Chapter 3
Undesired Chemical Alterations and Process-Related Causes. The Role of Thermal Control and the Management of Thermal Machines

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Abstract This chapter is explicitly dedicated to the description of several known sensorial alterations in foods and beverages because of technological preservation systems. Many possible alterations could be considered: the more diversified the choice of industrial processes (including also traditional preservation systems), the broader the field of interest. For this reason, authors have decided to consider the subcategory of sensorial failures in relation to thermally conducted processes only. Basically, the following chemical alterations are discussed in relation to thermal food processing: hydrolytic decomposition of fat molecules and proteins; ketonic and oxidative rancidity; homofermentative reactions; decomposition and modification of lateral chains in amino acids; protein denaturation; interactions between proteins and other organic molecules; enzymatic degradation of proteins; caramelisation of carbohydrates.

Keywords Denaturation · Enzymatic degradation · Food preservation · Hydrolysis · Mechanical process · Rancidity · Thermal dissipation

Abbreviations

UHT Ultra High Temperature
a_w Water activity

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3.1   Introduction to Chemical and Physical Modifications of Foods in the Modern World

One of the most interesting (and worrying) features of the modern food and beverage industry is correlated with the irreversible modification of certain chemical–physical parameters of produced foods. Actually, the variation of sensorial features in edible products is not a recent discovery (Cappelli and Vannucchi 1990); the same thing can be affirmed when speaking of microbiological alterations caused by the modification of edible 'culture media' (foods, beverages) after processing techniques (Parisi 2002, 2003). However, the current situation appears different if compared with the past: in other words, the increasing industrialisation worldwide has certainly modified the organoleptic representation of current foods and beverages (Parisi 2006, 2012, 2013) with partially forecasted results on the one hand and the concomitant confusion of selected brands and related features by normal consumers on the other side.

Anyway, many different chemical and physical modifications may be observed when speaking of industrial and artisanal foods and beverages. A preliminary discrimination should concern the frequency of certain variations on the chemical level, because an industrialised process is expected to modify intermediate edible masses from the start to the end of the whole 'chain' in a predictable way. In other words, the more standardised the process, the lower the amount of unexpected 'surprises' in the final product. On the other side, artisanal products—the heritage of old times and civilisations—are easily recognised as the carrier of many unexpected or unknown sensorial variations when discriminating between two or more similar products of different geographical or social origin (Gómez-Ruiz et al. 2002; Kupiec and Revell 1998; Tregear 2003).

By a general viewpoint, the industrial food product (Campbell 2009) is designed, produced and distributed with the aim of 'capturing' the attention of 'normal consumers' (Parisi 2004–2012) in a durable manner. For this reason, each product has to be obtained with the minor possible amount of sensorial variations when speaking of different production lots: different results could easily disorientate consumers. In fact, the simple and inevitable modification of sensorial features in food products is irreversible between the start and the end of the commercial life (within the so-called expiration or sell-by-date). Consequently, food technologists should compose their personalised idea of food or beverage product taking inevitable temporal variations of colours, aroma, taste, texture and shape into account. The occurrence of additional modifications has to be avoided. On the other side, artisanal foods are surely restricted in the modern world when speaking or market sales: the competition may be assured if a sort of shared organisation is put in place. Normally, these joint entities may assure elements such as legal protection of brands, communications, innovation and quality assurance (Jordana 2000). The last feature, quality assurance, should imply the good or acceptable knowledge of technological processes and related effects, including undesired alterations (or 'surprises').
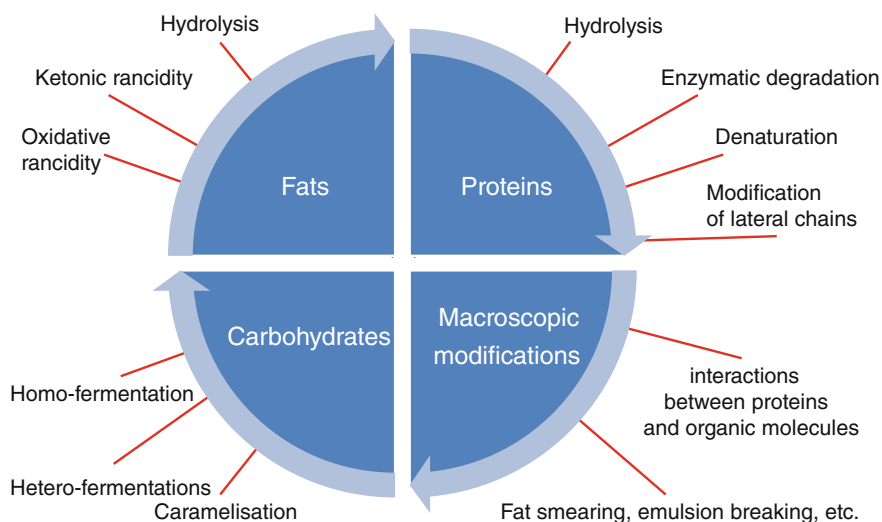
Food processing and related alterations

Fig. 3.1 A brief overview of chemical alterations during food processes. In spite of the possible influence of other causes, main chemical and physical alterations of foods and beverages are often originated during processing steps in the food industry, and during the remaining part of the whole food chain (storage, delivery and final distribution). Generally, the following procedures and reactions are considered when speaking of probable causes: controlled enzymatic reactions; controlled seasoning in open air; controlled homo- and hetero-fermentative processes; drying systems; mechanical processes; irradiation; high-temperature preservation; low-temperatures processes; MAP and CAS systems; semi-preservation (addition of salt, ethanol, sucrose, fatty oils, etc.); smoking treatments

This chapter is explicitly dedicated to the description of several known sensorial alterations in foods and beverages because of technological processes. Actually, the discussion is broad because many possible alterations could be considered: the more diversified the choice of industrial processes (including also traditional preservation systems), the broader the field of interest. For this reason, authors have decided to consider the subcategory of organoleptic failures or 'surprises' in relation to thermally conducted processes only (Fig. 3.1). In other words, the discussion does not concern chemical, physical, microbiological, sensorial alterations and synergistic results caused by processes without a clearly correlated thermal augment.

In fact, the following processes or systems may be taken into account when speaking of undesired food or beverage alterations (Cappelli and Vannucchi 1990; Delia et al. 2005; Fessmann 1999; Horner 1997; Micali et al. 2009; Parisi 2009; Rahman and Perera 1999; Šimko 2002):

- Controlled enzymatic reactions
- Controlled seasoning in open air (presence of selected microorganisms)
- Controlled seasoning in open air

- Controlled homo- and hetero-fermentative processes
- Drying systems including cryostatic processes, membrane- and microwave-assisted technologies and freeze-drying
- Mechanical processes (kneading, lamination, crushing, screwing, etc.)
- Radiation techniques
- Thermal systems with preservation purposes, high temperatures: pasteurisation, sterilisation, etc.
- Thermal processes with preservation purposes, use of low temperatures: refrigeration, freezing, etc.
- Thermal processes without preservation purposes, use of high temperatures: cooking, melting, etc.
- Other preservation techniques with the use of active systems and/or modified gases as food-contact atmosphere
- Chemical systems for the preservation of foods: addition of salt, ethanol, sucrose, fatty oils, etc.
- Smoking treatments for preserving foods and similar techniques without preservation meaning, also named 'aromatisation' procedures.

The above-mentioned systems are the basis of the modern industrial production when speaking of foods or beverages. Each processing step or sub-step has its own reason or reasons (advantages) in the ambit of the whole food process. On the other side, the occurrence of several defects may be ascribed to the same group of systems.

In general, the following chemical alterations should be discussed in relation to food processing and other causes:

(a) Hydrolytic decomposition of fat molecules, catalysed by microbial lipases, light exposure and water (Holliday et al. 1997; Macrae 1983; Sherwin 1978)
(b) Ketonic rancidity. Enzymatic reactions are involved; mould contamination is needed (Hamilton 2003; Kinderlerer 1993; Kinderlerer and Kellard 1984)
(c) Oxidative rancidity, catalysed by light exposure, presence of metals such as iron, copper, nickel and other metallic elements, thermal dissipation during mechanical processes, presence of peroxides and lipoxydases (Aluyor and Ori-Jesu 2008; Kochhar 1996; Mizutani and Hashimoto 2004; Tan and Man 2002)
(d) Homofermentative reactions (Marshall and Tamime 1997) with the production of ethanol, and hetero-fermentative processes (Pilone et al. 1991; Priest 1999) with these final products: ethanol, isopropyl alcohol, butyl alcohol, acetone, etc. These reactions are naturally catalysed by different microorganisms such as *Lactobacillaceae*, yeasts and moulds
(e) Decomposition and modification of lateral chains in amino acids (Stadtman and Levine 2003)
(f) Protein denaturation (Hendrickx et al. 1998; Lopez-Fandiño et al. 1996; Messens et al. 1997)
(g) Different interactions between proteins and other organic molecules. 'Maillard reaction' is one of the most known and studied examples (Fu et al. 1994; Martins et al. 2000)

(h) Hydrolysis of proteins (Sarmadi and Ismail 2010)
(i)   Enzymatic degradation of proteins (Cappelli and Vannucchi 1990)
(j)   Caramelisation of carbohydrates (Claude and Ubbink 2006; Friedman 1996; Izydorczyk 2005)
(k) Chemical–physical modifications such as fat smearing and breaking of emulsions (McCain 1991).

From the sensorial viewpoint, many of the above-mentioned reactions determine important variations. Generally, colours are modified in an irreversible way: the caramelisation and other reactions with carbohydrates as substrates are the cause of brown colours in foods (Cappelli and Vannucchi 1990). Moreover, the following chemicals are modified and/or destroyed:

– Vitamins. In particular, vitamin C, β-carotene and thiamine are progressively destroyed (Morris et al. 2004) because of oxidation, contact with water, and thermal processes such as blanching (vegetable products)
– Anthocyanins (Jackman et al. 1987; Patras et al. 2010)
– Carotenoids (Britton and Khachik 2009; Rodriguez-Amaya 1997).

The physical looking of foods and beverages can be notably modified: as an example, textural variations in several products such as certain cheeses and industrial pasta have been considered in the scientific literature and studied with the aim of providing better products (Parisi et al. 2006). Anyway, the reduction of hydric contents is one of the most known effects when speaking of textural variations and correlated effects (Barbieri et al. 2014). Certain superficial modifications, including also the simple aspect of products (example: geometrical shape) can be also observed and mathematically correlated with chemical–physical parameters. Finally, the aroma may be remarkably modified with pleasant or unpleasant results, depending on the process; naturally, alterative phenomena (degradation) can also determine the premature perishability of certain foods or beverages with claimed long durability. Should this situation be observed, production/storage/delivery steps and the distribution process (or the sum of independent and subsequent sub-steps in the food chain) would be studied carefully.

Anyway, the sum of possible modifications is very large and this book could not consider all options here. For this reason, chemical–physical modifications are discussed here if some food processing relationship is involved and the process tends to the increase of thermal values. In addition, some reflection is made here with concern to the importance of thermal control and the management of involved machinery and equipments in the modern food industry.

In general, the following list can be exhaustive enough when speaking of thermal augment:

• Mechanical processes with heat dispersion (lamination, extrusion, kneading, grinding, crushing, screwing, mixing, etc.)
• Thermal systems with preservation purposes, use of high temperatures: pasteurisation, sterilisation, etc.

- Thermal processes without preservation purposes, use of high temperatures: cooking, melting, etc.

It should be noted that mechanical processes are not carried out with the aim of increasing thermal values of intermediate food masses. On the other hand, the simple mechanical movement determines a certain energy dissipation because of compression, impact and friction forces, with the consequent heat dispersion into working masses (Hernandez-Izquierdo and Krochta 2008; Zardetto and Dalla Rosa 2006). This heat dissipation is usually observed in grinding operations on dry foods (coffee, cocoa, etc.), although other sectors such as meat processing industries may show similar problems (McCain 1991). For this reason, mechanical processes should be considered in this ambit despite the low impact in terms of thermal increase. On the other side, it has to be considered that thermal increases may become notable during the process with the consequent melting of grinded particles. Moreover, heat exposure of initial raw materials (rice) during grinding has been reported to have apparently some influence on the quality of finished products (Nishita and Bean 1982). Cryogens such as carbon dioxide snow are often used with the aim of reducing thermal augments.

3.2 Undesired Chemical Alterations and Thermal Processing Management

In general, each food processing technique has one, two or three distinct aims:

(a) The safe consumption of foods or beverages
(b) The easy preservation of claimed long-durability foods and beverages
(c) The increase of palatability, for peculiar foods (Fujimoto et al. 2015; Lepper 2013).

Basically, all thermal treatments including the simple cooking process can determine the following situations (Cappelli and Vannucchi 1990):

(1) Reduction of hydric amounts (in other words, augment of dry content in treated foods)
(2) Hydrolysis of proteins, carbohydrates and fatty molecules
(3) Inactivation of many enzymes (microbial origin) and toxins, elimination of a notable part of pathogen microorganisms and reduction of the total viable count of microbial agents in treated foods
(4) Demolition of certain amino acids, thermally unstable vitamins and organic compounds (proteins and carbohydrates) because of different reaction chains (Maillard reaction, etc.)
(5) Aqueous dissolution of mineral salts in the excess water (after treatment) with consequent diminution of nutritional values.

Therefore:

(a) Each thermally treated food loses a variable amount of the original weight, depending on its composition, treatment conditions (temperature and time cycles), and the dimension of treated foods and 'cooking' chambers

(b) Certain colorimetric variations can be easily observed because the original aqueous solid solution (the initial food) has been physically concentrated after treatment. As an example, the variation of colours can be easily ascribed to certain fatty molecules without the diluting effect of dissolving water

(c) The digestibility of main nutritional molecules—carbohydrates, proteins and fats—is notably increased, in spite of the presence of certain indigestible fractions

(d) Sensorial features are remarkably enhanced after treatment

(e) The nutritional value may be increased after treatment when speaking of available proteins, fats and carbohydrates; on the other side, vitamins and other nutritive principles are lost

(f) The presence of mineral salts may be reduced after treatment because of the dissolution in the existing (dissolving) water and/or in added cooking waters. This modification may be observed with the concomitant variation of chemical–physical properties of certain macromolecules such as proteins.

The category of thermal processes involves many techniques, and cooking is only the first and best known of these methods. With relation to the industry of vegetable foods, the 'blanching' process (cooking in boiling water, followed by the sudden 'shocking' step under cold water) can be extremely useful (Abu-Ghannam and Jaiswal 2015; Menon et al. 2015). A notable part of thermal processes are carried out with the aim of destroying microorganisms and inactivating toxins with a reliable degree of food safety: pasteurisation, sterilisation and related variations such as ultra high temperature (UHT) processing are widely used in the modern food industry.

Because of the importance of involved parameters on the management of thermal processes and related machinery, a brief description of the above-mentioned processes has been shown in Chap. 2.

However, the main objective of this chapter is the definition of undesired chemical and physical modifications in foods in strict relation to temperature, the main process parameter. For this reason, the above-mentioned modifications are discussed in Sect. 3.3 with reference to thermal values, where available. Before continuing, a little premise should be made with concern to mechanical treatments (kneading, lamination, crushing, screwing, etc.) and their possible effects on undesired chemical reactions.

3.3 Thermal Processing Parameters and Chemistry of Foods

3.3.1 Reduction of Hydric Amounts and Possible Damages After Cooking

Generally, the reduction of moisture in edible products is desired because the microbial spoilage is direct function of liquid water amounts. Actually, the bioavailable water corresponds to a (notable) fraction of the whole water amount that could be analytically determined. This fraction, named 'free water', is generally measured in an indirect way by means of another parameter: water activity (a_w). Moreover, many properties of foods—rheological behaviour, hygroscopicity, etc.—depend strictly on the physical state of water, and the aqueous solvent is not always liquid.

Normally, a_w for a selected food corresponds to the relative humidity of the air above the food surface, on condition that the food is really in equilibrium with the atmosphere. The basic relation is:

$$a_w = \frac{P_0}{P} \tag{3.1}$$

where P_0 is the vapour pressure for the food (a solution!) and P corresponds to the vapour pressure of pure water at the temperature of measurement (Walstra 2003). The importance of a_w values is correlated with the necessity of eliminating or reducing the microbiological risk, in the ambit of food safety measures (Delia et al. 2005; Ottaviani 2002; Parisi 2002). Anyway, the limit for a safe preservation of foods without the addition of chemical additives (as dissolved substances) or thermal treatments such as pasteurisation corresponds to $a_w \leq 0.65$ (Cappelli and Vannucchi 1990). Below this value, only saccharophilic yeasts could colonise foods.

Apart this consideration, it has been reported that normal reactions in foods have a different speed (in terms of kinetics) depending on the water content. In other words, the lower the amount of available water, the lower the speed of many possible reactions is; including biochemical processes (Walstra 2003). The basis of this reflection cannot be simple because many different subprocesses of different (chemical, physical, biological) origin can occur at the same time with different 'weights'.

Anyway, the reduction of moisture (this quantity is generally different from 'water') in foods is normally desired. With exclusive reference to thermal processes with strong augments of temperatures, it can be affirmed that the most used method concerns evaporation (Cappelli and Vannucchi 1990). In detail, the higher the removal of water molecules from a food product, the higher the quantity of produced (and consumed) heat for this work (Walstra 2003). The continuous removal of aqueous content becomes more and more difficult if a_w gets lower (and sorption

enthalpy increases). This phenomenon depends on the direct relationship between the remaining amount of water and the low a_w value, and the necessity of supplying continually sorption heat (also intended as sorption enthalpy) and the additional enthalpy of evaporation[1] during the process. For these reasons, drying processes are easier (and get good yields) at increasing temperatures (Cappelli and Vannucchi 1990; Walstra 2003).

The direct reduction of water can be performed by means of direct or indirect systems. Briefly, it can be affirmed that direct techniques concern the use of electric resistances and low pressures (under-vacuum processes), while indirect systems use heat exchangers and additional machinery (placed into reactors) for increasing heat diffusion in the mixture or intermediate mass. The problem of these processed concerns thermally sensible fluids (orange juices, milk, etc.). For this reason, many processes can be subdivided in different steps where the working pressure is constantly reduced with the aim of reducing evaporation temperatures until 40 or 50 °C. However, thermally sensible products cannot be treated in gradient processes: the reduction of evaporation temperatures has to be reached immediately (one step only). The diffusion of heat can be observed as the result of four different mechanisms: conduction, convection, radiation and distillation (Walstra 2003).

Apart chemical damages, the reduction or moisture can affect the textural appearance of products, but this property depends also on the composition of dry residues: proteins, fats and carbohydrates. Other physical modifications concern the rheology of intermediate foods and beverages, and the possible hygroscopicity. With concern to hygroscopic powders, the observed phenomenon corresponds to the normal tendency of the 'food/external atmosphere' system to a new equilibrium, because of the unbalanced ratio between a_w values (air has a higher a_w than foods). Consequently, the problem should be managed in terms of good (or bad) storage in dry warehouses.

Finally, some reflection could be made with relation to several defects (microbial spoilage is not discussed here) without direct connection to water losses. The removal of water produces a more rigid mass with peculiar properties. As a single example, the following defects have been reported with relation to poultry foods (Sams 2001):

(a) Abnormal emersion of fatty substances in certain meat products because of the excessive amount of fats and collagen in the raw materials or because of excessive heating temperatures (normal end-point values: 68.3–73.9 °C). Fat globules break the complex fat/protein matrix
(b) Possible discoloration in emulsified products because the minimum temperature (end-point: 68 °C) has not been reached.

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[1]This enthalpy is really higher than sorption heath during the process.

3.3.2 Hydrolysis of Proteins, Carbohydrates and Fatty Molecules

Proteins can be easily hydrolysed under high-temperature processes such as cooking; naturally, pasteurisation (temperatures under 100 °C) and sterilisation (example: more than 3 min at 121 °C) procedures may enhance the hydrolysis. In general, this reaction is a good result because of the increase of digestible proteins; anyway, the amount of hydrolised proteins depends strongly from the acidity of food masses.

Fat molecules can also be hydrolised—the industrial procedure is applied at 250 °C (pressure: 50 bar)—but the initial decomposition in glycerol and fatty acids is unfavourable: the obtained glycerol is subsequently decomposed again with the production of undesired acrolein and two water molecules (Cappelli and Vannucchi 1990). Moreover, several fatty acids may be detrimental when speaking of food palatability.

With relation to carbohydrates, starch molecules can be partially hydrolised in absence of water (temperatures should be around 160 °C) with the production of dextrins; following reactions may also produce single maltose molecules, with a notable augment of digestibility. For this reason, the hydrolysis of carbohydrates in foods does not seem important as risk factor.

3.3.3 Demolition of Nutrients. Maillard Reactions and Other Mechanisms

The action of high temperatures may destroy or modify certain amino acids with important effects. One of the most important and studied mechanisms concerns the transformation of cysteine and cystine residues at temperature >100 °C with the production of hydrogen sulphide. Actually, this simple reaction is part of the sum of reactions concerning the vast ambit of denaturation phenomena. Because of the rupture of disulphide bonds in certain proteins, the production of the typical 'rotten eggs' smell highlights the presence of amino acids rich in sulphur (Parisi 2012).

Other situations concern

– The modification of nitrogen-containing amino acids such as asparagines and glutamine with production of ammonia; modified amino acids may also form covalent bonds (Cappelli and Vannucchi 1990)
– Serin modification with the loss of one water molecule
– The oxidation of methionine to methionine sulphoxide (Narayan 1997)
– The production of certain mutagenic heterocyclic amines may be produced under cooking (temperatures >200 °C) when the presence of tryptophan is observed (Sugimura 1985).

The Maillard reaction can be also observed: this reaction starts as the initial condensation of glucose and a protein with the production of Schiff bases; after this step, a number of compounds can be irreversibly obtained with different (parallel and consecutive) reactions. These products include furfural, different aldimines and ketimes, and the group of brown-coloured melanoidins. The Maillard reaction is extremely complicated and the discussion could not be easily carried out in this Chapter. In addition, the influence of temperature values may not be simply expressed with the Arrhenius equation, but with the basic Eyring equation (Martins et al. 2000; Walstra 2003) containing activation enthalpy, entropy and Gibbs energy terms. In addition, it has been reported that the reaction between proteins and reducing carbohydrates may be observed at room and very low temperatures (Narayan 1997). For this reason, the complex of Maillard reactions is not further discussed here.

Moreover, proteins can easily interact during high-temperature processes with the formation of solid cross-linked matrices (Sams 2001): one of the most known examples is the gelation of myofibrillar proteins in processed poultry products.

Finally, it should be remembered that

(a) Mineral salts are easily dissolved into water; because of the removal of the aqueous solvent, the diminution of certain metals (calcium, etc.) can compromise the stability of partially bonded macromolecules such as proteins, with the consequent coagulation and precipitation

(b) The amount of vitamins is notably reduced under drastic conditions; for this reaction, oxidation can easily attack fatty molecules after treatment. In addition, the reaction between certain food proteins and oxidised fatty acids has been recently reported (Narayan 1997). The problem is not the expected oxidation of fatty acids after the destruction of natural antioxidants (vitamins, etc.), but the production of strongly bound complexes between these fatty acids and proteins. The sum of radicalic, cross-linking, oxidation and transfer reactions has been observed to produce similar complex at 90, 60 and 30 °C too (Narayan 1997). Apparently, the role of protein radicals is important; consequently, the reduction of protein damages in high-temperature (100 °C and above) processes seems crucial.

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