Moving from molecules, to structure, to texture perception

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Abstract

The two main food biopolymers, proteins and polysaccharides, are used in many foods to form structures that are the basis of product identity and quality. One of the main factors determining food quality is texture. A simplistic view of food is that molecules are assembled into structures, and the breakdown of structures during oral processing determines texture. There have been great advancements in understanding mechanisms for how proteins and polysaccharides assemble into structures, and factors determining the mechanical properties of the structures. In contrast, the understanding of how breakdown of structures during oral processing produces specific textural properties is still incomplete. Structuring processes based on protein-polysaccharide mixtures and emulsion gels are evaluated regarding structure formation and texture perception. It is shown that these systems can be designed to modulate specific textural properties. Finally, future trends in this area are discussed.

Key Words: protein, polysaccharide, emulsion, texture, structure, oral processing

1. Introduction

1.1 Food structure and texture in relation to food security

The world population is predicted to reach 9.7 billion people in 2050, with Africa and Asia representing 80% of the total (United Nations, 2015). In a 2010 report on the challenge of feeding 9 billion people, it was stated that “A threefold challenge now faces the world: Match the rapidly changing demand for food from a larger and more affluent population to its supply; do so in ways that are environmentally and socially sustainable; and ensure that the world’s poorest people are no longer hungry (Godfray et al., 2010).” Furthermore, “This challenge requires changes in the way food is produced, stored, processed, distributed, and accessed that are as radical as those that occurred during the 18th- and 19th-century Industrial and Agricultural Revolutions and the 20th-century Green Revolution (Godfray et al., 2010).” Therefore, a scientist starting a career today should consider their activities over the next 33 years (now until 2050) as part of a new revolution in food and agriculture that must meet the needs of an expanding population with greater land and environmental constraints. Rather than viewing agriculture, food production, and dietary needs as separate areas, the focus should be on a *food supply continuum* that maintains a healthy environment (agriculture) and world population. Understanding how food molecules assemble into structures, and how those structures contribute to food quality and health, will allow for optimization of the *food supply continuum* to individual, regional, and world needs.

Producing desirable food structures and textures can be approached from a food material science perspective where food is considered a material that is designed to provide specific properties. From that perspective, the goal is to assemble molecules into structures that are desirable to eat because they contribute positively to pleasure, nutrition, and health. The structures are assembled either by nature (e.g., fruits, vegetables, and fresh meats), or are manufactured (e.g., bread, cheese, and processed meats). There are currently many gaps in understanding how food structure contributes to hedonic and health goals. This knowledge is essential in addressing the predicted needs in agriculture and food, such as moving to a more plant-based diet. Therefore, one goal could be the ability to make a specific food type with a range of agriculture materials

(animal or plant) that could be optimized based on sustainable agriculture practices. This requires 1) defining the essential properties of the desired food structure and 2) understanding how that structure can be produced using a variety of molecules. In other words, how to move from molecules, to structure, to texture perception. The general approach of generating food structures with specific textural properties is outlined in Figure 1. It should be acknowledged that for centuries humans have been able to manufacture foods with desirable properties based on an incomplete or no knowledge regarding the chemical and physical mechanism involved. Craftsmanship is based on a keen eye and memory to produce a desired product. It also has a high potential for failure and waste. So a fundamental understanding of all processes is needed for efficient and effective food production.

1.2. A brief history of texture research

In the introduction to Scott Blair's classical monograph titled "Foodstuffs their Plasticity, Fluidity and Consistency," he made several key points that can be reevaluated at this junction 64 years post the original publication (Scott Blair, 1953). First was that investigating the rheological properties of foods was almost entirely empirical and that models showing springs and dash-pots had no direct links with molecules. He concluded, "we know practically nothing about what is really happening at the molecular level." A second element concerned the interest in using rheological properties to explain sensory perception of texture, which he termed "masticability." Two key points were made on this subject. The first being that known changes in sensory properties, such as toughness of apple skins, were (based on observation) not correlated with tensile strength or ductility of skins. This was offered as an indication that what is intuitive from a classical mechanical property testing framework may not hold true with textural assessment by humans. It was further mentioned that food quality is ultimately determined by subjective assessment of humans, and a chapter in the book addressed this area in what was termed "psycho-rheology of foodstuffs." Harper (1953) described psycho-rheology as assessing the human reactions to rheological behavior of materials. The psycho-rheology chapter covered early works on human perception of liquidity, semi-liquidity, solidity, roughness, and smoothness to name a few properties that can be described based on physical methods and sensory analysis. The investigations discussed were often on non-food materials;

however, the goal was the same - understanding how humans perceive mechanical/physical properties. The importance of multi-modal sensory input into texture perception was illustrated by mentioning that when experts judged the softness of wool that sight alone was more accurate than touch alone (Harper, 1953).

Scott Blair's monograph established the two extremes in length scales that would be the focus of texture research for subsequent years. The nanometer level where molecules alone (protein and polysaccharide), or in colloidal structures (e.g., emulsions and foams), can be described in great detail and characterized by physical and chemical methods. This "bottoms up" approach fits the dictum of Sir W. Thomason Kelvin in that "when you can measure what you are speaking about and express it in numbers you know something about it..." The other extreme of length scales is human perception of texture. In one of series of seminal papers from researchers at General Foods, Szczesniak (1963) proposed a classification system for textural characteristics. She used the term "structural elements" to describe structures observed at the molecular (nano-scale), microscopic (meso-scale), and visual (macro-scale) levels. This definition of food structure fits well with the current soft matter physics approach (see section 2.2) of describing food structure based on various length scales and consisting of measurable elements. Texture was proposed to be the combination of three "primary characteristics" – mechanical, geometrical, and other. *Mechanical* are evaluated by how a food responded to stress, *geometrical* are the macroscopic food structural elements assessed visually and possibly by oral sensations, and *others* includes those that are not attributed to mechanical or geometrical characteristics alone. The perception of moisture and fat content are given as examples of "other" properties.

As the world population is predicted to grow with Africa and Asia representing 80% of the total, another concern is how well texture terms translate globally across languages. In evaluating the equivalent of 54 English terms for specific sensory textural properties in 22 other languages, Drake (1989) found that the words fell into groups of viscous, plastic, elastic, compressible, cohesive, and adhesive. This suggests that a reductive view - focused on terms in different languages related to key physical properties - may allow for cross-language comparisons.

Mechanical properties (characteristics) of foods are commonly evaluated by instruments or by humans. When measuring mechanical properties of a food using a machine, there is variability in sensitivity and specificity among machines, but the general approach is to use one machine to assess a specific mechanical property (e.g., fracture stress). Texture testing by humans is different in several ways; one being that it generally requires assessment by a number of individuals to get a representative value. Therefore, unlike using one analytical instrument to measure a property, a group of human assessors will naturally vary in their ability and sensitivity to measure a texture property. The variability among groups can be exploited to determine e.g. the impact of gender, age, health status, or cultural context on texture perception, an aspect that cannot be studied using machines.

While much progress has been made in some areas since Scott Blair's observations in 1953, many of the same challenges face us today. The science of understanding how to assemble molecules into structures, and what determines the mechanical and other properties of structures, has advanced greatly in the intervening years. For example, see reviews by Dickinson (2006), Ikeda & Zhong (2012), van der Linden (2008), Douaire & Norton (2013), Mezzenga, Schurtenberger, Burbidge, & Michel (2005), van der Sman (2012), and Ubbink, Burbidge, and Mezzenga (2008), to name a few. In contrast, we still lack a comprehensive understanding of how those structures are perceived when eaten. The recent emergence of temporal assessment of physiological processes associated with eating (eating rate, muscle activity, jaw movement, and tongue movement), and sensory signals (e.g., **time intensity** and temporal dominance of **sensations**), is providing new insight into the dynamic nature of texture perception (Pineau et al., 2009; Çakır, et al. 2012b).

1.3 Moving from static to dynamic food texture models

Typical texture terms used in evaluating soft solid foods are hardness (firmness), friability (fracturability), cohesiveness, adhesiveness, moisture/fluid release, and residual. Instructions on how they are evaluated vary among investigations, but in general, hardness (effort for the first bite) and friability (number of particles) are “first bite” terms while cohesiveness and adhesiveness are evaluated after a number of chews. Moisture/fluid release can be evaluated at

first bite and/or after a number of chews. While a descriptive sensory analysis method may provide one number for each attribute, it is understood to be based on an evaluation after a given number of chews.

The awareness of the dynamic nature of texture has brought a new focus on the role of oral processing in texture perception. Oral processing consists of manipulation and transporting of food in preparation for swallowing (Chen, 2009). It involves of two aspects – physiological processes and transformation in food structure. The physiological processes include muscle activity for jaw opening and closing; direction, rate, and extent of tongue and jaw movement; and signals from mechanoreceptors from initial contact of food with the oral surfaces to swallowing. Food structure transformations vary widely among foods; with examples of particle generation in cookies and fluid release in cooked meats. Connections among elements of oral processing, transformations in food structure, and dynamic sensory perception are being investigated (Çakır et al., 2012b; Foster et al, 2011).

The dynamic nature of texture perception was captured by Hutchings & Lillford (1988) in their model showing oral processing as changes in food structure and degree of lubrication (e.g., bolus preparation) over time. While only relative changes in “degree of structure” were shown, an oyster required no structural change while tough dry meat required a high degree of change. Food texture is continuously assessed during oral processing. The temporal perception of food structure breakdown and associated texture terms was addressed by Lenfant, Loret, Pineau, Hartmann & Martin (2009) using the sensory method of *temporal dominance of sensations*. Six types of wheat flakes were evaluated and all ended with stickiness as the final, dominant sensation. Differences among wheat flakes were shown by the temporal patter of preceding textures being: hardness, grittiness (n=1); hardness, crackly, crisp (n=3); and crisp, brittle, light (n=2). This implies that three questions need to be answered to explain the cause for textural differences among samples. First, were there differences in food structure that could be seen prior to oral processing? In other words, were there quantifiable differences in initial structure that would imply variances in the breakdown pattern? Second, what were the differences in structural transformation during oral processing? Then finally, how were distinctive textural elements assessed during oral processing? Forming different structures using proteins,

polysaccharides and lipids, and associated oral processing and sensory perception (Figure 1), are addressed in the following sections.

2. Food structures

2.1 States of food matter

The four states of matter are gas, liquid, solid, and plasma. The majority of our foods fall into the categories of liquids and solids, with gases being primarily a processing factor (smoke) or a component of food (aroma, carbonation, air). Rheological properties can be used to provide a comprehensive characterization and differentiation among fluids and solids. In moving from mechanical characterization to texture perception, additional factors need to be considered. Subclassifications of semisolid, soft solid and hard solids provide more differentiation in regards to texture (Sherman, 1969; van Vliet, van Aken, de Jongh, & Hamer, 2009). Soft and hard solids require chewing and form particles during oral processing. While the distinction between soft and hard solids may be somewhat arbitrary, a good rule of thumb is that hard solids emit sound when fractured (van Vliet et al., 2009). Cheese and meat are examples of soft solids, while crackers, dry toast, carrots, and celery are hard solids. Beverages are the most common liquid food. Semisolids are materials between a liquid and a soft solid. Pudding, some yogurt and sauces, and ice cream are semisolids. A textural distinction between liquids and semisolids is that the latter usually requires some oral manipulation to transform the food into a bolus for swallowing, while liquids are mainly transported for short time periods when consumed.

2.2. Structure forming mechanisms

The ultimate goal in food structure formation is to create a structure with specific properties that relate to stability (shelf life), enjoyment, and delivery of compounds that benefit human health. For example, Norton, Moore & Fryer (2007) explain how food structuring and breakdown can be used in addressing obesity. Areas of science that historically have been used to provide theoretical and practical frameworks for designing food structures are colloids (Dickinson, 2006; Norton and Norton, 2010) and polymers (Slade & Levine, 1991); however, more recently the

overall concepts of soft matter physics have been used (Mezzenga et al., 2005; Ubbink et al., 2008; van der Sman, 2012). As noted by de Kruif (2012), soft matter includes liquids, colloids, polymers, foams, emulsions, gels, and granular material. This wide range of materials covers most if not all manufactured foods. van der Sman (2012) indicated a feature of soft matter is mesoscopic structures on the length scale of micrometers; a size that is important because it is a level that humans can sense. An expanded length scale and associated features is presented by Ubbink et al. (2008). One overall concept is that the mesoscopic structures can be “course-grained” and represented by properties, such as size, flexibility, and amphiphility (van der Sman, 2012). This is very important to concepts of food security because it implies that the meso-structural elements associated with desired properties can be created by various molecules. For example, if tofu texture is determined by a specific gel structure of soy proteins, then forming that gel structure with pea, wheat, or any other protein should produce the same texture (ingredient flexibility while maintaining texture perception). What is inferred in this hypothesis regarding structure and sensory texture is that the concept of “structure” is not a static image of molecular composition and spatial orientations, but a dynamic process of structural changes during oral processing (Hutchings & Lillford, 1988).

2.2.1. Gels. Proteins and polysaccharides are the two main biopolymers used in food to form structures, especially those with high water content. Gels are commonly used as model systems in investigating texture of soft solids because they can be formed with proteins, polysaccharides or mixtures thereof, and represent structure forming mechanisms for food products, such as cheese, yogurt, processed meats, and cooked eggs (Gwartney, Larick, & Foegeding, 2004; Vilgis, 2015).

In gels composed of a protein or polysaccharide network surrounded by water, the biopolymer network generally represents < 10% of the total mass. The simplest gel is one where individual polymers, or mixture of compatible polymers, are used to form the gel matrix. More complex mesostructures can be formed when using a mixture of proteins and polysaccharides under conditions that cause associative or segregative phase separation (see section 3). Likewise, complexity can be increased by adding a dispersed phase of liquid (oil or a separated second

aqueous phase), solid (fat, cocoa particles, crystals and others), or gas. These are often called *filled gels* and are similar to several food products (see section 4).

2.2.2. Jammed systems. Another mechanism for forming soft and hard solids is jamming. As defined by Jaeger (2015), “Jamming occurs when geometric constraints among neighboring particles prevent mobility and the applied stress is too small to cause yielding. Conversely, unjamming and the associated loss of aggregate rigidity occur when the free volume experienced by particles becomes large enough to allow for mobility or when a sufficiently large shear stress is applied to force the system to dilate and yield.” Cheddar cheese is a soft solid that has gel and jammed structural elements. In manufacturing, a gel is formed initially followed by a series of processing steps that cut the gel into macroscopic particles, remove fluids, and re-forms particles into a cohesive mass. With aging, the connectivity of the protein network decreases. This is seen with young cheese being springy with a low degree of breakdown after a few chews; however, after aging (9 months), the same cheese has a lower level of springiness and increased degree of breakdown (Rogers et al., 2009). This implies moving from a gel to a combination gel/jammed structure.

3. Structuring based on protein-polysaccharide mixtures

3.1. Phase separation vs. gelation as driving forces behind structure formation

The understanding of the mechanisms underlying the texture development by phase separation between (bio)polymers dates back to the sixties of the last century. As many reviews have been published on the thermodynamic principles underlying the interactions between biopolymers (Norton & Frith, 2001; Schmitt, Sanchez, Desobry-Banon, & Hardy, 1998; Schmitt & Turgeon, 2011; Tolstoguzov, 2003; Turgeon, Beaulieu, Schmitt, & Sanchez, 2003), only the basics are summarized in this paper. Cahn & Hilliard (1958) described the thermal fluctuations in composition or concentration as the driving force behind the formation of inhomogeneous structures. These inhomogeneities exist in the polymer composition (segregative phase separation; right hand side of Figure 2) or total polymer concentration (aggregative phase separation; left hand side of Figure 2). Typically, this instability and phase separation is

accompanied by a turbid appearance. The initial step of this process is driven by thermodynamic forces, e.g. the diffusion constants and incompatibility. The further coarsening of the structure is dominated by the hydrodynamics, interfacial tension, viscosity ratio between the phases and the overall viscosity (Siggia, 1979). The final microstructure of the mixture is the result of the interplay between phase separation and gelation of at least one of the two phases (Binder & Stauffer, 1974). In Figure 2 one biopolymer is given in blue, whereas the second or the polymer depleted aqueous phase is given in white. Upon increasing phase separation, the domains of the first biopolymer become more concentrated (darker blue) and more defined (more round structures). Upon gelation the microstructure is fixed and cannot coarsen further. When shear is applied during phase separation, elongated structures are formed in contrast to round. In general, slow gelation allows for a further coarsening of the microstructure.

As explained before, gelation of at least one of the two phases arrests the microstructure. This allows for developing gelled model foods to study texture, structure, and sensory relationships. When proteins and polysaccharides form the two biopolymer phases, gelation is generally obtained by the gelation of the protein phase. Over the years, different methods have been applied to induce protein gelation, including acidification, heating or cooling, salt addition, enzymatic cross linking, and high pressure treatment. These methods have been used for different proteins as well as for mixtures of these proteins with different polysaccharides.

3.2. Classification of the microstructure of different model protein-polysaccharide systems

Whey proteins have been and still are the workhorse of many research groups. Phase separated whey protein/polysaccharide gels were obtained by acidification (de Jong & van de Velde, 2007; Wassén et al. 2013) or by heating of whey protein/carrageenan mixtures (Çakır, Khan, & Foegeding, 2012a; Çakır, et al., 2012b). The microstructures of phase separated mixed protein/polysaccharide gels can be classified according to their morphology (Çakır et al. 2012a,b; van den Berg, van Vliet, van der Linden, van Boekel, & van de Velde, F. 2007; 2008a; van den Berg, Rosenberg, van Boekel, Rosenberg, & van de Velde, 2009). Figure 3 gives a schematic overview of the microstructure of homogeneous (H), protein continuous (PC), bicontinuous (BC), serum continuous (SC), coarse stranded (CS) and particulated (P) gels. The length scale of the

bicontinuous microstructure ranged from below 10 μm for acidified whey protein/gellan gum gels (de Jong & van de Velde, 2007) to above 50 μm for heat-set whey protein/carrageenan gels (Çakir & Foegeding, 2011). The serum continuous phase separated systems were formed at high concentrations of polysaccharide and comprised protein spheres embedded in the serum or polysaccharide phase. In the case of acidified whey protein mixtures with non-gelling locust bean gum or gellan gum below the gelation concentration, the samples were liquid-like and the size of the protein beads could be adjusted by cross-linking and pH adjustment (de Jong & van de Velde, 2007; Chojnicka-Paszun, Doussinault, & de Jongh, 2014). In the heat-set whey protein carrageenan gels, the high concentration of carrageenan turned the serum continuous gels into self-supporting gels, which were originally classified as carrageenan continuous (Çakir & Foegeding, 2011; Çakir et al. 2012b).

The research on mixed protein/polysaccharide gels is not limited to whey protein gels. Other milk proteins, such as caseinate, and animal proteins, such as gelatin, are used to prepare gels with different microstructures. The addition of guar gum to acidified sodium caseinate gels shifted the microstructure from homogenous for pure sodium caseinate towards protein continuous, bicontinuous and serum continuous microstructures. The type of microstructure obtained is strongly affected by the protein/guar gum ratio (Hidalgo et al., 2015). Very finely structured coarse stranded gels were obtained by rennet induced gelation of micellar casein in the presence of carrageenan (Wang et al., 2014). Different carrageenans (kappa-, iota- and lambda-carrageenan) showed different binding modes with the casein micelles and, thereby, showed a different impact on the gelation and microstructure. In contrast to globular proteins, such as whey proteins that form a gel upon heating, gelatin is thermoreversible and gels upon cooling of a molten gelatin solution. When mixed with various polysaccharides, phase separated microstructures can be obtained (Norton & Frith, 2001). As discussed before, the obtained microstructure is the balance between phase separation and gelation. For gelatin, the gelation rate is determined by the quenching temperature, i.e. the temperature below the gelation temperature of gelatin. Not only the quenching temperature, but also the molecular weight of the polysaccharide (dextran) affected the final microstructure (de Hoog & Tromp, 2003). Protein continuous, bicontinuous and serum continuous gelatin/dextran gels with a wide range of length scales were obtained by varying molecular weight of the dextran.

With the growing demand for food proteins, the development of plant proteins as a sustainable source of proteins has progressed (van de Velde, 2016). This development is not limited to single plant protein gels, but includes also mixed plant protein-polysaccharide gels. These plant protein-polysaccharide gels can also be classified according to the classes defined for the whey protein-polysaccharide gels (Figure 3). Homogeneous, protein continuous and bicontinuous microstructures were formed upon acidification of mixtures of soy protein isolate and gellan gum or xanthan gum (Chang Lib, Wanga, Bib, & Adhikari, 2014). Analogues to the acidified whey protein gels, the shift from homogeneous to protein continuous and bicontinuous microstructures was the result of increasing polysaccharide concentration. Heat-set soy protein / kappa-carrageenan mixed gels formed homogeneous, protein continuous and particulated microstructures depending on the soy protein to kappa-carrageenan ratio and the pH of the mixture (Cavallieri, Garcez, Takeuchi, & da Cunha, 2010). Heat-set soy protein gels were prepared with different galactomannans, i.e. guar gum, tara gum and locust bean gum (Monteiro, Rebelo, da Cruz e Silva, & Lopes-da-Silva, 2013). The microstructure changed from protein continuous for pure soy protein to protein continuous at different length scales for guar gum and tara gum to bicontinuous for locust bean gum. The gelation of plant proteins is not limited to heat- and acid-induced gelation, but also enzymatic cross linking by transglutaminase (TGase) has been described. Increasing amounts of sugar beet pectin shifts the microstructure of TGase cross linked soy protein gels from homogeneous to protein continuous and bicontinuous (Hou et al., 2015). Phase separation and microstructure formation is not limited to protein/polysaccharide mixtures, but also occurs in protein/protein mixtures. Bicontinuous heat-set whey protein gels can be obtained by the addition of gelatin (Ersch et al., 2016).

As described above, the classification of microstructures of phase separated gels is not limited to microstructures obtained by whey proteins. Common microstructures are generally observed for a wide range of gels based on animal and plant proteins gelled with a wide variety of methods. Pure protein gels are generally homogeneous or particulated. Upon addition of polysaccharide, the types of microstructures separate into two groups: coarse stranded on the one hand and protein continuous, bicontinuous and serum continuous on the other hand (Figure 4). Coarseness or characteristic length scale of the coarse stranded gels generally increases with increasing

polysaccharide concentration. Independent of the type of protein, the gels that form protein continuous, bicontinuous and serum continuous microstructures all follow this trend with increasing polysaccharide concentration. To be more specific, the trend is associated with increasing polysaccharide to protein ratio. It is generally observed that higher concentrations of polysaccharides are required when higher concentrations of protein are used to prepare the phase separated gels. On top of this major effect of concentration, the charge density, degree of substitution and molecular weight of the polysaccharides determine the final microstructure. Or phrased in a different way, these molecular properties determine the concentration window for each of the different microstructures.

The above described understanding of microstructure formation in mixed protein polysaccharide gels is explanative, i.e. once the microstructure is observed, the mechanism behind can be described. Prediction of the microstructure of mixed gels based on molecular properties of the proteins and polysaccharides and the selected gelation mechanism is in its infancy. Simulations on the thiol-disulfide exchange in β -lactoglobulin aggregation are available (Zeiler and Bolhuis, 2012; 2015), but until now not extended to mixed protein-polysaccharide gels. Ingredient flexibility, such as making whey, pea or gelatin-based tofu is still a cyclic development, whereby explanation of observed microstructures can be used to design the next development step.

3.3. Texture perception of mixed protein-polysaccharide gels

Texture attributes describing the properties of mixed gels can be grouped based on the stage in the oral processing: first bite, chew down and residual after swallowing (oral coating) (Brandt, Skinner, & Coleman, 1963; Foegeding, Çakır, & Koç, 2010). Sensory attributes generally used to describe semi- and soft solids according to the stage in the oral processing were summarized by Stieger and van de Velde (2013). The start of the oral processing is the compression of the gel between tongue and palate or biting through the gel with the incisors until the product falls apart: first bite. The firmness of the product determines whether it is palated or chewed. Generally, sensory firmness correlates well with mechanical parameters, such as stress at fracture and possibly Young's modulus, and physiological parameters, such as jaw muscles activity and vertical jaw amplitude (Çakır et al., 2012b). The sensory firmness is not affected by the

microstructure of the mixed gels, but by the overall ingredient concentration. However, microstructure has a significant impact on the first bite attributes water release (separation) and crumbliness (van den Berg et al., 2007; 2008a). Gels showing a homogeneous (H) or protein continuous (PC) microstructure generally are perceived as crumbly, whereas bicontinuous (BC) microstructures are perceived as separating or watery. Energy storage in the gel matrix is a prerequisite for the fracture mechanisms underlying crumbliness. As serum release or moisture expulsion under deformation results in energy dissipation, these two attributes are each other's opposite. High chewing frequencies were observed for crumbly gels and low chewing frequencies for gels that show energy dissipation due to moisture or serum release (Çakır et al., 2012b).

The rhythmical chewing phase, aimed at reducing the particle size of the food, is generally the longest part of oral processing. Particles formed upon oral processing are glued together by saliva and moisture from the gel to form a cohesive bolus that is safe to swallow. Sensory attributes related to this period of oral processing are referred to as chew down attributes. Watery, separating and moisture release are texture attributes related to fluid release during oral processing and perceived during chew down and to a lower extend at first bite. As described above, microstructure is dominant for the fluid release, with high ratings for the associated attributes in gels with a bicontinuous (BC) or coarse stranded (CS) microstructure. The degree of serum release is affected by differences in gel microstructure, such as pore size and micro-phase separation (Urbonaite, de Jongh, van der Linden, & Pouvreau, 2014). Moisture release was not correlated with chewing duration, number of chews or muscle activity, but negatively correlated with chewing frequency (Çakır et al., 2012b). Associated attributes, such as moist and refreshing, are perceived during chew down, but are more related to type of gelling agent rather than to the microstructure (Devezeaux de Lavergne, Strijbosch, van den Broek, van de Velde, & Stieger, 2016b).

Particle size and size distribution play an important role in perception of chew down texture attributes, such as powdery, grainy, and spreadable. Whereas powdery and grainy are directly related to particle size, spreadable is more indirectly related. Longer time periods during the jaw closing phase were observed for gels breaking down into larger, non-uniform particles, while jaw

opening was faster for those gels (Çakir et al., 2012b). These large particles were generally observed for homogeneous (H), protein continuous (PC) and bicontinuous gels (BC), and were formed by a distinct fracture path through the material as observed by confocal microscopy (van den Berg et al., 2008b). The number of particles counted by image analysis in spat-out boli correlated well with the texture attribute spreadable (van den Berg et al., 2008a). A large number of particles related to a high score for spreadable and micro-cracks throughout the microstructure and multiple fracture points. High spreadability is generally observed for coarse stranded gels (CS). After swallowing, the oral coating is formed by residues adhering to the tongue, teeth and oral tissues. With respect to mixed protein-polysaccharide gels, particle size and size distribution of the particles left after swallowing are the parameters affecting sensory perception the most (Çakir et al., 2012c). However, fat and/or emulsion droplets play a more important role in the after swallowing perception of texture attributes, as discussed in the next section.

Summarizing, the protein type, i.e. molecular structure, pH and salt conditions of the solution in combination with the gelation mechanism determines if a homogeneous protein gel or a particulated gel is formed. The polysaccharide type, i.e. molecular structure, molecular weight and charge density, determine if a protein continuous or a coarse stranded phase separated gels is formed upon gelation of the mixed solution of protein and polysaccharide. Upon increasing concentration of polysaccharide, the particulated and coarse stranded gels coarsened, whereas the protein continuous gels turned into bicontinuous and serum continuous upon increasing polysaccharide concentration.

As can be seen from the citations listed above, whey protein based mixed gels systems are still the workhorse of the research relating microstructure to oral processing and sensory perception. Although, microstructure formation is studied for plant proteins, the sensory properties of mixed plant protein-polysaccharide gels have not been studied in great detail. This knowledge gap needs to be filled in order to make a step towards the global challenge of ingredient flexibility and the transition from animal to plant based protein-rich foods.

4. Structuring based on emulsion gels

4.1. Emulsion droplet matrix interactions

More complex meso-structures of gelled foods are obtained when a dispersed phase, such as a liquid oil (i.e. emulsion droplets) or solid fat are added to protein, polysaccharide or mixed protein-polysaccharide gels. These gels are referred to as *gelled emulsions* when the emulsion droplets, typically coated with proteins at the interface, aggregate upon gelation to a form space filling networks of emulsion droplets. Alternatively, they can form *emulsion-filled gels* when the emulsion droplets are embedded into a gelled matrix formed by proteins, polysaccharides or mixtures thereof. Gelled emulsions and emulsion-filled gels are also often referred to as *emulsion gels*. Emulsion gels are an important class of typically semisolid and soft solid model foods that are representative for a broad variety of products, such as cheeses, ice creams, spreads, and processed meat. The meso-structure and resulting physical-chemical properties of emulsion-filled gels can be controlled by the properties of the added emulsion droplets and its interactions with the matrix. Volume fraction, size, rheological properties, and surface charge of the added emulsions droplets have been demonstrated to control the properties of emulsion-filled gels (van der Poel, 1958; van Vliet, 1988; Chen & Dickinson, 1998; 1999). The interactions between the emulsion droplets and the gel matrix determine to a large extent the properties and structure of the emulsion-filled gel. To modify the binding properties of emulsion droplets to the gel matrix, electrostatic interactions between droplets and matrix can be controlled, usually by changing the surface charge of the emulsion droplets by using different types of emulsifiers, or altering the pH of the matrix. The interactions between emulsion droplets and gel matrix can be controlled by the surface properties of the emulsion droplets. Emulsion droplets that do not interact with the matrix are termed unbound or inactive fillers. Emulsion droplets that interact with the matrix are termed bound or active fillers. Depending on the ratio between the modulus of the filler and the modulus of the gel matrix, fillers (emulsion droplets) either increase or decrease the modulus of the emulsion filled gel. When the modulus of the filler is smaller than the modulus of the matrix, bound and unbound emulsion droplets decrease the gel modulus with increasing filler volume fraction. When the modulus of the filler is larger than the modulus of the matrix, bound emulsion droplets increase the gel modulus with increasing filler volume fraction. Emulsion droplet size affects the modulus of the filler and can consequently affect the modulus of emulsion filled gels (McClements, Monahan, & Kinsella, 1993; Kim, Gohtani, Matsuno, & Yamano, 1999; Kim,

Renkema, & Van Vliet, 2001; Rosa, van Vliet, & van de Velde, 2006; Sala, van Vliet, Cohen Stuart, van de Velde, & van Aken, 2009). Emulsion gels can be prepared by various gelation methods including heating, acidification, enzymatic crosslinking, and renneting (Dickinson, 2012).

4.2. Structure heterogeneity formation and mechanical properties of emulsion gels

The lipids embedded in emulsion gels contribute to many aspects determining the gel properties such as structure, mechanical properties, and sensory texture (Akhtar, Murray, & Dickinson, 2006; Chung, Degner, & McClements, 2013; McClements & Demetriades, 1998; Chen & Dickinson, 1998; Sala, van Aken, Stuart, & van de Velde, 2007a; Sala et al., 2009). In the past decades, our understanding of the impact of the properties of emulsion droplets on structure formation and mechanical properties of emulsion gels has advanced considerably. In addition to factors previously mentioned, mechanical properties of emulsion gels are dependent on the spatial distribution of the emulsion droplets in the gel matrix, and the inhomogeneity of the gel matrix (Oliver, Scholten, & van Aken, 2015a; Oliver, Berndsen, van Aken, & Scholten, 2015b; van Aken, Oliver, & Scholten, 2015; Oliver, Wieck, & Scholten, 2016). The distribution of emulsion droplets can be altered by aggregation or clustering of droplets by controlling the gelation kinetics of the matrix to tune the degree of aggregation (van Aken et al., 2015). A schematic overview of the microstructures of emulsion gels is provided in Figure 5.

The clustering of emulsion droplets can increase gel strength by a factor of 2x (Oliver et al, 2015b). By engineering droplet distribution, mechanical properties of emulsion gels can be controlled allowing to maintain desired mechanical properties while reducing the content of the dispersed oil phase. Inhomogeneity can be induced by local phase separation of proteins leading to protein dense and protein poor areas or voids within the gel matrix (Ako, Nicolai, Durand, & Brotons, 2009). The degree of inhomogeneity can be controlled by the properties of the protein matrix (type of protein, concentration, pH and their mutual interactions). When inhomogeneity is increased by changing protein content and their interactions (Oliver et al., 2015a), gel strength can increase by a factor of 10x (Oliver et al., 2016). Little is known about the effects of inhomogeneity on oral processing behavior and texture perception. In conclusion, these studies

demonstrate how the microstructure of emulsion gels can be engineered to obtain specific mechanical properties.

4.3. Texture perception of emulsion gels in relation to structure and mechanical properties

The impact of emulsion droplet properties and its interactions with the gel matrix on texture perception, especially fat related sensations, such as creaminess, are still not fully understood. Several studies linked emulsion gel structure and large deformation properties to texture perception (Gwartney et al., 2004; Kim, Gohtani, & Yamano, 1996; Sala, van de Velde, Cohen Stuart, & van Aken, 2007b; Sala, de Wijk, van de Velde, & van Aken, 2008). Creaminess perception has been related to the type of gel matrix used, volume fraction of oil, and the release of emulsion droplets from the surrounding gel matrix during breakdown as it would occur during mastication (Chojnicka, Sala, de Kruif, & van de Velde, 2009; Sala et al., 2007a). Increasing the volume fraction of the dispersed oil and reducing emulsion droplet size can enhance creaminess perception (Sala et al., 2007b). Release of emulsion droplets from the gel matrix during mechanical breakdown is larger when emulsion droplets are unbound to the matrix compared to when emulsion droplets are bound to the matrix (Chojnicka et al., 2009; Sala et al., 2007b). The increase in oil release from gels with unbound emulsion droplets leads to an increase of the oil fraction that is deposited on the tongue surface during mastication (Camacho, Liu, van der Linden, Stieger, & van der Velde, 2015). Fattiness perception of emulsion gels increased with increasing mastication time and decreased after expectoration of the boli (Camacho et al., 2015). More droplet coalescence was observed in emulsion-filled gels with unbound droplets than in emulsion-filled gels with bound droplets. The increase in coalescence related to a decrease in friction, which related to an enhancement of the perception of fat-related sensory attributes (Liu, Stieger, van der Linden, & van der Velde, 2015). Coalescence of fat droplets in emulsion-filled gels can further be increased by increasing the solid fat content of the emulsion droplets (Liu et al., 2015). Emulsion droplet-matrix interactions can influence the friction properties of broken down emulsion gels, which influence the texture perception of fat-related attributes (Chojnicka et al., 2009). Thus, the release of emulsion droplets from the gel matrix and the structure transformations occurring during oral processing (i.e. droplet coalescence) influence the

mechanical and tribological bolus properties and consequently the oil fraction deposited on the tongue, which determines dynamic texture perception.

Phase transitions of the gel matrix influence texture perception of emulsion-filled gels. Gelatin based emulsion-filled gels melt upon heating to temperatures below 35°C (depending on gelatin grade and concentration), so melting will start at temperatures below mouth temperature (Inoue, Sasai, Shiga, & Moritaka, 2009). The melting of the gelatin matrix enhances creaminess considerably (Sala et al. 2008; Devezeaux de Lavergne et al., 2015a). Liquefaction of starch based soft emulsion gel matrices occurs by hydrolysis of salivary amylase (Janssen et al., 2007), and this transition also influences texture perception. Melting mixed agar/gelatin emulsion-filled gels display higher friction forces than mixed carrageenan/locust bean gum and mixed high/low acyl gellan emulsion-filled gels, which correlated to an increased stickiness and creaminess perception (Devezeaux de Lavergne, Strijbosch, van den Broek, van de Velde, & Stieger, 2016b).

To gain a better understanding of texture perception of emulsion gels in relation to structure and mechanical properties, the structural transitions of emulsion gels during mastication and the oral processing behavior of emulsion gels have to be considered (Pasca et al, 2013; Koç et al., 2013; Devezeaux de Lavergne et al., 2015a; Devezeaux de Lavergne et al., 2015b; Devezeaux de Lavergne et al., 2016a; Devezeaux de Lavergne et al., 2016b). Texture perception of emulsion gels is a highly dynamic process and changes continuously during oral processing. Different sensory attributes are perceived during different stages of mastication, such as firm in the beginning, elastic and sticky in the middle, and grainy or creamy at the end of oral processing depending on the mechanical properties of the emulsion gels (Devezeaux de Lavergne et al., 2015b). Often texture attributes perceived at the beginning of mastication, such as hardness/firmness, are correlated with mechanical properties, such as fracture stress. These attributes are primarily related to large deformation properties of foods (rheology dominant). Texture attributes, such as chewiness, stickiness and creaminess, are often perceived in the masticatory phase. During this phase of oral processing, the bolus is subjected to shear, flow, and displacement, which are strongly influenced by both the rheological and tribological behavior of the bolus. In the last phase of oral processing, the bolus is swallowed and perception is

determined more by the tribological behavior of food residuals and their interactions with saliva (Prakash, Tan, & Chen, 2013). At the end of oral processing, texture attributes of emulsion gels were mainly correlated to bolus flowability, linked to lubrication properties of the boli and the number of gel fragments in the bolus (Devezeaux de Lavergne et al., 2015b). Thus, texture perception of emulsion gels is dynamic and entails a “transition from rheology-dominant processes to tribology dominant processes” (Stokes, Boehm, & Baier, 2013).

4.4. Digestion of emulsion gels in relation to structure and mechanical properties

The structure and mechanical properties of emulsion gels do not only influence texture perception, which contributes to food quality and pleasure of eating, but also influence digestion and lipolysis, which contribute to nutrition and health. As the digestion of emulsions and emulsion gels has been reviewed previously (Golding & Wooster, 2010; Golding et al., 2011; Lairon, 2009; McClements, Decker, Park, & Weiss, 2008; Singh, Ye, & Horne, 2009), this section provides a brief summary of a limited number of recent studies focusing on the digestion of model emulsion gels in relation to structure and mechanical properties. For gelatin-based emulsion-filled gels, the lipolysis kinetics are related to the gel microstructure (Sarkar et al. 2015). A relationship between speed of fat digestion and gel porosity was observed, suggesting that fat digestion is controlled by the ability of the emulsion gel’s microstructure to slow lipase diffusion to the interface of fat droplets (Sarkar et al. 2015). For agar-based emulsion-filled gels, it has been suggested that the gelation of emulsions may delay lipid hydrolysis (Wang, Neves, Kobayashi, Uemura, & Nakajima, 2013). Soft boli of emulsion gels disintegrated faster than hard boli of emulsion gels under *in vitro* simulated gastric digestion conditions (Guo, Ye, Lad, Dalgleish, & Singh, 2014a). Only soft emulsion gel boli displayed oil droplet coalescence. The differences in the breakdown of soft and hard emulsion gels under *in vitro* simulated gastric digestion conditions were attributed to the different structures of the emulsion gel (Guo et al., 2014a). During oral processing, the release of oil droplets from emulsion gels increased with increasing oil droplet size (Guo, Ye, Lad, Dalgleish, & Singh, 2014b). More oil coalescence was observed for emulsion gels with larger oil droplets. Under gastric digestion conditions, high degrees of coalescence and phase separation of oil droplets occurred in emulsion gels containing larger oil droplets leading to slow gastric emptying (Guo et al., 2014b). Insight into the

relationships between structure, mechanical and physical-chemical properties of emulsion gels and digestion helps to develop foods that deliver nutrients to specific parts of the gastrointestinal tract and control digestion.

5. Outlook and future trends

A long-standing goal in textural studies has been to instrumentally measure properties of a food *ex vivo* that can predict *in vivo* perception of texture (Scott Blair, 1953; Sherman, 1969; Szczesniak, Brandt & Friedman, 1963). In other words, knowledge of textural transformations during oral processing (Figure 1) would not be needed to determine the magnitude of a specific textural property. Generally speaking, first bite attributes are well described by mechanical properties and microstructure of model food gels. While properties such as hardness and juiciness may be predicted directly from *ex vivo* measurements, this could be a *fait accompli* reflecting how the human is instructed to assess the property rather than the normal sensory experience. For example, if hardness is an evaluation of the force required to complete one chew, are two foods with the same instrumentally measured hardness going to be perceived as similar in hardness by consumers? The change in hardness from first bite to swallow would be very different between brittle and deformable structures – even though they may have started at similar first bite hardness. Likewise, properties, such as cohesiveness and adhesiveness, that are evaluated after several chews and involve saliva and bolus formation, are not logically associated with just structural elements of the starting food material. Therefore, oral processing needs to be considered in overall perception of texture.

Structural transformations are generally assessed by so called “chew and spit” methods where samples are chewed for various amounts of time, removed from the mouth, and analyzed for various properties. These methods have historically been more a part of the dental literature where a persons ability to chew food was evaluated as the reduction in particle size of a test or model food after a number of chews or the number of chews required to achieve a give particle size reduction (Fontij-Tekamp et al. 2000; Helkimo, Carlsson, & Helkimo, 1978). More recently they have been used to help understand texture perception (Young, Cheong, Hedderley,

Morgenstern, & James, 2013; van den Berg et al., 2008a; Devezeaux de Lavergne et al., 2015a; Devezeaux de Lavergne et al., 2015b; Devezeaux de Lavergne et al., 2016b).

As implied previously, there is more control over assessing rheological and mechanical properties of foods that when using human subjects, because of natural variation in the latter. For example, the type of oral processing activities for semisolids may vary among individuals. In evaluating cake texture, about half the panelists bit into the sample while the other compressed it between the tongue and hard palate (Boyd and Sherman, 1975). Moreover, asking panelist to evaluate a texture property has been shown to cause adjustments in oral processing. Force-time curves for chewing a piece of cheese are longer and with different magnitude of key transitions when panelists are asked to evaluate hardness as compared to when just requested to chew the sample (Dan, Okamoto, Wada, Dan & Kohyama, 2007). In general, asking subjects to analytical assess the texture changes oral behavior. For example, the total mastication time increases considerably when panelists use temporal dominance of **sensations** (TDS) to assess texture, as compared to free mastication behavior (Devezeaux de Lavergne et al., 2015a).

Most studies so far used TDS to link dynamic texture perception to mechanical properties and structure of (model) foods. An alternative sensory method, Temporal-Check-All-That-Apply (TCATA), allows identifying several sensory attributes that are concurrently perceived. It was suggested that TCATA may provide a more detailed description of the dynamics of the sensory characteristics of foods (Ares et al., 2015). It was also suggested that the TDS concept of dominance might decrease detailed description and discrimination of attributes that are simultaneously perceived, particularly when dealing with multiple sensory modalities. Similar to TDS, TCATA allows obtaining sensory trajectories to described perception over time for foods (Castura, Antúnez, Giménez, & Ares, 2016). The potential of TCATA in comparison to TDS to link dynamic texture perception to the mechanical properties and structure of foods has not been explored yet.

6. Conclusions

A fundamental knowledge of how molecules form structures, and how structures are transformed into texture perception and delivery of nutrients is essential to designing an optimized the food supply continuum. Much progress has been made on understanding how proteins and polysaccharides form structures with specific spatial and mechanical properties. However, if there is going to be a shift to more reliance on vegetable than animal proteins, then there is a gap because the majority of research has been done on dairy proteins. There is limited knowledge available on their behavior and sensory perception of plant based structures. There is also a need to move away from primarily focusing on structural links to “first bite” sensory properties and evaluate the entire process from first bite to bolus. Understanding how the dynamics of structural transformation during oral processing provides the signals for sensory perception of texture is key to developing a rational approach to food structure design.

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Figure 2. Schematic representation of the behavior of mixed biopolymer mixtures.

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Figure 4. Transitions in microstructures of mixed protein-polysaccharide gels with changes in composition. Abbreviations for structure types are the same as in Figure 3. Protein rich domains are given in blue.

Figure 5. Microstructures formed in emulsion gels (adopted from Olivier et al., 2015a). Protein rich domains are indicated in blue, and fat droplets are red. A. homogeneous gel without emulsion droplets; B. emulsion gel with homogeneous droplet distribution in homogeneous gel matrix; C. emulsion gel with inhomogeneous droplet aggregates in homogeneous gel matrix; D. heterogeneous, particulated gel without emulsion droplets; E. emulsion gel with heterogeneous droplet distribution in heterogeneous particulated gel matrix.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

A

B

C

D

E