. The initial oxidation products that get accumulated in triacylglycerols are hydroperoxides, which are said to be unstable substances. Owing to overexposure to high temperature, they might undergo cleavage to form a variety of secondary oxidation products. These secondary oxidation products, which include lipophilic aldehydes such as alkanals, alkenals, alkadienals, and hydroxylalkenals are said to be absorbed readily through the diet. Implementing cross-checks and control measures in the supply chain might help to curb abuses and malpractices prevailing in the frying industry. Methods of analysis based on HPLC and GLC have been developed so far could help to cross-check the emergence of toxic elements in the food supply chain.

The worst of the worst scenario is the case of the same oil being used repeatedly in frying operation until it becomes dark in appearance and extremely poor in quality. According to past studies, frying medium reaching the abused stage leads to several health risks including cancer. This happens frequently because of the lack of awareness among the food vendors regarding food quality and hygienic practices. In fact, the great majority of these vendors are largely self-starters without formal education or training in food processing or food quality and safety. Concern over the exposure of above-said compounds has led to regulatory initiatives by some countries limiting the toxin levels in foodstuffs. However, this aspect of food safety has been largely overlooked in several developing nations due to lack of awareness about the risk of exposure. As a part of the implementation of 'toyyiban' concept, it may be necessary to provide food science education and training for those who require it.

7 Conclusion

Lipid oxidation can be categorized into several subgroups such as autoxidation, photo-oxidation and enzyme-catalyzed oxidation. Among them, autoxidation is the most common mechanism which has three distinct stages (initiation, propagation and termination). The nature of lipid oxidation greatly influenced by both intrinsic and extrinsic factors. The composition of triacylglycerol and minor constituents in present oils are the main intrinsic factors while moisture, atmospheric oxygen, storage temperature, light and food processing operations such as frying, roasting and cooking are the major extrinsic factors. Particularly, the type of frying oil, frying temperature and duration, type of fryer, type of food and antioxidants play crucial roles in lipid oxidation during frying process. Lipids undergo oxidation, resulting in a diverse range of oxidation products including fatty acids, aldehydes, ketones, carboxylic acids, alcohols esters, epoxies, phenols, sulfoxides, sulfones, anhydrides and quinones. These products affect the nutritional value, sensory quality and safety of food. Moreover, these products pose a risk of non-communicable diseases, including cancers, cardiovascular diseases and diabetes. However, several approaches can be employed to minimize lipid oxidation. These include reducing the exposure to oxygen, decreasing/lowering the degree of unsaturation in oils, using free radical scavenging antioxidants, incorporating singlet oxygen quenchers, blocking light exposure, reducing storage temperature, and adding suitable metal chelators. One significant food quality is the formation of carcinogens in frying oils, which affects both producers and consumers. Implementing cross-checks and food analysis in the supply chain can help prevent abuses and malpractices in the frying industry. As the term of "toyyiban" refers to the safety and cleanliness of the food, it must be implemented to eliminate harmful elements of lipid oxidation in compliance with the Islamic law. The application of toyyiban concept would also help align with one of the United Nations' Sustainable Development Goals (SDGs), promoting responsible production and consumption.

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