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REVIEW

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Deep-fried flavor: characteristics, formation mechanisms, and influencing factors

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ABSTRACT

Deep-fried flavor, involving fatty, sweet, burnt, and grilled odors, is an important factor leading to the popularity of deep-fried foods. Comparing with flavors from other conventional and innovative thermal treatments, deep-fried flavor is characterized by a rich variety of volatile species (e.g. aldehydes, alcohols, ketones, hydrocarbons, carboxylic acids, furans, pyrazines, and pyridines), intricate formation mechanisms, and a stronger attraction to consumers. By means of comprehensively literature research, this article critically reviews deep-fried flavor deriving from lipid oxidation, Maillard reaction, hydrolysis and amino acid degradation, with a special emphasis to discuss the involvement of lipid oxidation products in the Maillard pathway to form fried volatiles via secondary processes (e.g. fragmentation, rearrangement, and degradation). The reactions are interacted and influenced by various factors, such as frying oils (e.g. fatty acid composition and oil type), food components (e.g. amino acid and sugar), frying conditions (e.g. oxygen concentration, frying time, temperature, pH, and moisture content), and frying types (e.g. vacuum frying and air frying). Overall, well understanding of chemistry origins of deep-fried volatiles is meaningful to economically manipulate the frying process, optimize the fried flavor, and improve the safety and consumer acceptance of deep-fried foods.

Background

Deep-fried foods are very popular in the household kitchens, restaurants, and catering industry, due to their palatable taste, acceptable cost, and fast and easy preparation steps. Deep-fat frying is an intricate process, in which foods are completely immersed into hot oils (150–190 °C) to trigger a simultaneous heat and mass transfer among oil, food, and air to result in food dehydration. Meanwhile, the food temperature is heterogeneous (the peripheral region is close to oil temperature; whereas the food core is around 100 °C) and increases slowly by limited oil penetration. As a result, a desirable deep-fried food with attractive flavor, golden color, and advantageous texture (the interior is juiciness, and the exterior/dehydrated surface is crispy) is prepared (Choe and Min 2007; Ngadi and Xue 2009). During deep-fat frying, mass transfer and lipid exchanges are major physical alterations contributing to the fried food quality (e.g. composition, texture, and palatability) (Dobarganes, Marquez-Ruiz, and Velasco 2000). Various chemical reactions (e.g. lipid oxidation, starch gelatinization, sugar dehydration, protein denaturation, polymerization, hydrolysis, and Maillard reaction) take place to produce volatile/non-volatile and soluble/non-soluble matters to darken food color, generate aroma, and develop unique texture, by which the deep-fried flavor is highly affected by lipid oxidation and Maillard reaction (Kalogeropoulos et al. 2007).

Flavor is a decisive sensory property to determine the prevalence of food products and provide an indication of physical/chemical alterations suffered within food materials. Volatile compounds are a basic element to decide flavor perception by reacting/binding with more than 400 rhodopsin-like G protein accompanied with seven transmembrane helix receptors, which translate external chemical stimuli into internal information to be read by nervous system and transmit signals to the brain that compares, classifies, and reflects flavor sensation (Stier 2000). It is assumed that the receptors have special coding strategies to cope volatiles and then yield valuable insight to encode the nature of volatile information to stimulate cerebral cortex (Schmuker et al. 2007). Although advanced technologies [e.g. gas chromatography–mass spectrometry (GC–MS), gas chromatography–olfactometry (GC–O), and descriptive sensory analysis] have been applied over last decades to comprehensively characterize the sensory acceptability of food on a molecular level, the interaction between odorants and their cognate receptor proteins and the mechanism behind olfactory decoding to transmit stimuli to cerebral cortex to generate preference response of food flavor are still mysteries (Dunkel et al. 2014). Therefore, elucidating the receptor codes to relevant odorants and olfactory signals perceived by the brain during food consumption will be an important milestone to understand the consumer acceptability of deep-fried flavor. But here is no doubt that deep-fried flavor is an

important factor contributing to the popularity of deep-fried foods.

Volatile constituents from animal fats (Chyau and Mau 2001; Tang et al. 1983), palm oil (Martin and Ames 2001; Wagner and Grosch 1998), soybean oil (Chyau and Mau 2001; Petersen et al. 2013), corn oil (Chun and Ho 1997; Macku and Shibamoto 1991), sunflower oil (Ramirez et al. 2004; Warner, Orr, and Glynn 1997), and rapeseed oil (Peng et al. 2017; Thurer and Granvogl 2016) during simulated deep-fat frying with/without foods have been extensively investigated, and over 200 compounds including acids, hydrocarbons, alcohols, aldehydes, ketones, pyrazines, and furans were detected (Table 1). However, the fundamental reactions (e.g. lipid oxidation and Maillard reaction) contributing to the development of desirable fried flavor is scarcely comprehensively summarized in the literature. Systematically elucidating formation mechanisms of deep-fried volatiles is instructive important to monitor frying process, examine perceived flavor, and evaluate the appreciation of fried food products. This article reviews the characteristics of deep-fried flavor and its formation mechanisms, and discusses the roles of frying media, food components, frying conditions, and frying types on the development of desirable deep-fried volatiles, therefore, to provide meaningful instructions to optimize frying process and promote the development of healthy and palatable deep-fried foods.

Characteristics of deep-fried flavor

Deep-fried flavor vs. flavors generated by other heat treatments

In general, thermal treatments (e.g. boiling, steaming, baking, roasting, and frying) are widely used to inactivate detrimental constituents (e.g. food-borne pathogens and natural toxins), prolong shelf-life, enhance digestibility and bioavailability of nutrients, and improve sensory properties (e.g. texture and flavor). Because of variances on the temperature, cooking time, and heat transfer medium, they prepare food products with distinguished different flavor profiles (van Boekel et al. 2010). In practice, a very milder flavor is usually achieved by boiling and steaming, in which water (as liquid or steam) is a heat transfer medium at relative low temperatures ($\sim$100 °C) to retain more moisture but reduce fat content of cooked products (Nieva-Echevarria et al. 2017). As dry heating methods, baking and roasting are performed at a temperature range of 150–250 °C on a small quantity of oil base to induce roasted, burnt, and caramel aromas (Jousse et al. 2002). During frying, oil/fat is an excellent heat transfer medium and plays an essential sensory role to carry, enhance, and promote oily, fatty, and nutty flavors (Shabbir et al. 2015). Take potatoes as an example, lipoxygenase activated oxidation is a major source of volatiles (e.g. 2,6-nonadienal) for raw potatoes (Josephson and Lindsay 1987). Boiled potatoes present a relative blank and stale flavor (e.g. 2,6-nonadienal and 4-heptenal) by the degradation and inactivation of enzymes during boiling (Petersen, Poll, and Larsen 1998). The volatiles (e.g. methoxy-alklpyrazines, methional, and 2,4-decadienal) of

baked potatoes are mainly from Maillard reaction (Oruna-Concha, Bakker, and Ames 2002). However, the flavor formation in the fried potatoes is very complicated, which not only from chemical reactions (e.g. lipid oxidation and Maillard reaction) within potatoes and frying media, but also principally driving from complex interactions between reactive intermediates (Whitfield and Mottram 1992; Martin and Ames 2001). For example, saturated/unsaturated aldehydes (lipid oxidation products) can react with hydrogen sulfide (a Strecker degradation product of amino acids) to produce oily and roasted odorants [e.g. 3-(methylthio)propanal, 2-ethyl-3,5-dimethylpyrazine, and 2-nonenal] (Pokorny 1981; Whitfield and Mottram 1992).

Recently, different thermal treatments are analyzed to exhibit distinct notes contributing to food flavors. Nieva-Echevarria et al. (2017) announced steaming promoted stronger lipid hydrolysis and oxidation to stimulate more volatile release (e.g. 1-penten-3-ol, propanal, hexanal, 2-butanone, and benzaldehyde) from sea bass meat than boiling. Utama et al. (2018) compared roasting and boiling to cook Hanwoo brisket, and found roasting was a more efficient way to facilitate the release of aroma with abundant aldehydes (e.g. 2-methylbutanal, pentanal, and heptanal) and pyrazines (e.g. 2,5-dimethylpyrazine), because of severe hydroperoxides degradation and inosinic acid loss. Shi et al. (2017) claimed flavor profiles (e.g. roasted, beany, woody, and bitterness odors) of deep-fried peanuts were similar as roasted peanuts, but the roasted odor of fried peanuts lasted longer during storage by a great exposure of inner parenchyma cells during frying. Yang and coworkers (Yang et al. 2017) estimated the effect of baking and deep-frying on the formation of aroma compounds in pork loins, and noticed that a number of significant pyrazines (e.g. 2,5-dimethylpyrazine, 2,6-dimethylpyrazine, 2-methyl-3-propylpyrazine) that expressed roasted and nutty odors were missing in the baked pork loin as compared with deep-fried pork loin. Nevertheless, because of intensive pyrolysis of proteins and Maillard reaction, heterocyclic amines (carcinogenic compounds) in the baked samples were much higher than the deep-fried ones.

Types of volatiles impacting deep-fried flavor

Aldehydes

Aldehydes, including saturated (alkanals) and unsaturated (alkenals and alkadienals) ones, are the most common volatiles developed by the frying process through lipid oxidation to exhibit oily, green, paint, metallic, and beany flavors. In general, thermal degradation and hydrolysis of triacylglycerols easily occur under frying conditions to produce free fatty acids, which then generate different types of hydroperoxides (e.g. 8-, 9-, 10-, 11-, 12-, and 13-hydroperoxide) according to the reaction sites on carbon-carbon double bonds via β-homolysis cleavage, keto-enol tautomerism, or isomerization (Yin, Xu, and Porter 2011). Subsequently, each hydroperoxide can be decomposed to volatiles (e.g. aldehydes, hydrocarbons, ketones, alcohols, and carboxylic acids) with low molecular mass (Zhang et al. 2015). So, the increase of

Table 1. Study examples of deep-fried volatiles under simulated frying conditions.

Lipid resources	Food resources	Frying conditions	Chemical reactions	Volatiles	References
Propanal	Glycine	Fried at 180 °C	Maillard reaction	3,5-Dimethyl-2-ethylpyridine, 3,5-dimethyl-4-ethylpyridine, and 3,5-dimethylpyridine	Suyama and Adachi (1980)
Beef fat (90%) with cottonseed oil (10%)	Chicken meat	Fried at 185 °C for 8 min	Maillard reaction Lipid oxidation Strecker degradation	2-Methylhexadecane, 2-methylpyrazine, pentanol, 3-ethylpyridine, 2-methyl-4-ethylthiazole, hexanal, 2-butanone, 2-buthylfuran, pentanoic acid, 4,5-dimethyloxazole, 2-methylthiophene, and 2-methylpyrrole	Tang et al. (1983)
Fri-al shortening	Potato slices	Fried at 190 °C for 1.5–3.5 min	Maillard reaction Lipid oxidation	2,5-dimethyl-4-butyloxazole, 2-methyl-4-pentyloxazole, 2-pentyl-4-methyloxazole, and 2-butyl-4-propyl-5-methyloxazole	Carlin et al. (1986)
Corn oil	Zein + corn starch	Fried at 180 °C for 30 min	Maillard reaction Carotenoid decomposition Lipid oxidation	2-Methyl-3-pentylpyrazine, 2,5-dimethyl-3-pentylpyrazine, toluene, isophorone, 2-methylpentanal, and 2-methylbutanal	Huang et al. (1987)
2,4-Decadienal	Cysteine Glutathione	Fried at 180 °C for 1 h at pH 7.5	Maillard reaction	2-Hexylthiophen, 2-butylthiophene, 3,5-dimethyl-1,2,4-trithiolane, and 3,5-dimethyl-1,2,4-trithiolane	Zhang and Ho (1989)
Corn oil	Cysteine	Fried at 180 °C for 4 h	Maillard reaction Lipid oxidation	1-Pentanol, heptane, 2-heptanone, 2-methylthiazolidine, octane, 1-ethylpyrrole, 2-pentylfuran, and butylbenzene	Macku and Shibamoto (1991)
Corn oil	Glutamine/ asparagine + glucose	Fried at 180–183 °C for 1 min at pH 8	Maillard reaction Lipid oxidation	Ethylpyrazine, trimethylpyrazine, 2-acetyl-6-methylpyrazine, cyclopentapyrazine, 3-butylpyridine, and 2-pentylpyridine	Chun and Ho (1997)
Cottonseed oil High-oleic sunflower oil	Potato chips	Fried at 187–192 °C for 130 s	Lipid oxidation	Hexanal, pentanal, octanal, nonanal, and 2,4-decadienal	Warner, Orr, and Glynn (1997)
Palm oil Coconut fat	French fries	Fried at 170 °C for 3 min	Lipid oxidation	2-Ethyl-3,5-dimethylpyrazine, 3-ethyl-2,5-dimethylpyrazine, 2,3-diethyl-5-methylpyrazine, 2,4-decadienal, methylpropanal, 2-methylbutanal, 3-methylbutanal, and methanethiol	Wagner and Grosch (1998)
Soybean oil Corn oil Lard	Shallot	Fried at 150–160 °C for 5 min	Amino acid degradation Lipid oxidation	2-Hexenal, 2-pentylfuran, 2-heptenal, nonanal, 2-octenal, 1-octen-3-ol, 1-heptanol, 2,4-heptadienal, 2-decenal, 2-undecenal, and 2,4-decadienal	Chyau and Mau (2001)
Palm olein	Potato slices	Fried at 180 °C for 2.5 min	Strecker degradation Lipoxygenase-mediated oxidation	Methylpropanal, 3-methylbutanal, 2-methylbutanal, penylacetaldehyde, dimethyl disulfide, methylpyrazine, 2-ethyl-5(3)-methylpyrazine, 3-ethyl-2,5-dimethylpyrazine, hexanal, and 2,4-decadienal	Martin and Ames (2001)
Olive oil Sunflower oil Pig lard Butter	Pork loin chops	Fried at 160 °C for 2 min, and turned over after 1 min of frying	Lipid oxidation Strecker degradation Maillard reaction	Hexanal, pentanal, benzaldehyde, decanal, heptan-2-one, nonan-2-one, 2-methylbutanal, and 3-(methylthio)propanal	Ramirez et al. (2004)
Hydrogenated vegetable oil	French fries	Fried at 180 °C for 4 min	Sugar degradation Maillard reaction Lipid oxidation	3-Methylbutanal, hexanal, 2-methylpropanal, pyridine, heptanal, phenylacetaldehyde, 2,5-diethylpyrazine, 2,4-heptadienal, and 5-ethyl-2,3-dimethylpyrazine	van Loon et al. (2005)
Rapeseed oil Safflower oil Perilla oil	Sliced potato	Fried at 150–185 °C for 3 min over 8 h continuous frying	Lipid degradation Maillard reaction	Propanal, acrolein, 2-heptenal, 2,4-heptadienal, nonanal, and 2,4-decadienal	Katsuta et al. (2008)
Sunflower oil Rapeseed oil Palm olein	French fries	Fried at 170 °C for 4 min, and the oil was kept at this temperature for 12 h on 3 days	Lipid oxidation	Nonanal, 2,4-decadienal, 2,4-heptadienal, 1-octen-3-ol, 2-decenal, hexanal, 2-octenal, 3-octanone, and 2-decanone,	Petersen et al. (2013)
Soybean oil	Wheat dough Chicken breast meat	Wheat dough and chicken breast were fried at 180 °C for 5	Lipid oxidation Hydrolysis Isomerization	Hexanal, nonanal, 2-pentenal, 2-heptenal, 2,4-heptadienal, 2,4-decadienal, benzaldehyde, 3-	Zhang et al. (2015); Zhang et al. (2018)

(continued)

Table 1. Continued.

Lipid resources	Food resources	Frying conditions	Chemical reactions	Volatiles	References
		and 10 min, respectively; the frying was performed for 8 h/day over 7 days	Strecker degradation Maillard reaction	methylbutanal, 4-oxononanal, 3-nonen-2-one, 2-propylpyridine, 1-octen-3-ol, 3-dodecene, and 2-pentylfuran	
Coconut oil Olive oil Rapeseed oil Safflower oil Linseed oil	Potato chips	Fried at 180°C for 2.5 min	Lipid oxidation Sugar degradation Maillard reaction	2,4-Decadienal, 2,3-diethyl-5-methylpyrazine, 2-ethyl-3,5-dimethylpyrazine, 2-ethyl-3,6-dimethylpyrazine, 2-methylbutanal, 3-methylbutanal, and 3-(methylthio)propanal	Thurer and Granvogl (2016)
–	Chinese chestnuts	Fried at 240°C for 15 min	Maillard reaction Amino acid degradation Saccharides degradation	Hexanal, furfural, 3-methyl-1-butanol acetate, 2-hydroxy-2-cyclopenten-1-one, 4-hydroxy-2-butanone, 6-metyl-5-hepten-2-one, octanal, nonanal, and decanal	Li et al. (2016)
Combinations of high-oleic sunflower oil, high-oleic rapeseed oil, palm mid fraction, and hydrogenated rapeseed oil	Donut	Fried at 170°C for 1.5 min on each side, and the frying media was heated for 8 h/day over 10 days	Lipid oxidation Lipid degradation	Hexanal, heptanal, octanal, 2-octenal, nonanal, decanal, 2-decenal, 2,4-decadienal, and 2-undecenal	Merkle et al. (2016)
Olive pomace oil Palm oil	Potato chips	Fried at 180°C for 9 min	Lipid oxidation Lipid degradation	Heptane, octane, hexanal, nonanal, 2-heptenal, pentanol, 1-octen-3-ol, 1-hexanoic acid, 2-penthylfuran, 2-heptanone, and 2-octanone	ben Hammouda et al. (2017)
Extra-virgin olive oil Peanut oil Canola oil	Potato chips	Fried at 175°C for 6 min with an intermittent frying frequency of 30 min for 8 h	Lipid oxidation	Heptane, toluene, dimethylbenzene, toluene, 1-pentanol, 2-pentylfuran, hexanal, octanal, nonanal, decanal, 2-heptenal, 2-decenal, 2-undecenal, 2,4-decadienal, and 2,4-heptadienal	Molina-Garcia et al. (2017)
Palm oil Rapeseed oil Sunflower oil Soybean oil	Potato strips Pork loin strips	Fried for 10 min	Lipid oxidation	Formaldehyde, acetaldehyde, acrolein, propanal, 2-butenal, butanal, pentanal, hexanal, 2-heptenal, 2,4-nonadienal, 2-nonenal, 2,4-decadienal, and nonanal	Peng et al. (2017)
Corn oil	Pork loin	Fried at 180°C for 5 min	Maillard reaction Lipid oxidation Vitamin degradation	Hexanal, nonanal, benzaldehyde, 2-pentylfuran, 2,5-dimethylpyrazine, 2,6-dimethylpyrazine, 6-methyl-2-ethylpyrazine, trimethylpyrazine, and 2-methyl-3-propylpyrazine	Yang et al. (2017)

aldehydes is in accordance with the oxidation of unsaturated fatty acids in frying oils and the degradation of their oxidative intermediates. The formation of alkanals is positively associated with the content of oleic acid, whereas alkenals are correlated with the abundance of linoleic acid and linolenic acids (Perkins 2007). Thus, it is proposed to consider aldehydes as a suitable marker to detect oxidative degradation to alternate common methods (e.g. total polar compounds, anisidine value, and peroxide value) (Molina-Garcia et al. 2017). However, the production of aldehydes could be depressed by the complex reactions happened during deep-fat frying. Zhang and coworkers (Zhang et al. 2015) claimed (E)-2-hexenal and (E,E)-2,4-decadienal were increased under wheat dough frying, but were consumed by more chemical reactions (e.g. further oxidation, degradation, and Maillard reaction) as chicken breast meat was fried. In practice, 3-(methylthio)propanal (cooked potato-like smelling), 2- and 3-methylbutanal (malty smelling) contribute to the deep-fried flavor in more than 50% of fried products, followed by (E,E)-2,4-decadienal (fatty smelling), hexanal (green and grassy smelling), 1-octene-3-one (mushroom-like smelling), acetic acid (vinegar-like smelling), acetaldehyde (fresh fruity smelling), (E)-2-nonenal (cardboard-like smelling), and 2-acetyl-1-pyrroline (roasty smelling) (Dunkel et al. 2014).

Hydrocarbons, ketones, alcohols, and carboxylic acids

In practice, hydrocarbons, ketones, alcohols, and carboxylic acids are formed in small quantities during deep-fat frying (Molina-Garcia et al. 2017). Saturated hydrocarbons (alkanes, including straight-chain, side-chain, and cycloalkanes) and unsaturated hydrocarbons (alkenes, including straight-chain monoenes, substituted monoenes, straight-chain dienes, substituted dienes, and cyclic monoenes, cyclic dienes, and configurational isomers) are presented in the volatile fraction during deep-fat frying via oxidative decomposition of unsaturated fatty acids, free radical reactions (e.g. cyclization and combination), and thermal decarboxylation of saturated fatty acids (Chung, Eiserich, and Shibamoto 1993; van der Klis et al. 2011). Similar as aldehydes, not all hydrocarbons elevate in concentrations with increasing frying time. For example, tetradecane and pentadecane were decreased with the time prolongation in the later stage of deep-fat frying, due to the complex reactions occurring (Zhang et al. 2018). A great evaporation of alkanes could also be happened after frying since their boiling points closing to frying temperatures (Uriarte and Guillen 2010). Cyclic alkenes (e.g. cyclohexenes and cyclooctenes) were discontinuously detected during deep-fat frying process by their thermal instability (Nawar 1998).

Thermal peroxidation of saturated fatty acids, keto-enol tautomerization of hydroperoxides, further oxidation of hydrocarbons, decomposition and intramolecular electron rearrangement of peroxides from unsaturated fatty acids contribute to the production of ketones (e.g. methyl ketones and cyclic ketones) (Chang, Peterson, and Ho 1978; Yin, Xu, and Porter 2011). The addition of food components (e.g. amino acids and sugars) possibly impacts the profile of volatiles by reducing the quantities and concentrations of ketones during deep-fat frying. Scientifically, the oxidative degradation of oils yields the products with hydroperoxyl, peroxyl, carbonyl, and hydroxyl groups that tend to react with amino and amide-containing compounds to induce covalent bonding complexes (Pokorny and Kolakowska 2011). For example, 3-nonen-2-one and 1(3H)-isobenzofuranone were dramatically decreased and even undetectable during the soybean oil frying with wheat dough and chicken breast meat as compared with the heating of soybean oil (Zhang et al. 2018).

Alcohols (including saturated, unsaturated, and cyclic ones) are formed through oxidative decomposition of unsaturated fatty acids and positional isomerization of double bonds on carbon chains (Selke, Rohwedder, and Dutton 1977; Zhang et al. 2018), in which 1-octen-3-ol (a mushroom-like odor) from the decomposition of linoleic acid-10-hydroperoxide is a most reported alcohol formed by deep-fat frying (Asaaf, Hadar, and Dosoretz 1997). Since food components can either promote the oxidation of unsaturated fatty acids in the oils or result in the intensive boiling to promote loss of volatiles, alcohols may be elevated or reduced during deep-fat frying. Moreover, carboxylic acids can be resulted from lipid oxidation and secondary decomposition of lipid oxidative products (e.g. hexanal and 2,4-decadienal) (Molina-Garcia et al. 2017). Even though carboxylic acids are presented before and during all sessions of deep-fat frying, they provide little contribution to the deep-fried flavor (Dominguez et al. 2014). In the work done by ben Hammouda and coworkers (2017), most of carboxylic acids remained consistent in concentrations, except 1-hexanoic acid, which has fatty odor and is originated from the secondary degradation of hexanal and 2,4-decadienal (Molina-Gracia et al. 2017).

Furans, pyrazines, and pyridines

Furans, pyrazines, and pyridines are well known as typical aroma sources presented at trace amounts in the frying oil and fried food products. The appearances and concentrations of these compounds are mainly related to frying time, temperature, and ingredients complexity of foods (especially amino acids and reducing sugars). Furans are induced from multiple routes during deep-fat frying, such as thermal degradation of carbohydrates, Maillard reaction between amino acids and reducing sugars, and lipid oxidation (ben Hammouda et al. 2017; Zhang et al. 2018). They usually provide deep-fried foods with desirable aroma described as sweet, burnt, and caramel-like odors (Van Ba et al. 2012). 2-Pentylfuran is the most commonly studied volatile furan, which is generated by methyl linoleate hydroperoxide degradation from linoleic acid oxidation (Vichi et al. 2003). So, 2-pentylfuran concentration is particularly determined by the amount of linoleic acid in the oils. Notably, pyrazines are structure-odor active volatiles that perform high olfactory impact at very low concentrations. 2-Ethyl-3,5-dimethylpyrazine, 2-ethenyl-3,5-dimethylyrazine, and 2,3-diethyl-5-methylpyrazine exhibit baked potato-like smelling with the lowest human odor thresholds of 0.007–0.018 μg/kg (Wagner et al, 1999). In terms of pyridines, Strecker degradation during the Maillard reaction

between amino acids and reducing sugars or lipid oxidation products attributes to their production under frying conditions. Illustrating in an example of alkylpyridines, in which the dehydration condensation reaction between 2,4-alkadienals and amino compounds results in unstable Schiff bases (aldimines) to form alkylpyridines via intramolecular cyclization and homolysis under high temperatures (Adams et al. 2011).

Food components have significant influences on the formation of volatiles during deep-fat frying. They generally increase the intensity and complexity of reactions to produce more volatiles under the conditions of high temperature and occurrence of oxygen, but further hydroperoxidation, Maillard reaction, and thermal degradation could take place between food components and production intermediates to expedite the release of unstable volatiles. Therefore, elucidating volatile compositions is technically challenging but instructive importance to evaluate the sensory quality of deep-fried products.

Odor impacts of deep-fried volatiles

Although the olfactory receptors are exposed to ten thousand of flavor compounds in the environment, flavor perception relies on odor thresholds of volatiles, and only the volatiles at concentrations above their odor thresholds can activate the olfactory system (Table 2) (Dunkel et al. 2014). Whetstine, Cadwallader, and Drake (2005) determined phenylacetaldehyde (a threshold of 0.002 mg/kg in water) at 3.7 mg/kg was more predominant than 2-phenethanol and phenylacetic acid that were near or below threshold levels to be responsible for rosy/floral flavor in the cheddar cheese. It is noticed that unsaturated aldehydes (e.g. 2,4-decadienal) with low odor thresholds provide strong deep-fried flavor at relative low concentrations. On the contrary, alcohols and hydrocarbons with high odor thresholds are not considered as important contributors to the deep-fried flavor (Ho and Chen 1994). In fact, compounds within a same chemical family even perform different odor intensities. Pyrazines exhibit baked cereal odor, but ethylpyrazines (with low odor thresholds) are considered as a more important flavor contributor than methylpyrazines (with high odor thresholds) (Jousse et al. 2002; Reineccius 1990). In fact, threshold values can be tremendously varied depending upon experimental format (e.g. ASTM methodology), number of panelists, detection media (e.g. air, water, and buffer), and definitions [detection threshold (the lowest value to most sensitive subject) vs. recognition threshold (the highest value to most insensitive subject)] (Meilgaard, Civille, and Carr 1999).

In addition, odorant threshold is determined based on the olfactory screening of a single volatile molecule, and is not respective to food matrix. Therefore, odor activity value (OAV, a ratio of volatile concentration in the food to its odorant threshold in a media) is universally applied for the judgment of odor sensitivity and the contribution of individual odorant to a given food aroma. It is expected that the volatile with higher OAV (at least > 1) is a more important contributor to the corresponding flavor (Wagner and Grosch 1998). Buttery and Ling (1972) concluded the most important odorant was methional (with the highest OAV) in the fried potato flavor, followed by phenylacetaldehyde, 2- and 3-methylbutanal, 3-ethyl-2,5-dimethyl-pyrazine, 2,4-decadienal, 1-penten-3-one, and hexanal (with the lowest OAV). Nevertheless, the absence of methional in the imitated flavor of French fries was not detected by sensory panels, due to the flavor masking effect by other odorants (e.g. 2-ethyl-3,5-dimethylpyrazine, 3-ethyl-2,5-dimehylpyrazine, and 2-methylbutanal) (Wagner and Grosch 1998). Indeed, the relationship between flavor perception by consumers and volatile chemicals are complicated and also dependent on some minor important parameters (e.g. chemical stability, partition coefficient, volatility, and molecular weight) rather than threshold value and OAV (Jousse et al. 2002). For instance, molecular weights of volatiles are negatively correlated with the sense of odor, in which formaldehyde provides a strong sting and pungency odor, but aldehydes with ≥ 7 carbon atoms behave lower odor intensities (Stier 2000). Therefore, it is essential to comprehensively consider various elements (e.g. OAV, threshold value, concentration, and molecular weight) to define the sensation efficiency of volatiles.

Formation of deep-fried flavor

Lipid oxidation and Maillard reaction are two most predominant chemical reactions contributing to the development of deep-fried flavor, which are determined by extraordinary complex volatiles in various quantities as discussed in last section. As shown in Fig. 1, both reactions follow similar reaction procedures with the production of key intermediates (e.g. hydroperoxides in lipid oxidation, and Amadori rearrangement products in Maillard reaction) to be later decomposed, degraded, cyclized, and rearranged to generate volatile compounds responsible for food flavor. Apart from the evaluation of individual reaction, due to the formation of chemical compounds with similar functional groups, more studies have investigated the interaction between lipid oxidation (or intermediates/products) and Maillard reaction (or intermediates/products), particularly on the participation of lipid oxidation products in Maillard reaction to create certain volatiles (e.g. pyrazines, pyridines, and sulfur-containing compounds) responsible for deep-fried flavor. In addition, hydrolysis of triacylglycerols and thermal degradation of amino acids play a minor important role to facilitate or assist the formation of volatiles during deep-fat frying.

Lipid oxidation

In the deep-fat frying operations, water molecules in the fried food evaporates as a steam and leave the empty space within the food structure, then the pressure difference between the outer circumstance and inner structure of the fried food results in the oil absorption in the fried product when it is removed from the frying oil. (Mehta and Swinburn 2001). So, the volatile molecules in the frying oils

Table 2. Examples of common volatile compounds responsible for desirable deep-fried flavor.

| Volatile compounds	Odor description	Precursors	Formation mechanism	Odor threshold (mg/m³, in air)	References
Hexanal	Fatty, oily, and grassy	Linoleic acid	Breakdown of 13-diene hydroperoxide that is from the oxidation of linoleic acid	0.23	Laska and Ringh (2010)
Nonanal	Fatty, paint, and soapy	Oleic acid	β-Homolysis of 10-hydroperoxide from oleic acid oxidation	0.0026	Yang et al. (2008)
Pentanal	Woody, bitter, and oily	Linoleic acid	Breakdown of 13-hydroperoxy-9,11-octadecadienoic acid from linoleic acid degradation	0.85	Laska and Ringh (2010)
Benzaldehyde	Sweet and vanilla-like	Linoleic acid	Oxidation and hydrolysis of linoleic acid under heating and occurrence of oxygen	0.085	Yang et al. (2008)
2-Methylpropanal	Malty	Valine	Strecker degradation of valine or Maillard reaction between valine and reducing sugars	0.001	Nagata (2003)
(E)-2-Decenal	Paint, fatty, and fishy	Oleic acid	β-Homolysis of 9-hydroperoxide from oleic acid oxidation	0.0027	Yang et al. (2008)
(E)-2-Nonenal	Fatty and cardboard-like	Linoleic acid	Isomerization of 3-nonenal, which is a combination product of 1,3-nonadienyl radical (decomposed from 9-hydroperoxide) and hydroxyl radical (from linoleic acid oxidation)	0.00009	Yang et al. (2008)
(E)-2-Heptenal	Pungent, tallowy, and oxidized	Linoleic acid	β-Homolysis of 12-hydroperoxide from the oxidation of linoleic acid	2.8	von Ranson and Belitz, (1992)
(E)-2-Octenal	Sweet and oily	Linoleic acid	Oxidative decomposition of 3-nonenal that is formed by the β-homolysis of 9-hydroperoxide from linoleic acid oxidation	0.0027	Yang et al. (2008)
(E)-2-Hexenal	Lawn and almond-like	Linolenic acid	Isomerization and electronic rearrangement of 3-hexenal, which is formed by keto-enol tautomerism of the combination product between 1,3-hexadienyl radicals degraded from the oxidation of linolenic acid (to produce 12-hydroperoxide and hydroxyl radical)	0.0031	Yang et al. (2008)
Undecenal	Oxidized, and paint	Oleic acid	β-Homolysis of 8-hydroperoxide from oleic acid oxidation	0.14	von Ranson and Belitz, (1992)
2-Methylbutanal	Malty	Isoleucine	Strecker degradation of isoleucine	0.1	Bartschat and Mosandl (1998)
3-Methylbutanal	Roasted	Leucine / Linolenic acid	Nonenzymatic Strecker degradation of leucine / Oxidative decomposition of linolenic acid	0.00035	Nagata (2003)
(E,E)-2,4-Decadienal	Deep-fried and fatty	Linoleic acid	Cleavage of 9-diene hydroperoxide that is from the oxidation of linoleic acid	0.0023	Yang et al. (2008)
(E,E)-2,4-Heptadienal	Fatty, nutty, and rancid	Linolenic acid	β-Homolysis of 12-hydroperoxide from linolenic acid oxidation	0.057	Hall and Andersson (1983)
(E,E)-2,4-Nonadienal	Fatty	Linolenic acid	Peroxidation of 3-nonenal generated from the decomposition of 9-hydroperoxide that is a product of linolenic acid oxidation	0.0002	Yang et al. (2008)
4,5-Epoxy-(E)-2-decenal	Crumb flavor	2,4-Decadienal	Oxidation of 2,4-decadienal	0.0000006-0.0000025	Schieberle and Buettner (2001)
1-Octen-3-ol	Mushroom-like	Oleic acid	Cleavage from methyl oleate hydroperoxides that is generated by the oxidation of oleic acid	0.0027	Yang et al. (2008)
1-Octen-3-one	Metallic and mushroom-like	Arachidonic acid	β-Scission reaction of 12-hydroperoxide from the oxidation of arachidonic acid	0.00003-0.00012	Schieberle and Buettner (2001)
2,5-Dimethylpyrazine	Roasted and grilled	Asparagine	Strecker degradation of asparagine	1.82	Wagner et al. (1999)
2,3-Dimethylpyrazine	–	Glutamine	Strecker degradation of glutamine	0.88	Wagner et al. (1999)
2,3-Diethyl-5-methylpyrazine	Earthy	–	A condensation between two aminoketones, two aminoaldehydes, or an aminoketone and an aminoaldehyde, followed by the addition of acetaldehyde to the corresponding dihydropyrazines as intermediates	0.00001-0.000014	Wagner et al. (1999)
2-Pentylpyridine	Grassy aroma	2,4-Decadienal	Maillard reaction between ammonia and 2,4-decadienal	0.0002	Schieberle (1993)
Methional	Potato-like	Methionine	Degradation of methionine	0.06	Gijs et al. (2000)
2-Pentyl-4-methyl-5-ethyloxazole	Sweet and lactone-like	–	Strecker degradation of amino ketones that are from the condensation of α-dicarbonyl compounds with amino acids	–	Carlin et al. (1986)
2-Pentylfuran	Sweet, burnt, and caramel-like	Linoleic acid	Cyclization of vinyl hydroperoxide (generated from the cleavage of 9-hydroxyradical of linoleic acid) via alkoxy radicals	0.019	Yang et al. (2008)
2-Ethylfuran	Sweet and rancid	2-Hexenal	Oxidation of 2-hexenal at allylic position followed by cyclization	–	ben Hammouda et al. (2017)
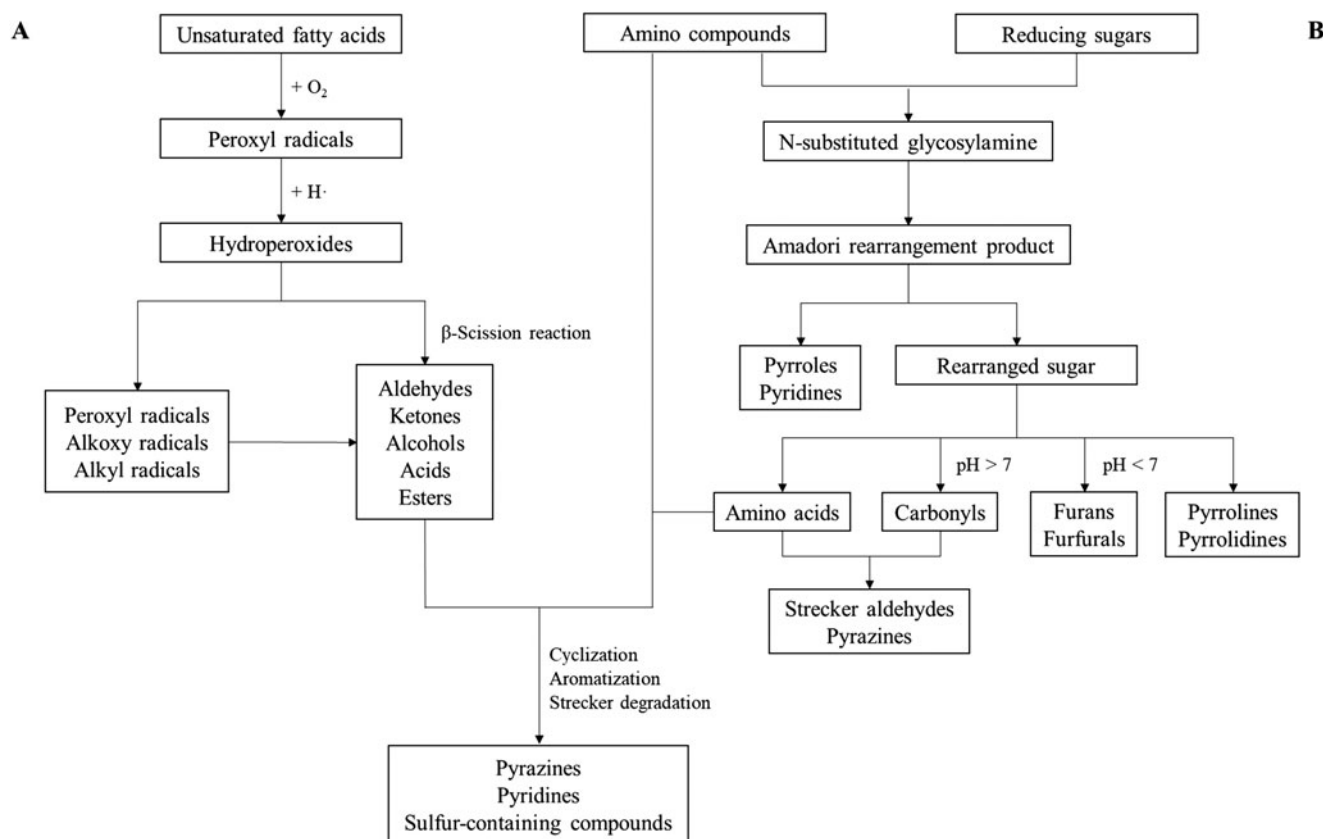
Figure 1. Lipid oxidation (A) and Maillard reaction (B) pathways in the development of deep-fried volatiles.

rising from lipid oxidation directly influence the flavor of fried foods by the oil uptake. Although volatile species in the frying media present at low concentrations (parts per million levels) and might be evaporated (with steam or smoke) or decomposed (by the reaction with other volatile compounds or food components), they are still extremely important to the odor of deep-fried foods (Nawar 1985). It is well known lipid oxidation involves three steps: initiation, propagation, and termination. During initiation, the abstraction of hydrogen from unsaturated fatty acids (e.g. linoleic acid) produces the alkyl radical, on which the free radical is delocalized on the carbon chain, and upon molecular rearrangement, to form conjugated double bonds (McClements and Decker 2007). The weakest carbon-hydrogen (C–H) bond is firstly attacked to release a free radical. For instance, the hydrogen on carbon 11 is removed to form a radical at carbon 11 on linoleic acid (Fig. 2). Thus, the strength of C–H bond explains the oxidation rate of various fatty acids, at which stearic acid (the weakest C–H bond: 100 kcal/mol) < oleic acid (the weakest C–H bond: 75 kcal/mol) < linoleic acid (the weakest C–H bond: 50 kcal/mol) < linolenic acid (the weakest C–H bond: 25 kcal/mol) (Min and Boff 2002).

Subsequently, the alkyl radical (a reduction potential of 600 mV) rapidly reacts with oxygen (at triplet state) to produce a peroxyl radical (a reduction potential of 1000 mV), which has high energy to promote the abstraction of hydrogen from another unsaturated fatty acids. Therefore, the addition of hydrogen on the peroxyl radical produces hydroperoxides, and this step is known as propagation (Fig. 2). In

theory, the hydrogen abstraction on linolenic acid (at C-11 and C-14) is 20-30 times faster than linoleic acid (at C-11), to generate four conjugated diene hydroperoxides located at C-9, C-12, C-13, and C-16, whereas the intermediate radical on linoleic acid reacts with oxygen to produce conjugated 9- and 13-diene hydroperoxides. The oxidation of oleic acid is 10 times slower than linoleic acid to produce a mixture of 8-, 9-, 10-, and 11-allylic hydroperoxides (Ho and Chen 1994; Whitfield and Mottram 1992). However, the hydroperoxides are very unstable and broken into alkoxy radicals by homolytic cleavage of the –OOH group, which then undergoes β-scission reaction on the C–C bond to form volatiles (e.g. aldehydes, alkene, alkane, and alcohols) (Ho and Chen 1994; Whitfield and Mottram 1992). Therefore, the volatile structure individually depends on the saturation (saturated vs. unsaturated) of alkyl chain and the breaking position of C–C bond by β-scission reaction (Whitfield and Mottram 1992). In termination, the combination of two radicals (e.g. the combination of peroxyl radical and alkoxyl radical, and the combination between alkyl radicals) occurs to form non-radical volatile species (e.g. aldehydes, ketones, hydrocarbons, lactones, alcohols, acids, and esters) (Fig. 1) (McClements and Decker 2007).

Maillard reaction

Maillard reaction, a nonenzymatic glycosidation of amino acid/proteins, is recognized as another most important chemical reaction taking place during deep-fat frying to determine the deep-fried flavor (Zamora and Hidalgo 2005).

Figure 2. Formation of 2-pentylpyridine from Maillard reaction between an amino acid and 2,4-decadienal (B) that is produced by the oxidation of linoleic acid (A) (Choe and Min 2007; Zhang and Ho 1989).

It is an extraordinary complex reaction starting with a free amino group (in amino acid, peptides, or proteins) and a reducing sugar (e.g. glucose, fructose, and lactose) to yield a N-substituted glycosylamine, which then undergoes an Amadori rearrangement to produce Amadori rearrangement product (ARP). Subsequently, ARP can either be cyclized to generate nitrogen-containing heterocyclic compounds (e.g. pyrroles and pyridines) or be broken into rearranged sugars,

which are super critical for the formation of volatiles via four pathways: (1) the rearranged sugars suffer keto-enol tautomerization (with 1-desoxy-2,3-diketones and/or 3-desoxy-1,2-diketones) to give back the original amino acid for further Maillard reaction; (2) the cyclization of the rearranged sugars occurs (in acid) to result in oxygen-containing heterocyclic compounds (e.g. furans and furfurals); (3) the rearranged sugars can be cleaved into dicarbonyl fragments

that react with amine groups from Strecker degradation of amino acids [involving oxidative deamination and decarboxylation of an α-amino acid (e.g. valine, leucine, and serine) in the presence of a dicarbonyl compound] to provide Strecker aldehydes (e.g. 2-methylpropanal, 3-methylbutanal, and 2-hydroxyethanal) and pyrazines (e.g. 2,5-dimethylpyrazine, 2,5-dimethyl-3-ethylpyrazine, and 2,3,5-trimethylpyrazine) in a base; and (4) the interaction between dicarbonyls (from the rearranged sugars) and proline/hydroxyproline can produce pyrrolines and pyrrolidines (Fig. 1) (Hodge 1953; Jousse et al. 2002; Mottram 1994). Flavor compounds produced by Maillard reaction mostly consist of aliphatic substances (e.g. aldehydes, ketones, and fatty acids) and heterocyclic derivatives (containing oxygen, sulfur, and/or nitrogen, e.g. furans, pyrroles, pyridines, and pyrazines), but heterocyclic derivatives with lower odor thresholds are more important to carry typical flavors than aliphatic substances at higher concentrations (Pokorny 1980).

In practice, the typical deep-fried flavor accounts for simultaneous changes on both frying oil and food components resulting from lipid oxidation, Maillard reaction, and the interactions initiated by intermediates/end-products from both reactions. Most of researches have built deep-fried model systems to investigate the participation of lipid oxidation products in Maillard reaction (Chun and Ho 1997; Macku and Shibamoto 1991; Peng et al. 2017; Ramirez et al. 2004; Zhang et al. 2015; Zhang and Ho 1989). In the work by Zhang and Ho (1989), an aldehydic group on 2,4-decadienal (a well-known fried flavor compound from lipid oxidation) was reacted with amino groups on glutathione or cysteine to produce 2-pentylpyridine (Fig. 2) via an electrocyclic reaction and aromatization at a typical frying condition (180 °C). But cysteine performed a poor reactivity, due to its degradation to form acetaldehyde, ammonia, and hydrogen sulfide. A similar result was also demonstrated by the addition of linoleic acid to a mixture of glucose and methionine under high temperatures. Linoleic acid was not only oxidized to produce 2,4-decadienal that was further re-used to form 2-pentylfuran, but also accelerated Strecker degradation of methionine to provide methional (Mandin, Duckham, and Ames 1999). Negroni, D'Agostin, and Arnoldi (2001) established Maillard model systems containing xylose/glucose and lysine with different frying oils (e.g. olive oil, canola oil, and sunflower oil) to study the impact of frying media on the development of fried flavor. They found unsaturation degrees of the oils expressed significant effects on pyrazines. More pyrazines generally formed in the system with olive oil, followed by canola oil and sunflower oil, but the production of 2-methylpyrazine, 2,5-dimethylpyrazine, and 2,3-dimethylpyrazine were more sensitive to sunflower oil (containing more linoleic acid). Therefore, lipid oxidation products can react with amino groups or Strecker degradation products of amino acids (e.g. ammonia, ammonium sulfide, and hydrogen sulfide) to generate heterocyclic volatiles containing ≥ 1 nitrogen/sulfur atoms on a long-chain alkyl substituent (containing ≥ 4 carbon atoms) to influence deep-fried flavor (Whitfield and Mottram 1992).

Hydrolysis and amino acid degradation

Apart from lipid oxidation and Maillard reaction, both hydrolysis and amino acid degradation independently and/or cooperatively assist the formation of deep-fried flavor. Hydrolysis of triacylglycerol to result in diacylglycerol, monoacylglycerol, glycerol, and free fatty acids is another source to facilitate the formation of deep-fried flavor. When food is fried in the oil, water molecules gradually evaporate and subside to the food under a bubbling action. So, the water with weak nucleophilicity attacks the ester linkage on the triacylglycerol to produce monoacylglycerols and diacylglycerols that can be further decomposed to glycerol (Chung, Lee, and Choe 2004). However, the specific breakage position is hard to figure out, which might be affected by carbon-carbon double bond position, carbon-chain length, moisture content, temperature, and unknown factors (Zhang et al. 2012). Subsequently, the glycerol can either evaporate at 150 °C or conduct hydrolysis to produce free fatty acids, which not only promote lipid oxidation to yield more volatiles, but also make the oil less acceptable for deep-fat frying by the generation of off-flavors (Naz et al. 2005). Hydrolysis is much more readily in the oils with short and unsaturated fatty acids than the oils with long and saturated fatty acids, because of their better solubility in water (Nawar 1969). Fortunately, oil replenishment is an effective way to slow down the hydrolysis and prolong the shelf-life of frying oils (Romero, Cuesta, and Sanchez-Muniz 1998). Stevenson, Vaisey-Genser, and Eskin (1984) claimed that the content of free fatty acids should be kept lower than 0.05-0.08% in the frying oil.

Thermal degradation of amino acids is observed during deep-fat frying to produce volatiles. Amino acids degradation can occur in two pathways to produce glycoaldehydes via Strecker degradation or to generate acetaldehydes through decarboxylation to loss ammonia molecules, which are then processed to volatiles in an assistance with aldol condensation and cyclization (Locas and Yaylayan 2004). Methionine was degraded into dimethyl disulfide and dimethyl trisulfide in the work by Mandin, Duckham, and Ames (1999), and the addition of linoleic acid resulted in unexpected higher levels (13–19 folds) of dimethyl sulfides. Therefore, linoleic acid oxidation facilitated the conversion of methionine sulfoxide, which was more readily to produce sulfides than methionine alone. Furthermore, both serine and cysteine can be metabolized to glycolaldehydes and acetaldehydes at the frying condition to be further converted to aldotetrose derivatives via aldol condensation, which are then cyclized to furan (Seok et al. 2015). However, aspartic acid, threonine, and α-alanine are not suitable to this mechanism, and they require the presence of reducing sugar to produce Strecker aldehydes (Yaylayan 2006). Overall, the deep-fat frying is a very complicated physicochemical process that involves intensive reactions and various substances under tremendous influences from external contributors. The volatiles with small molecular masses not only evaporate in the air, but are also re-absorbed in the frying system to participate and promote further complex reactions to regenerate more volatile compounds.

Factors affecting deep-fried flavor

According to the introduction of chemistry origins of fried volatiles in last section, lipid oxidation, Maillard reaction, hydrolysis, and amino acid degradation are mainly initiated by the macronutrients in frying oils (e.g. lipids) and food itself (e.g. carbohydrates and proteins) to form numerous volatiles to determine deep-fried flavor. So, frying oils and food components have great impacts on the formation of deep-fried volatiles. In addition, a number of relevant reactive conditions extensively influence the generation of volatiles in two categories: (1) similar to the reactants (e.g. amino acids and sugars in foods, and fatty acids in frying oils), pH determines the reaction pathways to regulate the nature of volatiles; (2) the frying temperature, time, oxygen concentration, moisture content, and frying type mainly exhibit influences on the kinetics of the reaction, rather than volatile varieties.

Frying oils

Since lipid oxidation is a protagonist in the formation of deep-fried flavor, the frying oil constructed by triacylglycerols and minor components (e.g. free fatty acids, phenols, sterols, and phospholipids) shows a great impact on the sensory characteristics of fried products (Cao et al. 2017). Specifically, fatty acid composition and oil type are paramount factors to influence the manufacture of volatiles by oxidative degradation of frying media (Whitfield and Mottram 1992). For instance, oleic acid is oxidatively degraded to yield higher amounts of unsaturated aldehydes (e.g. 2-decenal and 2-undecenal), followed by lower quantities of saturated aldehydes (e.g. nonanal and octanal) and hydrocarbons (e.g. octane and heptane) (Nawar 1998). The oxidation of linoleic acid generates various volatile compounds (e.g. 2,4-decadienal, 2,4-nonadienal, 2,4-octadienal, 2-heptenal, 2-octenal, and hexanal) to be more important in the determination of fried flavor (Buttery 1989). 2,4-Decadienal is the most representative deep-fried flavor compound, which comes from the breakdown of 9-diene hydroperoxide resulting from the oxidation of linoleic acid and is possibly further oxidized to produce (E)-4,5-epoxy-(E)-2-decenal (having a crumb flavor) (Schieberle and Grosch 1991). In addition, typical volatile compounds from the oxidation of linolenic acid are 2,4-heptadienal, 2,4-hexadienal, 2-pentenal, butenal, propanal, and acrolein (Nawar 1998; Katsuta et al. 2008).

According to a sensory analysis, potato chips fried in the canola oil with 64% oleic acid, 24% linoleic acid, and 3% linolenic acid released a stronger fried-potato flavor than them prepared in the canola oil with 78% oleic acid, 9% linoleic acid, and 4% linolenic acid, but the reason was not characterized in depth (Warner et al. 1994). Same group of researchers further reported hexanal, 2,4-decadienal, pentanal, octanal, and nonanal were representative volatiles to monitor the changes of fried flavor. The concentrations of hexanal, 2,4-decadienal, and pentanal (the most prominent volatiles originating from linoleic acid oxidation) were raised as oleic acid and linoleic acid were decreased and increased,

respectively. But, octanal and nonanal (originating from oleic acid oxidation) were not significantly changed (Warner, Orr, and Glynn 1997). This phenomenon is also proved by Wagner and Grosch (1998), who stated more decadienal isomers [(E,E)- and (E,Z)-2,4-decadienal] were liberated from French fries prepared by palm oil (10.5% linoleic acid) than those from coconut fat (1.9% linoleic acid). In the study by Peng et al. (2017), sunflower oil (rich in linoleic acid) performed higher emission of aldehydes (e.g. butanal, hexanal, 2-heptenal, 2,4-nonadienal, and 2,4-decedienal) than rapeseed oil (rich in oleic acid) and palm oil (rich in saturated fatty acids). As a result, the frying oils containing 16–42% oleic acid and 37–55% linoleic acid is suggested to prepare fried foods with moderate flavor intensity and advantageous flavor quality (Warner, Orr, and Glynn 1997).

Regarding to linolenic acid, Frankel, Warner, and Moulton (1985) claimed soybean oil (linolenic acid $\geq$ 8.5%) was extremely destroyed to release undesirable pungent, fishy, and paraffin-like odor during bread frying at 190 °C. The rejection of linolenic acid is also explained in deep-fried potatoes by linseed oil (55% linolenic acid) to produce extreme high quantities (about 5 times more) of toxicological relevant compounds (e.g. acrylamide and glycidamide) than coconut and safflower oils (<1% linolenic acid) (Thurer and Granvogl 2016). Considering on these results, lowering linolenic acid (< 3%) has been settled as a goal for plant breeders to improve oxidative stability, sensory quality of frying oils, and safety of deep-fried food products (Liu and White 1992).

Furthermore, Ramirez and coworkers (2004) evaluated volatile compositions of pork loin chops fried in conventional fats [e.g. butter and pig lard; dominated by saturated fatty acids (~53%)] and vegetable oils [e.g. olive oil and sunflower oil; dominated by unsaturated fatty acids (~86%)]. More aliphatic aldehydes (e.g. hexanal and pentanal) from lipid oxidation were detected as the degree of unsaturation elevated in the vegetable oils, whereas unique Strecker aldehydes (e.g. 2-methylbutanal and benzeacetaldehyde) produced by Strecker degradation of amino acids (e.g. isoleucine and phenylalanine) were released from the conventional fats fried chops. On the contrary, fried volatiles are not always dependent on the frying oil. The formation of pyrazines (e.g. 2,3-diethyl-5-methylpyrazine and 2-ethyl-3,5-dimethylpyrazine) was not very pronounced by coconut oil, olive oil, rapeseed oil, and safflower oil in the study by Thurer and Granvogl (2016). This was also confirmed by Wagner and Grosch (1998), who concluded the differences on alkylpyrazines from French fries prepared by palm oil and coconut oil were not significant. In consequence, more factors might be acted to influence deep-fried flavor other than frying medium.

Food components

As a matter of fact, the flavor of deep-fried food depends much on the food components rather than the frying oil (Dobarganes, Marquez-Ruiz, and Velasco 2000). For instance, nitrogen-containing volatiles (e.g. 2-pentylpyridine,

2-propylpyridine, and pyrrole-3-butyronitrile) were only detected in deep-fried chicken breast meat rather than the fried wheat dough in the work by Zhang et al. (2018). Peng et al. (2017) studied the flavors of deep-fried pork loins and potatoes, and concluded acetaldehyde, butanal, hexanal, and 2,4-decadienal in the fried pork loins were significantly higher than those in fried potatoes. So, the complexity of food components profoundly affects the production of deep-fried flavor. Lu, Kuhnle, and Cheng (2018) found deep-fried chicken meatballs had less heterocyclic amines than the fried beef meatballs, because a high level of nonheme iron in beef reacted with hydroxyl and peroxyl radicals to accelerate lipid oxidation and Maillard reaction as compared with chicken (Gibis 2016). Although carbonyl groups on lipid oxidation products are identified to replace the reducing sugar in Maillard reaction (Mandin, Duckham, and Ames 1999; Zhang and Ho 1989), their importance is questioned by Adams and coworkers (2011), who found glucose was more competitive than 2-hexenal to react with lysine for the volatile production. This is also proved by Miyagi and Ogaki (2014a), who stated free amino acids and sucrose (decomposed to glucose and fructose) contents were reduced by 89% and 25% during frying by Maillard reaction and caramelization, respectively. Hence, reducing sugar is still essential to provoke the liberation of aroma compounds during deep-fat frying.

Moreover, the influence of amino acids on the flavor should not be overlooked by their significant sensory attributes. For example, alanine contributes to sweet and malt characters of odor; more lysine increases nutty and burnt odors; methionine raises cooked potato flavor; and the presence of tryptophan is necessary for the burnt meat smell (Pokorny 1980). A total of 29 pyrazines and pyridines in the mixtures of glutamine/asparagine and glucose fried in corn oil were analyzed by Chun and Ho (1997), who presented more alkylpyrazines and pyridines (particularly 2-pentylpyridine) were released from glutamine model system, because a stronger deamination reaction of glutamine occurred to provide ammonia under frying conditions. Theoretically, however, asparagine is more susceptible to both deamidation and deamination to generate ammonia under high temperatures (Sohn and Ho 1995). This indicates the degradation of amino acids can be alternated by the deep-fat frying to affect the production of volatiles. From the view point of nucleophilicity, lysine (pK_a = 8.95) exhibited poorer performance on the conversion of hexanal (78% unreacted) to 2-butyl-2-octenal (a ham-like flavor compound) than glycine (pK_a = 9.78), which was more readily accessible to Maillard reaction (only 5% hexanal was left) (Adams et al. 2011). In addition, the fried flavor is also differentiated between acidic and basic amino acids. In the work by Kavousi et al. (2015), acidic amino acids (e.g. glutamic acid and aspartic acid) lowered the pH to push the degradation pathway of Amadori products towards 5-hydroxymethylfurfural, whereas basic amino acids (e.g. lysine, arginine, and histidine) increased pH (> 6) and decelerated the formation of 5-hydroxymehylfurfural to a non-detectable level. Hence,

food components play an indispensable role in the formation of deep-fried flavor.

Frying conditions

A proper frying condition (e.g. oxygen concentration, frying time, frying temperature, and pH) is critical to develop a desirable deep-fried flavor. In literature, a typical fried flavor is induced by an optimum oxygen concentration to encourage a proper lipid oxidation rate, whereas too low and too high oxygen contents result in weak fried flavor and off-flavor, respectively (Pokorny 1989). Chyau and Mau (2001) explained a lighter fried shallot flavor in medium-chain triglycerides was caused by little oxygen involvement as compared with the stronger flavor in soybean oil and corn oil. Additionally, the concentration of volatiles gradually climbs up as the frying time is prolonged. Take 2-nonenal as an example, which was gently raised during wheat dough frying in soybean oil. But this is not always true, an initial increase followed by a decrease of 2-nonenal was observed when chicken breast meat was fried, due to an elimination action between volatiles (Zhang et al. 2015). Adams and coworkers (2011) also demonstrated the volatile concentration was declined by twice when the heating time was extended to 120 min in a Maillard model established by lipid oxidation products (e.g. hexanal and 2,4-decadienal) and amino acids (e.g. lysine and glycine) with/without glucose. Similar phenomenon is also observed on the deep-fried peanuts, at which the deep-fried peanuts aroma increased with frying time (2–15 min), but the highest overall liking score was given when the peanuts were fried for 4–9 min (Miyagi and Ogaki 2014b).

Frying temperature is another vitally important aspect to determine sensory characteristics of deep-fried foods, at which higher temperature substantially facilitates complex chemical reactions (by opening structures of reactants then being ready for the reactions) to stimulate the generation of volatiles. Frying temperature of safflower oil was decreased from 180 °C to 140 °C and resulted in a huge loss (almost 86%) of key odorants [e.g. 2,4-decadienal, 3-methylbutanal, and 3-(methylthio)propanal] (Thurer and Granvogl 2016). Mandin, Duckham, and Ames (1999) claimed lower reaction temperature (100–105 °C) was responsible for the very light cooked flavor in the mixtures of linoleic acid, glucose, and methionine, due to the absence of pyrazines, furans, thiazoles, and oxazoles. Regardless of potential toxicological compounds (e.g. acrylamide), relative high temperature is corroborated to improve odor impression of deep-fried foods.

Moisture is an inevitable factor participating in a complex series of reactions (e.g. hydrolysis, oxidation, isomerization, and Maillard reaction) to give rise to flavor formation in the deep-fat frying. In most cases, moisture is considered as a negative point to induce undesirable off-flavor in the fried products and frying oil by intensive deterioration reactions to rancidify frying oils (Bazina and He 2018). Mandin, Duckham, and Ames (1999) identified an absence of pyrazines, pyridines, and thiazoles under high moisture conditions resulting from negative effects of Maillard reaction. In

addition, deep-fried banana chips with higher moisture content exhibited worse odor acceptability to testers (Ammawath et al. 2001). However, an innovative frying was developed by Ma and coworkers (Ma et al. 2016), who used the mixture of oil and water as an alternate to pure oil. In this case, the oil was an upper layer at frying temperatures, whereas the water was a lower layer below 55 °C, so, the food descended into water before carbonization at the high temperature of oil, and was finally removed through plughole in the fryer. Hence, the rate of oil deterioration was mitigated by the presence of water during the frying, meanwhile desirable fried sensory characteristics were maintained. As a result, the application ways of moisture will have a great influence on the development of deep-fried flavor.

Moreover, pH is a predominant factor to influence volatile production by affecting ARPs degradation to alternate kinetics of Maillard reaction (Jousse et al. 2002). At pH ≤ 7, 1,2 enolization of ARPs attributes to the formation of furfural (with pentoses) and hydroxymethylfurfural (with hexoses), whereas 2,3 enolization at pH > 7 produces 4-hydroxy-5-methyl-2,3-dihydrofuran-3-one and fission products (e.g. acetal, pyruvaldehyde, and diacetyl) (Martins, Jongen, and van Boekel 2000). Yaylayan and Huyghues-Despointes (1996) mentioned ARPs can be degraded to acetic acid and pyruvaldehyde with free amino acids under basic conditions, so, the pH > 7 is considered as a major pathway for flavor formation via Maillard reaction. Meanwhile, pH also affects carbohydrates hydrolysis and the formation of unprotonated amino acid (a reactive form for Maillard reaction). Starch is hydrolyzed faster at acidic pHs to promote the release of carbonyl groups joining into Maillard reaction (BeMiller 1965). At lower pHs ($<$pK_as of amino groups), less unprotonated amino acids are available to result in a milder Maillard reaction (Martins, Jongen, and van Boekel 2000). For example, the production of 5-hydroxymethylfurfural was gradually diminished as the pH increased from 4 to 10, since 1,2-enolisation pathway was blocked by more protonated form of glutamic acid (Kavousi et al. 2015).

Frying type

It is well known conventional frying includes stir-frying, pan-frying, and deep-frying, which are distinguished by the amount of oil used. Stir-frying and pan-frying normally requires small amounts of oils and longer cooking time (5–10 min), whereas the food is submerged into a large amount of oil at extreme high temperatures to induce violent reactions during deep-fat frying (Gertz, Klostermann, and Kochhar 2000). Although volatiles from different frying types are resembling same, the highest emissions of volatiles, especially the typical fried volatiles (e.g. hexanal and 2,4-decadienal), are found by deep-frying, followed by pan-frying and stir-frying (Peng et al. 2017). Deep-fried flavor, basically composed of a number of volatiles (Table 2), is described as oily, fruity, grassy, buttery, burnt, nutty and fishy odor (Nayak et al. 2016).

Today, consumers are more interested in healthy diet and are seeking for food products with extraordinary quality in the appearance, nutrition, safety, functionality, and sensory property. Vacuum frying attracts more attentions by the application of lower temperature, minimum contact with oxygen, and lower oil absorption to produce fried products with less oil content and higher retention of natural colors and flavors (Moreira 2014). It is the only frying technology that can be applied on delicate food sources with high sugar content (e.g. fruit and vegetables). As compared with deep-fat frying at atmosphere, vacuum frying operates at lower temperature (90–150 °C) under pressure <6.65 kPa to decrease the boiling point of water (Moreira, Castell-Perez, and Barrufet 1999). As demonstrated by Aladedunye and Przybylski (2009), Crosa et al. (2014), and Nazarbakhsh et al. (2014), vacuum frying showed superior protections than atmospheric frying on canola oil, sunflower oil, and high-oleic acid sunflower oil to retard oxidative deterioration, which was assessed by total polar compounds, anisidine value, fatty acids composition, and antioxidative activity. In spite of diverse studies are available to use vacuum frying on fruits and vegetables, its applications on meat products are scarce. Teruel and coworkers (Teruel et al. 2014) intended to prepare healthier fried chicken nuggets by using vacuum frying (at 130–150 °C) as an alternative process to deep-fat frying (at 165 °C). Vacuum fried chicken nuggets at 150 °C for 6 min obtained better organoleptic characteristics (including color, crispness, juiciness, oiliness, and overall acceptance) than the deep fried one, because of the extreme high score on the crispy texture. But the juiciness and the whiteness (L value) in color were undesirable by the loss of water content in the vacuum fried chicken nuggets. In this study, little information was available on the flavor except oiliness, which was not significantly affected by frying type. An opposite conclusion was reported by Yang et al. (2012) on the rice bran oil fried sweet potato snacks prepared by vacuum frying (at 90 °C under 20 kPa) and atmospheric frying (at 170 °C for 2.5 min). Vacuum fried snacks had better acceptability on color and flavor, whereas the deep-fat fried snacks were crispier. Similar results were also proved by Troncoso, Pedreschi, and Zuniga (2009), in which sensory attributes, flavor quality and physicochemical features of fried potato slices were significantly improved by vacuum frying, even though better flavor was achieved on the control sample (Desiree potato slices prepared by traditional frying at 120–140 °C without pretreatments). But certain volatile compounds were not evaluated to explain the flavor difference in both studies. Therefore, the measurement of volatile profile is still necessary for the feasibility analysis of vacuum frying in the food manufacturing.

Air frying is another novel alternative technique to deep-fat frying, which is developed to match the requirements from consumers on the preparation of fried foods with lower fat content and similar sensory features as conventional frying. An emulsion of oil droplets in hot air is created when food products are fried in the air frying chamber, so the product homogeneously contacts with hot air and oil to be dehydrated and forms a typical crust structure of fried products (Andres et al. 2013). Air frying is able to reduce fat content in the fried food up to 90% in comparison with

deep-fried ones (Sansano et al. 2015). Yang and coworkers (2017) found the variety and total amount of aroma-active compounds of air-fried pork loin were only 1/5 and 1/2 of these compounds as in the deep-fried one, in which 2-pentylfuran, 2,5-dimethylpyrazine, 2,6-dimethylpyrazine, 2-methyl-3-propylpyrazine, 6-methyl-2-ethylpyrazine, and trimethylpyrazine were not presented in the air-fried samples. This might be attributed to the lower rate of Maillard reaction and lipid oxidation as initial reducing sugar and frying oil contents were increased and decreased, respectively (Sansano et al. 2015). Caetano and coworkers (Caetano et al. 2018) announced similar findings on sweet potato chips fried in canola oil, in which the deep-fried chips showed much better sensory acceptance to induce higher purchase intent by consumers than the air-fried chips. Although air fried products present lower fat and moisture contents with better texture properties (e.g. crispiness), weaker aroma features hinder the extension of air frying in the fast food and manufacturing industries. So, a better scientific understanding to improve organoleptic sensation of air fried foods is essential to provide sufficient evidences to alternate deep-fat frying.

Overall, compared to other frying types, deep-fat frying is still a superior thermal treatment to provide a great quality on sensory characteristics (e.g. color, texture, and aroma), because of the usage of oils in large quantities, fast heat transfer, dehydrated environment, short cooking time, and complex chemical reactions, and to finally produce a very attractive deep-fried flavor (Ghidurus et al. 2010). Most operational suggestions on deep-fat frying are previously concentrated on the reduction of off-flavors resulting from excessive lipid oxidation. Stevenson, Vaisey-Genser, and Eskin (1984) recommended a replenishment of frying oil by 15–25% capacity of the fryer should be done to lower the application of silicones (an antifoaming agent) and limit off-flavors. Przybylski and Eskin (1988) suggested flushing nitrogen (15 min) or carbon dioxide (5 min) to frying oils prior to frying can significantly prevent oil oxidation to reduce rancid flavor. However, practical steps and concerns on the manipulation of major chemical reactions (e.g. lipid oxidation and Maillard reaction) are rarely inquired to optimize deep-fried flavor.

Potential harmful products in deep-fried flavor

Many studies have been announced deep-fat frying favors the generation of undesirable toxicological relevant compounds [e.g. acrylamide, acrolein, glycidamide, α, β-unsaturated aldehydes, and heterocyclic aromatic amines (HAAs)] by violent chemical reactions at high temperatures to potentially threaten human health. Mutagenic and carcinogenic HAAs are most commonly studied potential toxic chemicals originating from thermal processed protein-rich products. They include thermic-HAAs (resulting from creatine, free amino acids and monosaccharides through Maillard reaction at 150–250 °C) and pyrolytic-HAAs (possibly resulting from amino acids pyrolysis at temperature > 250 °C) (Jagerstad et al. 1998; Matsumoto, Yoshida, and Tomita 1981). In the

25 identified HAAs, 2-amino-3,8-dimethylimidazo[4,5-f]quinoxaline [MeIQx, possible carcinogenic to human (Groups 2B)], 2-amino-1-methyl-6-phenylimidazo[4,5-b]pyridine (PhIP, Groups 2B), and 2-amino-3-methylimidazo[4,5-f]quinoline [IQ, probable carcinogenic to human (Group 2A)] are the most abundant HAAs exposed in the cooked foods (IARC 1993). A number of researches observed the formation of HAAs drastically promoted as the frying temperature and time increased (Shabbir et al. 2015). In addition, acrolein (Group 3: not classifiable to carcinogenic toxicants) and furan (Group 2B) generated during deep-fat frying via multiple pathways (e.g. carbohydrates and amino acids degradations, lipid oxidation, and Maillard reaction) also have potential adverse effects on human health (IARC 1993). Since the toxicity of aldehydes (best known deep-fried volatiles) is highly dependent on their abilities to introduce inter- and/or intramolecular cross-linkages to modify proteins and nucleic acids and interfere cell metabolisms (Witz 1989), further establishments on regulations and classification of aldehydes will be fundamental to provide a clear guidance on deep-fat frying.

In current, majority of researchers intended to lower frying temperatures, apply pretreatments (e.g. microwaving and edible coating), and limit the involvement of oxygen (by vacuum fryers), in order to prevent the production of these food-borne toxicants (Hosseini et al. 2016; Thurer and Granvogl 2016; Yang, Achaerandio, and Pujola 2016; Zamora et al. 2016; Zhang et al. 2012), but the intensity of fried flavor would be drastically declined to decrease the sensory quality of deep-fried products. For example, the total content of HAAs were decreased by ∼70% in the air-fried pork loin, but its sensory satisfaction was greatly lowered in comparison with deep-fried one (Yang, Achaerandio, and Pujola 2016). Nevertheless, Haskaraca et al. (2014) did not find any detectable effect on the HAAs by adding green tea extract into the fried chicken meat, but lowering the number of frying cycles led to a reduction of HAAs contents in the grass carp with a negative organoleptic response (Wang et al. 2015).

Future studies

The development of deep-fried flavor is an intricate process induced by chemical reactions (e.g. lipid oxidation, Maillard reaction, hydrolysis, and amino acid degradation) between food components (e.g. proteins and reducing sugars) and frying oils, in which lipid oxidation products involved Maillard pathway is recently studied to illustrate the complexity of flavor production in simulated food systems. However, the absence of essential conditioning elements (e.g. pH and water activity) in the simulated system decreases the authenticity of chemical reactions taking place during deep-fat frying and negatively impacts the comprehensive study of deep-fried flavor. Further studies in this field should also be extended on the reactions of more lipid oxidation products (e.g. nonanal, 2-heptenal, and 2,4-heptadienal) with amino acids, to possibly identify novel potent flavor compounds that have not been detected

out in the fried foods. In addition, aliphatic aldehydes and ketones (from lipid oxidation) have been identified to react with reducing sugars to produce acetals and ketals that were decomposed to volatiles [e.g. alkylfurans, 2-furfurals, and 3(2H)-furanones] (Whitfield and Mottram 1992). Apparently, the interactions between lipid oxidation products and reducing sugars have not been investigated during deep-fat frying, which might be valuable to explain deep-fried flavor.

Factors influencing the fried flavor mainly include fatty acid composition, amino acid, frying temperature, pH, moisture, and frying type. Comparing with oleic acid, linoleic acid oxidation releases more various volatiles responsible for typical deep-fried flavor, whereas linolenic acid is more susceptible to oxidation and potentially generates toxicological chemicals, so, it is suggested linoleic acid content is better to be twice than oleic acid in the frying oil with < 3% of linolenic acid. Depending on deamination reaction, nucleophilicity, and pKa values of amine groups, amino acids in foods behave differently to determine the fried flavor. From the view point of frying condition, the formation of volatiles is more favorable in a basic environment at higher temperatures. Therefore, frying media and frying conditions must be carefully selected to optimize the volatile formation during deep-fat frying. In fact, flavor perception is exceedingly complicated, not only relies on the sense of individual volatile compound, but also integrates additive, synergistic, and masking actions between volatiles (Nawar 1998). Thus, a simple analysis of volatile composition is insufficient to understand the complex flavor pattern, and repeated examinations during frying process and investigation of combined groups of volatile interactions are necessary to reflect the continuous volatile changes and comprehensively clarify the fried flavor perception.

While, one side effect of deep-fat frying is the losses of macronutrients (e.g. amino acids and sugars) through Maillard reaction, caramelization, and Strecker degradation. Oluwaniyi, Dosumu, and Awolola (2010) reported deep-fat frying greatly reduced total amino acids content in *Scomber scombrus* by 2–19%, in which available lysine was notably damaged by palm oil frying. They also noticed Maillard reaction between amino groups and aldehyde groups (on reducing sugars) or carbonyls (on oxidized fats), cross-linking reactions, and protein–protein interactions at frying temperatures made amino acids metabolically unavailable. In conclusion, balancing frying conditions and regulating frying materials to minimize the production of toxicological relevant substances with a simultaneous maintenance of ideal deep-fried flavor is a critical challenge, but it is vitally important to substantially improve food quality and safety to increase consumer acceptance and economic benefits of deep-fried foods in the future.

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