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Lipid Oxidation, Maillard Reaction, and Their Possible Interrelation in Selected Foods and Model Systems – A Review

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Abstract

In food matrices, two major reactions occur: lipid oxidation and the Maillard reaction. Both involve a series of reactions that produce complex mixtures of compounds in varying concentrations. These compounds show desirable and undesirable effects on the flavor, color, texture, and nutritional profile of food. This article focuses on the influence of lipid oxidation products on the Maillard pathway, the effect of Maillard reaction products on the lipid oxidation pathway, and the presence of common intermediates (such as glyoxal and methylglyoxal) in both reactions. Lipid oxidation produces analogous carbonyl compounds, which take part in the Maillard reaction and enhance the production of advanced glycation end products (such as Nε-carboxymethyllysine and Nε-carboxyethyllysine). These relationships can be understood through the oxidation of linoleic acid to 4,5-epoxy-2-alkenals (carbonyl compounds), which react with amino acids to produce high-molecular-weight polymerized compounds, such as 2-(1-hydroxyalkyl)pyrroles. On the other hand, Maillard reaction products are identified with free radical scavenging ability or antioxidant activity in some foods and model systems. These findings help in understanding the importance of reducing lipid oxidation in foods, and controlled Maillard reactions produce antioxidant compounds, which are useful in the improvement of food quality.

Keywords: Antioxidant compounds, common intermediates, lipid oxidation, maillard reaction

INTRODUCTION

As modern lifestyles become increasingly fast-paced, individuals are often occupied with their daily routines, leading to a growing reliance on processed foods. These foods typically contain significant amounts of fats or oils, carbohydrates, and proteins. During processing and storage, various chemical reactions occur among these constituents, influencing the food's characteristics. Lipid oxidation and Maillard browning are the most important chemical reactions responsible for alterations in color, flavor, texture, and nutritional value. Understanding these reactions is essential for improving food quality, ensuring safety, and maintaining nutritional benefits.

Lipid oxidation is one of the fundamental chemical reactions occurring in both unprocessed and processed foods. This reaction affects the organoleptic and nutritional characteristics, resulting in a decline in quality and shortening the shelf life of foods. It proceeds as a free-radical chain reaction, consisting of initiation, propagation, and termination stages [1]. During this process, unsaturated fatty acids are oxidized to form odorless and tasteless hydroperoxides. These hydroperoxides are unstable and further decompose into flavorful carbonyls and other compounds. These compounds are sometimes desirable for the formation of aromas in typical cheeses or fried

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foods [2]. Quality attributes including appearance, texture, consistency, taste, flavor, and aroma may deteriorate as a result [3]. In addition, the formation of toxic compounds and a reduction in nutritional quality have also been reported [4]. Controlling the progression of lipid oxidation in foods is a key factor in preserving quality and prolonging shelf life.

Initiation of the Maillard reaction occurs through the interaction of the reactive carbonyl group of a sugar with the nucleophilic amino group of an amino acid. This initial step leads to the formation of several reactive carbonyl compounds, which play a crucial role in the development of advanced glycation end products (AGEs) in processed foods. There are two generation pathways of AGEs, which include endogenous and dietary AGEs [5]. Glycation pathways are the primary routes for the formation of AGEs such as *N*ε-carboxymethyllysine (CML), *N*ε-carboxyethyllysine (CEL), and pyrraline. The formation of AGEs through the Maillard reaction pathway begins with the formation of a Schiff base, followed by an Amadori rearrangement product, which can further promote the generation of dicarbonyl compounds [6, 7]. These dicarbonyl compounds can then react with lysine or arginine residues to produce various AGEs [8].

Recent literature shows that reactive carbonyls are not only derived from carbohydrates. Lipid oxidation is also a significant source of reactive carbonyl compounds, which are commonly formed during food processing and storage. In the chain reaction of lipid oxidation, the formation of these compounds contributes to the undesirable flavors associated with oxidative rancidity in foods. Glyoxal (GO), methylglyoxal (MGO), D-glucosone (GLO), and 3-deoxyglucosone (3-DG) are common intermediate compounds produced during both Maillard browning and lipid oxidation. These compounds can further react with amino acids to form AGEs (CML, CEL, and pyrraline, etc.) [9, 10].

In processed foods, the presence of synthetic antioxidants has become a major concern because they may be harmful to human health. Consequently, there is a growing need for natural alternatives. Previous studies on Maillard browning have shown that MRPs can function as antioxidants. Several MRPs are identified with the capability of scavenging free radicals and chelating metal ions in emulsions [11]. These MRPs could therefore be used as natural antioxidants in foods to counteract lipid oxidation. In addition, MRPs show emulsifying activity; these MRPs made a partition between the interface and the continuous phase [12].

Numerous studies have thoroughly examined the individual reactions responsible for food browning; however, their mutual interactions have received relatively limited attention in scientific literature. This review discusses the fundamentals of two major nonenzymatic browning reactions: lipid oxidation and the Maillard reaction, their occurrence in foods (meat, fish, bakery products, and model systems), the intermediate and end products formed, their interrelation, and their effects on food quality. This information is helpful in understanding the importance of these two reactions in other significant food commodities, such as milk and milk products.

LIPID OXIDATION

The principal reaction involved in lipid oxidative deterioration is the autoxidation of fats, though in foods, lipids are susceptible to oxidation through both enzymatic and nonenzymatic mechanisms. The autoxidation or nonenzymatic oxidation of fats advances via free radical mechanisms and is a common chemical reaction during food processing and storage. It predominantly involves the interaction between mainly unsaturated fatty acids and oxygen. The mechanisms underlying autoxidation and the widely accepted explanation is the free radical chain theory [13]. According to this theory, the oxidation of unsaturated fatty acids occurs as a chain reaction and is autocatalytic, with oxidation products acting as catalysts, thereby increasing the reaction rate as the process progresses. The steps involved can be summarized in a simplified scheme. The latter provided a straightforward and convincing explanation for autoxidation, highlighting that oxygen-mediated oxidation reactions in food systems are commonly referred to as autoxidation [1]. This reaction not only reduces the nutritional value and shortens the shelf life of food products but, in some cases, also leads to the formation of toxic and carcinogenic compounds [14, 15].

During the initial stages of lipid oxidation, formation of lipid hydroperoxides occurs. These primary lipid oxidation products increase and eventually decrease due to their conversion into secondary oxidation products as the food begins to deteriorate. These secondary products include hydrocarbons, acids, lactones, esters, ketones, and aldehydes. The autoxidation of food lipids is accelerated by factors such as heat, light, singlet oxygen, and trace metal catalysts, particularly copper and iron [16].

Initiation

$RH \rightarrow R^* + H^*$

$RH + O_2 \rightarrow RO^* + HO^*$

The initiation of the reaction involves the abstraction of hydrogen to form a free radical species in the presence of an initiator. It may take place by hydroperoxide decomposition, metal catalysis, singlet oxygen, or by exposure to light.

Propagation

$R^* + O_2 \rightarrow ROO^*$

$ROO^* + RH \rightarrow ROOH + R^*$

The addition of oxygen occurs at the carbon atom adjacent to the double bonds (methylene carbon), resulting in the formation of peroxy radicals (ROO*), and these in turn abstract hydrogen from a methylene groups (RH) of the other fatty acid molecules to yield hydroperoxides (ROOH) and another free radicals (*R). The newly formed free radicals combine with oxygen, leading to a repetition of the reaction sequence. In addition to the formation of hydroperoxides, other types of reactions may occur. The peroxides may decompose to carbonyls, form polymers, or react with proteins, vitamins, pigments, and other components [17, 18]. The hydroperoxides are intermediates in the oxidation process. These compounds are unstable and break down to a variety of products such as aldehydes, ketones, acids, and alcohols, which contribute to rancid flavors [19].

Termination

$R^* + R^* \rightarrow R - R$

$R^* + ROO^* \rightarrow ROOR$

$ROO^* + ROO^* \rightarrow ROOR + O_2$

These reactions lead to the formation of inactive stable end products.

The autoxidation of linoleic acid is shown in Figure 1. Autoxidation of linoleic acid in the presence of metal catalysis, singlet oxygen, or by exposure to light produces the free radical, which is unstable, and shifting of the double bond produces two cases. The reduction of linoleic acid produces hydroperoxides such as 9-hydroxy-10,12-octadecadienoic acid and 13-hydroxy-9,11-octadecadienoic acid. These hydroperoxides are involved in the production of carbonyl compounds like 4,5-epoxy-2-alkenals (Case 2).

MAILLARD REACTION

The Maillard reaction is a nonenzymatic chemical reaction involving the interaction of amino groups of amino acids and carbonyl groups of reducing sugars (e.g., glucose, fructose, maltose, and lactose). The reaction is named after the French biochemist Louis-Camille Maillard (1878–1936), who first described the formation of brown pigments or melanoidins upon the heating of a solution of glucose and lysine. The Maillard reaction as a sugar–amine reaction should be distinguished from caramelization, which occurs when pure sugars are heated to form brown pigments [20]. In baked and fried foods, the reaction is beneficial, whereas in concentrated and dried foods, it is undesirable. A widely accepted mechanism for the reaction has been published [21]. This nonenzymatic glycosylation process proceeds through three main stages.

In the early stage, carbonyl groups react with free ε-amino groups through condensation, resulting in the formation of unstable Schiff base compounds [22]. These compounds then undergo cyclization and in turn form reversible *N*-substituted glycosylamines. When derived from aldoses, *N*-glycosylamines form Amadori compounds (1-amino-1-deoxyketoses), whereas those from ketoses lead to the formation of Heyns compounds (2-amino-2-deoxyaldoses) [23]. Although these intermediates do not contribute to browning, they may lower the nutritional quality by reducing the availability of amino acids [24].

The intermediary stage starts with the decomposition of Amadori and Heyns compounds via different routes [25]. The 1,2-enolization of these compounds produces furfural or hydroxymethylfurfural (HMF) under acidic conditions, whereas 2,3-enolization leads to the formation of reductones (e.g., 4-hydroxy-5-methyl-2,3-dihydrofuran-3-one, HMFone) and fission products (e.g., glyoxal (GO), methylglyoxal (MGO), and acetol) under alkaline conditions. The formed dicarbonyl compounds can interact with amino acids to produce aldehydes and aminoketones through Strecker degradation [26], or react with arginyl and lysyl residues to form advanced glycation end products (AGEs), including *N*ε-(carboxymethyl)lysine (CML) and *N*ε-(carboxyethyl)lysine (CEL) [27]. In this stage, a yellowish color, flavor formation, and increased reducing power are observed [11].

In the final stage, intermediate-stage reactive compounds can undergo a series of reactions including retro-aldolization, isomerization, rearrangement, and condensation, which lead to the formation of brown, nitrogen-containing, high-molecular-weight polymers referred to as melanoidins [28]. The chemical structures and formation mechanisms of these compounds are still not completely understood. In most thermally processed foods, the final stage of the Maillard reaction is chiefly responsible for color and flavor development.

The common amino acid lysine, present in many foods, is involved in the formation of harmful AGEs. Lysine-derived AGEs, including CML, CEL, and pyrraline, are formed not only through the degradation of Amadori products (fructoselysine) during the Maillard reaction but also via the direct reaction of lysine with reactive dicarbonyl compounds such as GO, MGO, and 3-deoxyglucosone (3-DG) [5, 30, 31]. Acrylamide, a harmful AGE contaminant in foods, is generated from the reaction between asparagine and carbonyl compounds during the Maillard reaction and is recognized as a probable human carcinogen. Another compound, 5-hydroxymethylfurfural (HMF), which forms as an intermediate in the Maillard reaction, can be converted into other mutagenic compounds.

EFFECT OF LIPID AUTOXIDATION ON MAILLARD BROWNING

The role of lipid oxidation in nonenzymatic browning was first highlighted [32], showing that acetaldehyde could induce protein browning. Furthermore, [33] described the formation of covalent bonds between oxidized lipid chains and amino acids or proteins without necessarily breaking the lipid chain. This process involves reactions between the reactive groups of amino acid residues and secondary products of lipid peroxidation. The electrophilic nature of free radicals (hydroperoxides, peroxyl, and alkoxyl radicals, carbonyl compounds, and epoxides) generated during lipid oxidation leads to interactions with nucleophiles such as free amino acids, peptides, and proteins. These compounds are derived from the decomposition of lipid hydroperoxides. The carbonyl compounds formed from oxidized lipids readily condense with free amino groups of proteins, forming Schiff bases, which play a pivotal role in the progression of nonenzymatic browning. These Schiff bases undergo polymerization through aldol condensation, producing dimers, trimers, tetramers, and complex high-molecular-weight oxypolymers (macromolecules). Such polymeric brown materials are inherently unstable [34]. Their degradation, through mechanisms like macromolecular scission or dehydration, results in the release of new volatile compounds that significantly influence the flavor profiles of foods during cooking and processing. Moreover, the amino compound can also detach from the polymer, which could partly explain the wide variation in nitrogen content found in the polymer [35].

Researchers have also reported that three types of free radical species are produced in the interaction between amino acids and methylglyoxal (MGO), including cross-linked radical cations, superoxide radical anions, and MGO radical anions. This cross-linked radical cation is a precursor of AGE formation [36].

Figure 2 shows the formation of higher melanoidin-like polymers or brown polymers. These polymers are produced through the polymerization of 2-(1-hydroxyalkyl)pyrroles, which are formed from the reaction between 4,5-epoxy-2-alkenals (or other carbonyl compounds) and amino compounds.

Meat

The effect of different frying time–temperature combinations on the formation of AGEs through the Maillard reaction and the role of lipid oxidation in batter-coated meat was studied. Researchers found that prolonged high-temperature frying significantly increases AGE formation in batter-coated meat products. The effect of lipid oxidation on the Maillard reaction was demonstrated by the estimation of thiobarbituric acid reactive substances (TBARS value). They found increasing CML and CEL content as the TBARS value increases [37]. Lipid oxidation combined with the Maillard reaction in braised chicken processing showed that the content of protein carbonyl and TBARS value continued rising with the increase in frying, boiling, and sterilization time. Data show that frying for 3 min compared with 0 min increased the carbonyl value by 1.83 nmol/g protein and the TBARS value by 2.69 mg malondialdehyde (MDA)/kg meat. In the case of boiling for 60 min compared with 0 min, the protein carbonyl content increased by 1.14 nmol/g protein and the TBARS value increased by 1.16 mg MDA/kg meat. The sterilization of chicken for 30 min compared with 0 min increased the protein carbonyl by 1.27 nmol/g protein and the TBARS value by 1.18 mg MDA/kg meat [38].

Bread

Studies focused on the effects of medium-chain triacylglycerols on the Maillard reaction during bread baking. In the experiment, different fatty acids were added at the rate of 0.2 mg/kg during the preparation of bread dough. The level of AGE formation increased as unsaturation in fatty acids increased, such as 18.7 percent (C8:0), 35.2 percent (C12:0), 38.2 percent (C18:0), 68.7 percent (C18:1), 80.1 percent (C18:2), 80.8 percent (C20:4), and 89.8 percent (C22:4) compared to bread dough without added fatty acids. These findings indicate that the oxidation of unsaturated fatty acids is more likely to enhance the Maillard reaction [39].

Coffee Beans

Roasted coffee beans are a source of acrylamide. In coffee, the major fatty acid linoleic acid, along with palmitic acid, is oxidized easily during roasting [40, 41]. These oxidized fats help in the formation of acrylamide. The influence of lipid oxidation on the formation of acrylamide in coffee during roasting was studied. *They found that lipid oxidation products such as 2-octenal, 2,4-decadienal, 2,4-heptadienal, 4-hydroxynonenal, and 4,5-epoxy-2-decenal concentration increased steadily and linearly as coffee beans roasting progressed [42].* This linear relation after some roasting time indicates that this compound acts as a carbonyl compound, participating in the Maillard reaction and contributing to acrylamide formation.

Model System

The effect of lipid oxidation on the formation of CML in a model system was studied. Researchers found a higher amount of CML in a protein–glucose model system compared to the protein–linoleic acid system. These results indicate that glucose is a major precursor of CML formation compared to linoleic acid. In the model system containing protein–glucose–linoleic acid, the CML content was higher than in the protein–glucose system followed by the protein–linoleic acid system. The model system consisting of protein–linoleic acid–sodium nitrite showed the lowest CML content among all, which suggests that the presence of lipid enhances CML formation, but sodium nitrate inhibits it [43]. The contribution of linoleic acid to the formation of AGEs in model systems during heat treatment was examined. They compared the model system containing glucose–lysine with glucose–lysine–linoleic acid and glucose–lysine–stearic acid. The level of AGEs was estimated by measuring the *AGE-fl* using a fluorescence spectrum on a Hitachi FL-4600 fluorometer. The results showed that AGE formation was higher in the glucose–lysine–linoleic acid system than in the other two model systems [44].

EFFECT OF MAILLARD BROWNING ON LIPID AUTOXIDATION

Oxidative stress in lipids leads to detrimental changes in food flavor, taste, and appearance, thereby shortening the shelf life of food products. These defects in food can be mitigated with the help of antioxidants. The Maillard reaction produces compounds (e.g., hydroxyl, superoxide, peroxyl radicals, pyrrole, and other reductones) with antioxidant activity. MRPs possess antioxidant activity through hydrogen or electron donation (reducing power) and metal ion chelation. The reducing power of MRPs has been reported in multiple ways. Reductones can interrupt the radical chain reaction by donating a hydrogen atom. Amadori compounds and advanced glycation end products act as electron donors [45]. The Maillard reaction changes the structure of glycated protein and exposes amino acids (e.g., tryptophan, valine, methionine, tyrosine, and phenylalanine) with electron-donating abilities [46]. The metal chelating ability of MRPs is due to compounds such as pyrrole and MRPs containing hydroxyl, ketone, and thiol groups, as well as anionic melanoidins and heterocyclic products [47, 48]. Other antioxidant activities comprise hydroxyl radical scavenging, superoxide anion radical scavenging, and oxygen radical absorbance capacity.

Food

The generation of MRPs from whey protein powder increases with gamma irradiation in the range of 0–20 kGy. MRPs derived from whey protein powder exhibited hydroxyl and superoxide anion radical scavenging activity, along with iron-chelating abilities. This antioxidant activity was evaluated using DPPH and β-carotene assays [49]. The impact of different concentrations of MRPs on lipid oxidation in PUFA-enriched cooked beef patties was investigated. Researchers tested a range of MRP concentrations (0, 0.5, 1, 2, 3, and 5 percent) and observed clear antioxidant effects. For instance, after 15 days of storage, the sample without any antioxidant had a peroxide value of 20.55 mmol O2/kg. Interestingly, when 3 percent MRPs were added, the peroxide value dropped markedly to 3.51 mmol O2/kg. This corresponds to an 83 percent reduction in hydroperoxide formation, indicating strong inhibition of primary oxidation. Furthermore, they noted that the TBARS value, which reflects secondary lipid oxidation, was reduced by 85 percent at the 3 percent MRP level compared to the untreated control. These results suggest that MRPs, particularly at a 3 percent concentration, are highly effective in controlling lipid oxidation in meat systems enriched with polyunsaturated fatty acids [50].

Model System

A model system consisting of histidine and glucose was used to determine the antioxidant activity of MRPs. They found that heating the mixture at 120 °C for 10, 20, and 30 minutes led to a significant increase in antioxidant activity due to the formation of MRPs. Beyond 30 minutes of heating, a reduction in antioxidant activity was observed, likely due to the degradation of MRPs [51]. MRPs produced from basic amino acids exhibited higher antioxidant activity than those derived from neutral, aromatic, sulfur-containing, and acidic amino acids in a methyl linoleate model system. They found that MRPs formed from glucose + lysine and glucose + glycine showed effective antioxidative potential when prepared by heating at 100–110 °C for 2 hours [52]. The antioxidant activity of Maillard reaction products (MRPs) generated from a glucose–casein model system was evaluated using a sunflower oil-in-water emulsion. MRPs formed at 90 °C for 12 h demonstrated the lowest peroxide value, indicating effective inhibition of primary oxidation. Furthermore, these MRPs showed a strong ability to suppress secondary oxidation, with TBARS inhibition ranging from 39.05 percent to 88.66 percent. These glucose–casein derived MRPs exhibited superior antioxidant potential compared to the synthetic antioxidant TBHQ (0.02 percent) [53].

CONCLUSION

Findings from meat, bread, coffee beans, and model systems suggest that lipid oxidation does not end with the formation of secondary and tertiary oxidation products. In the presence of amino compounds, these lipid oxidation products (reactive carbonyls) further participate in reactions to produce carbonyl–amine compounds, such as MRPs. The presence of oil and its oxidation products accelerates the rate of the Maillard reaction by providing additional reactive intermediates and enhances the production of CML and CEL. Consequently, the Maillard reaction and lipid oxidation are not isolated processes but are interconnected through shared intermediates and reaction mechanisms. This interaction should be integrated into a general carbonyl reaction framework that can be initiated by either lipids or carbohydrates. On the other hand, Maillard reaction products also exhibit antioxidant properties, which have been evaluated using DPPH radical scavenging activity. The results demonstrated that antioxidant activity increased in parallel with the browning intensity of the samples, indicating that MRPs can influence lipid oxidation dynamics. These results highlight that lipid oxidation and the Maillard reaction are closely interconnected and both should be considered together to fully understand the formation of Maillard reaction products in the presence of lipids and vice versa.

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