

Microstructure, composition and their relationship with molecular mobility, food quality
and stability

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ABSTRACT

Food stability is a critical parameter for both consumers and producers, since it assures safety, nutritional and sensorial quality of foodstuffs, and at the same time maximizes shelf-life.

For long water activity, a_w , was considered a determinant parameter in food stability and physical properties. This concept was challenged with the revolutionary approach to the study of food systems using the glass transition concept. Recently, scientific research suggests that molecular mobility is a fundamental approach to fully attain food physical properties and stability. Current literature suggests that stability can only be fully grasped if molecular mobility and structure are taken into consideration, i.e. an appropriate understanding of the behavior of food products requires knowledge of its composition, structure and molecular dynamics, through the three-dimensional arrangement of the various structural elements and their interactions.

Food systems are complex mixtures of water, biopolymers, low-molecular weight ingredients, and colloid particles, and the molecular mobility between these different components reflects on the stability of such systems, determining the physical state, microstructure and composition, which impacts on food characteristics.

Particularly, food water content, location and interactions with other components are critical in microbial growth, degradation reactions and sensorial aspects. Understanding changes in water location and mobility represents a significant step in food stability knowledge, once that water “availability” profoundly affects chemical, physical and microbiological quality of foods.

Key words: Food stability, Microstructure, Molecular dynamics, Water availability, Sensorial quality, Safety, Bioavailability

1. Introduction

Food stability and also nutritional and sensorial quality was always a critical parameter for different stakeholders. With respect to industry, stability allows maximizing shelf-life, and simultaneously minimizes waste along the distribution chain, increasing profit and reducing the environmental impact [1-5]. For consumers, stability assures safety, nutritional and sensorial quality of food products and answers to the increasing demand for a diversity of ready-to-eat food with fresh appearance and health-promoting properties [5, 6]. Nowadays, consumer's expectations from a food product are even more critical. Desirable sensory perception (liking) and healthy components at a reasonable cost are mandatory conditions. The current desire to alter food composition for health concerns (reduction in salt, fat, and calories, and increase in bioactive compounds) has brought to light the challenges involved in altering composition and maintaining consumer acceptance [7].

Since long time that scientists believed that by controlling a_w was determinant to assure all these food properties [4, 5]. Meanwhile, other concepts, such as glass transition temperature, have emerged as important parameters to be taken into consideration [5, 10, 11]. Nevertheless, current literature suggests that food quality and stability can only be fully grasped if microstructure and molecular dynamics are taken into consideration, i.e. an appropriate understanding of the behavior of food products requires knowledge of its composition, structure/ microstructure and molecular dynamics, through the three-dimensional arrangement of the various structural elements and their interactions [8].

Literature studies demonstrate that food microstructure contributes largely to sensorial and nutritional food quality and also its stability (Figure 1) [8, 12-17].

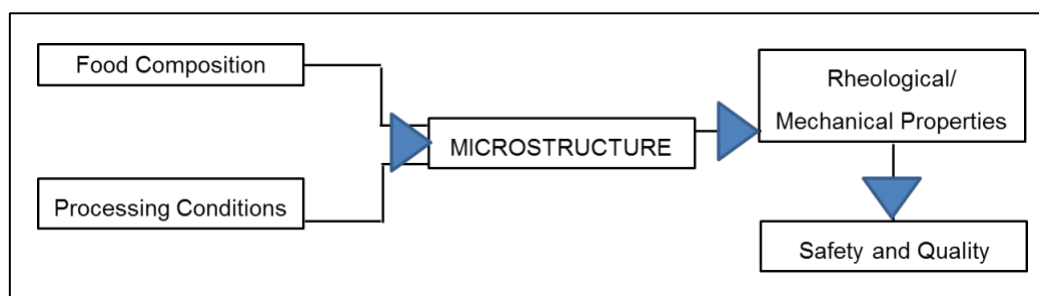


Figure 1: Interrelationship between composition and processing on microstructure formation in foods and its quality attributes.

Microstructure recognizes that foods are highly structured and heterogeneous materials, and the types of such structural units and their interactions are decisive for the food physical behavior and functional properties, such as texture or sensorial attributes, and also physical and chemical stability during storage [5]. Research studies recognize the association between food structure and functionality, and that this is a critical point for a proper control of food quality and safety aspects [12]. This means that several of the apparent features of food, such as mechanical and electromagnetic properties, mass and heat transfer, as well as sensorial, nutritional and safety quality are strongly dependent on their complex microstructure [8]. Also, the majority of food elements that critically participate in transport properties, physical and rheological behavior, textural and sensorial characteristic are below the 100 μm range [8]. This causes that further improvements on the quality of existing foods, and the creation of new products to satisfy expanding consumer's demands, should be largely based on interventions at a microscopic level.

Designing the food structure during processing can also affect the behavior during shelf-life. By physically separating the reactants in microstructural locations can for example control the biochemical activity by avoiding the reactants to be in contact, thus minimizing the development of off-flavors and browning reactions [5, 8]. Food microstructure can also be altered by controlling various intermolecular and inter-particle interactions among the different ingredients during processing and storage. This can be considered an important topic in different fields of food science and engineering, such as for the exploitation of food-grade delivery systems where the development of quantitative structure-function relationships is of utmost importance to develop rational design and efficient production of such systems [18]. Engineering structures require knowledge on the molecular organization of the ingredients (short- and long-range molecule assemblies) and physical properties, such as charge density, hydrophobicity, molecular size and conformation under different environmental conditions [19]. The expression "structure–function", nowadays widely used, describes basically the way in which physicochemical and functional properties of foods are related to their structure [12].

This chapter aims at reviewing some of the main aspects related to microstructure and composition and its interference in food matrices molecular dynamics, quality and stability.

2. Structural elements in foods

As was mentioned above, an appropriate understanding of the behavior of food products requires knowledge of its structures/ microstructures [17]. These structures are based on different structural elements, such as water, biopolymers (proteins and polysaccharides) and low molecular weight ingredients, oil droplets, fat crystals, granules, strands, micelles and interfaces, with various spatial arrangements and different interactions. These structural elements, in various combinations and proportions, can exist in different states (glassy/rubbery/crystalline and solubilized) even at uniform temperatures and water activities, which will necessarily affect the macroscopic food quality attributes and behavior along the storage period [5, 20]. These highly structured and heterogeneous elements influence the water/solute interactions and hence the water availability to participate in microbial growth and degradation reactions [12]. In fact, these intermolecular dynamics in which the water molecules play a very important role can determine the structure of the food material at the beginning of a given process and during processing [21]. Also, the effective water diffusivity in foods, as well as free water content, highly depend on pore structure or particle size distribution [5, 22-24]. Figure 2 shows some food structural elements and relevant length scales.

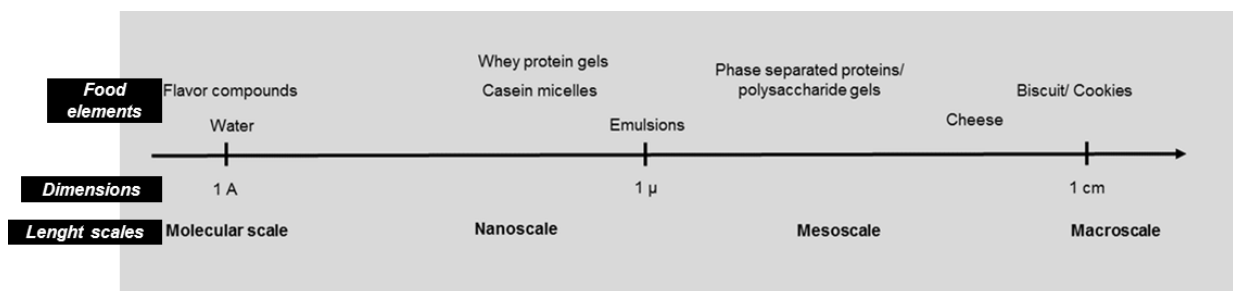


Figure 2. Examples of structural elements on foods with corresponding relevant length scales (adapted from [7]).

In order to obtain the highest quality food products, controlling the formation of structural elements can be a crucial step for determining final quality and shelf life. The formation of structural elements, and consequently the structure/ microstructure of a food product, can provide the desired rheological properties of the food, like hardness, stiffness or snap, and contribute to specific organoleptic properties, such as melt-down rate, cooling effect, among others. Zhao et al. (2017), for example, reported the closely relationship between hardness and microscopic structure of acetic acid pretreated and non-pretreated cooked potato slices [25]. The microstructural elements impact shelf life of many food products. Chocolate is a classical example, where the formation of bloom (i.e. the white haze that sometimes appears on chocolate) is related to a recrystallization event where cocoa butter crystals transform from one polymorph to another [26].

3. Molecular dynamics, microstructure and stability

As mentioned before, several of the food effective features are strictly dependent on their complex microstructure. Several macroscopic properties are also controlled by microscopic elements. By restricting the mobility/ dynamics of reactants, as well as separating those in different compartments, it is possible to avoid chemical or biological degradation reactions [10, 27], This is the case when foods are in glassy state, which is characterized by a very high viscosity, where physical and chemical stability abruptly increases as a consequence of the molecular mobility/ dynamics reduction.

Actually, it is generally accepted that an appropriate food behavior understanding requires not only knowledge on its composition, but also on its microstructure and molecular dynamics, through the three-dimensional arrangement of the various structural elements and their interactions [5].

Molecular mobility/ dynamics have been pointed as the actual most promising strategy for characterizing multicomponent systems, like foods. Analysis of systems at a molecular scale have demonstrated to be an useful methodology for investigating complex geometries and molecules, as well as studying structural and dynamic properties [21].

Moreover, the knowledge of molecular dynamics is determinant for assessing physicochemical and microbiological stability of food systems and is dependent both on composition and microstructure [28, 29]. In literature it is possible to find some examples that related molecular dynamics, with food stability and microstructure. Fundo et al. (2016) evaluated the relation between water molecular dynamics, measured in terms of transverse relaxation times with a NMR methodology, and fresh-cut pear firmness and microstructure. This study reported that the cell wall degradation together with cell structure alteration/ loss (e.g. sclereids spreading along the matrix), both observed along the storage period by microscope images, allows firmness modifications with impact on free volume and on the leakage of cellular osmotic solutes into the apoplastic space, which then result in altered water mobility availability [30].

3.1. Aspects of water molecular dynamics

Water is probably one of the most important food components in impacting food physicochemical and microbiological attributes, shelf-life and deteriorative changes [1, 10, 24, 31, 32]. Water is considered as the most important solvent, dispersion medium and plasticizer in biological systems, like foods [33]. It affects reactions, can be substrate and a product of reactions, and is involved in nutrients transport and dissolution of salts and other solutes. It establishes pH, acts as a polymer plasticizer and modulates viscosity or osmotic pressure [34]. Therefore, determination of water content is one of the most frequent analyses in the food industry laboratories [32]. However, various foods with the same water content differ in stability [35], which demonstrates that the sole value of “water content” in a food does not inform about the nature of water [5, 32, 35]. In fact, in a food matrix water molecules can be available or not to participate in degradation reactions [32]. The knowledge of each of these fractions is important, specifically because available water, its location and the interactions with the other food components (like proteins and polysaccharides) are responsible for the physicochemical and microbiological properties and stability of foods [31, 33]. As such, besides water content in a food material, it is important to understand the water state and dynamics for proper comprehension of properties and stability of food products.

Water mobility/ dynamics can thus be described as a manifestation how freely water molecules can participate in reactions or how easily water molecules diffuse to reaction sites to participate in reactions [5, 36]. Both conditions are profoundly related with food products matrices. Presence of molecules of different molecular weight and solubility in water can have a marked influence on water mobility/ dynamics, as this is dependent on the physiochemical properties of other non-aqueous food constituents and their interactions with water and among themselves [36, 37]. In this way it is possible to infer that, in a food matrix, water dynamics is dependent on matrix microstructure but can also be responsible for its development.

3.2. Measuring water dynamics

Nuclear magnetic resonance spectroscopy (NMR) has evolved to become a powerful tool to show the structure and dynamics of food constituents in solid state. NMR is recognized as one of the main analytical methodologies that gives a complete view of the foodstuffs metabolites and, together with suitable statistical analysis, provides relevant information in terms of food quality, processing and safety.

Despite of the fact that this technique is not based on image reproduction and analysis, it may be very useful in estimating physicochemical changes and understanding the structures and dynamics of complex macromolecular systems [38].

Specifically, ^1H NMR has been used to investigate water dynamics and physical structures of foods through analysis of nuclear magnetization relaxation times [5, 39]. In NMR, samples are submitted to a static magnetic field and the protons are excited by means of a radiofrequency pulse. The analysis of the signal emitted while the samples return to equilibrium (FID) allows the determination of spin-lattice (T_1) and spin-spin (T_2) relaxation times. This later variable is related with the mobility of the protons in the sample matrix [5].

Foods and biological materials consist largely of water and macromolecules rich in protons. Since water protons are major contributors to the proton relaxation, the interactions between water and macromolecules represent the most important factors affecting the proton relaxation process [40]. Thus, the application of NMR technique may be very useful in predicting physicochemical changes and understanding structures and dynamics of complex macromolecular systems like foods in solutions and/ or in solid state [41].

4. Relevance of food microstructure design for achieving nutritional and sensorial quality

Food microstructure is an essential parameter that must be taken into consideration for designing and developing healthier food products with improved sensorial characteristics and stability [14]. The knowledge on food microstructure aspects is crucial for food scientists and engineers, since it can be related with specific aspects of food consumption, such as nutrients bioavailability, flavor release or texture perception.

4.1. Nutrients bioavailability

Nowadays, one of the major goals of food science is to assess the nutrient bioavailability and how much of an ingested nutrient is efficiently absorbed by the human body. Food nutrients are often placed in natural cellular compartments or within assemblies produced during processing, and they need to be released during digestion so they can be absorbed to the gut [42].

Several studies have shown evidences that food matrixes or food microstructures, and also the interactions between food components, play a major role in the release and bioavailability of several nutrients and allergenic substances [13,16, 42-47]. As general examples, there are studies that showed that food matrix components, such as fiber, could decrease carotenoid absorption [44], others found good bioavailability of ferulic acid in the presence of bran [13]. There are also evidences that food microstructure influences the bioavailability of some minerals [45, 46]. Figure 3 presents different food matrices that are able to affect some nutrients bioavailability.

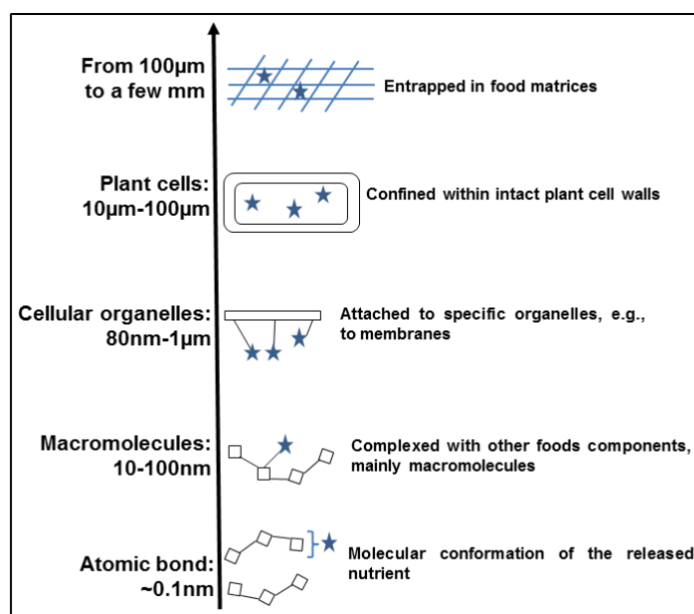


Figure 3. Different mechanisms that can disturb nutrients (★) bioavailability in different food microstructures (adapted from [42]).

As an example [16], a study investigating the relationship between carotenoids bioaccessibility and the structural organization of plant tissues. This work was concentrated on β -carotene and lycopene bioaccessibility in plain carrot and tomato based samples, in decomposed carrot and tomato samples of different sizes, and in carrot and tomato based fractions having various amount of structural barriers surrounding the carotenoid. It was possible to identify an inverse correlation between food structure integrity and carotenoid bioaccessibility. Moreover, it was possible to point out that the chloroplast structure and the cell walls are the major barriers for carotenoids bioaccessibility in these vegetables, and these barriers should be affected or destroyed in order to increase carotenoids accessibility, during the digestion. Treatments, like thermal, affecting structural barriers have also influence on the food final structure and should thus be optimized in order to deliver products with an acceptable quality both from nutritional and structural point of view [16]. Another example, focused on the release of protein, lipid and vitamin E from almond seeds during digestion, revealed that bioaccessibility is improved by increased residence time in the gut and is regulated by almond cell walls. Almond cell walls, when physically intact, play primary roles in influencing nutrient bioaccessibility, acting as a physical barrier, encapsulating nutrients and hindering the rate and extent of nutrient release [47].

Knowledge on food microstructure is very important not only to design and develop new products, establishing its stability along storage, but also to ensure the bioavailability of different compounds.

4.2. Impact of microstructure on volatile flavor release

Flavor is considered one of the most important food quality attributes determining consumer's acceptability. Food microstructure can play an important role in flavor retention and/ or flavor release during food processing and consumption [48], once (micro)structure complexity is related with the interactions between food macromolecules, which in turn are responsible for the different affinity of the volatile compounds to food matrix, by modifying the nature and the number of binding sites that are available to the aroma compound [49]. Due to foods complexity, it is often difficult to have conclusive results on the effect of texture and microstructure on the flavor behavior. However, literature reported that this parameter has generally a higher effect on the food flavor release before consumption than during ingesting [48]. Literature reports some examples about the effect of food inhomogeneity on flavor behavior (retention and release), e.g. hard candy manufacturing, or the entrapment of the volatile compounds in the lipid and hydrophilic phase during baking crackers [48].

4.3. Texture perception and structure/ microstructure

Understanding the relationship between food texture perception and food structure/ microstructure is of increasing importance for companies wishing to produce texturally attractive food products [50].

Food texture is a cognitive property assigned to foods on the basis of how senses interact with the food by vision, touch and oral processing [7]. This concept involves all the rheological and structural (geometric and surface) attributes of the food products [51]. Sensations perceived in-mouth by consumers immediately evidence that they are detecting many complex structural changes taking place throughout chewing and swallowing [15]. Therefore, food texture is perceived during the conversion of food structure into a bolus through a complex series of oral manipulations including ingestion, processing and swallowing [52, 53]. During oral processing structure is broken down with forces applied by teeth and/ or tongue, and it is the food structure

the parameter that determines mechanical properties, and mechanical properties determine processes needed before swallowing [52, 53].

The complexity of texture, as a multi-parameter property, makes difficult the knowledge about structure relationships. However, strong evidences exist for some associations including between texture and hardness, moisture release and crumbliness [7]. On one hand, food structure is also considered complex and consequently difficult to understand, with respect to structural elements producing specific textural properties. But, on the other hand, it is possible to relate specific structural elements to textural properties [7].

As an example, a research study demonstrated that under oral processing starch-containing foods will undergo enzymatic breakdown, reducing its hardness [54]. Also, transitions associated to the temperature, like for example melting, can modify food structure during mastication, which alter its perceived texture throughout the oral process [7].

There is also the potential of using induced droplet aggregation to control the properties of model food dispersions containing mixtures of protein coated and starch granules. The understanding of the influence of calcium content on the microstructure and physicochemical properties of these systems demonstrated the relevance of microstructure on the design of reduced fat foods and provided an indebt knowledge of the influence of the structural organization of fat droplets and starch granules in complex mixed dispersions on their physicochemical properties. This may be relevant for the rational development of reduced fat products with improved physicochemical properties and sensorial attributes [55].

5. Using microscopy techniques for quantifying microstructure

Microscope techniques are an essential methodology to evaluated food microstructure since the human visual system is not well prepared to make objective and quantitative determinations of the image features seen under the lens of a microscope.

In literature it is possible to find a great number of different techniques that can be used in order to simplify food microstructure analysis.

Microscopy (optical or light, electron and atomic microscopy) and other imaging techniques generate data in the form of an image. They are an extension of the visual examination of foods that has been practiced by consumers and food processors. Microscopy techniques vary in method of image production, resolution and type of signal detected, and give a particular type of structural information that is unique to the technique used [56]. Using different techniques together allows, in some cases, obtaining complementary results.

Light microscope has been shown to be an ubiquitous and versatile tool in food science. The use of this technique to evaluate food microstructures allows getting a qualitative description of the samples structures. On the other hand, the electron microscope includes an assortment of different techniques and is usually considered crucial for the biology and physics fields. The use of this microscopy to promote the understanding of foods and the support in the development of new food products has been increasing upwards. Electron microscope is a considerably improved resolution technique when compared, for instance, with light microscope. Image formation is similar in both techniques, but the illumination source is electron focused with magnetic lenses rather than photons focused with glass lenses [56]. Despite being considered as mostly destructive methodologies, required for samples preparation, light and electron microscopies are the most used to analyze food microstructures. These techniques are user friendly and less expensive when compare with other methods like magnetic resonance imaging (MRI) and atomic force microscopy, that are limited to specific applications [57]. Actually, a great number of other diverse techniques are being developed in order to better visualize, quantify and understand food microstructure [26, 57]. These innovative techniques allow to obtain non-destructive and a non-invasive 3D imaging at resolutions higher than 1 μm and also a quantitative characterization. X-ray micro-computed tomography is an example of an innovative radiographic technique applied to food microstructure analysis.

6. Conclusions

The growing concern of society with respect to public health is the huge challenge for today's food industries. The well-established link of eating patterns with some chronic diseases,

such as obesity or diabetes, focused the companies for increasingly producing products by reducing high levels of fat, sugar and salt. This often comes at a consequence for taste and texture, and since the consumer will not accept any compromise on quality, also here the industry urgently needs to redesign many existing food microstructures. All these requirements have led to a renewed interest in relations between food structures and consumer relevant quality features.

Following this information, the knowledge of the relationship between structure/ microstructure and function and how structure can be manipulated in order to achieve proper functionality is of utmost importance. Structure/ microstructure of a food product may be responsible for changing the bioavailability of relevant food components, flavor release, or texture perception in-mouth. Also, structure/ microstructure is an important characteristic to determine a food matrix stability, since can influence the molecular dynamics of the systems, namely the water molecular dynamics, and be responsible for the occurrence or not of degradative reactions by the promotion of the contact between the different components. Despite of NMR technique is not based on image tools, it can be considered as a powerful methodology to evaluate the matrix structure/ microstructure. However, nowadays a great number of non-invasive image techniques are being developed in order to better observe and quantify the microstructure of a food product.

Further work on food microstructure/ functionality is a crucial and necessary approach to fully attain the manufacture of high-quality, healthy and tasty foods.

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