Texture: The Physics of Mouthfeel – Spreadable Food and Inulin Particle Gels

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Introduction
Most food can be classified as “soft matter”, which has to be processed in the mouth by chewing, mastication and bolus formation. Even “hard matter”, such as crystalline chocolates or glasses, and amorphous matter, like hard caramels or potato chips, show “soft properties”; either they melt at slightly lower temperatures present in the mouth, they dissolve relatively quickly in the saliva, or their ultimate fracture properties are weak enough for them to be broken between tongue and palate. Consequently, pure physical processes induce a dominant part of the consistency and “mouthfeeling”, particularly mechanical processes; how do solids break?; how do the particles, through mastication, become smaller and smaller?; how are they wetted by the saliva?; how do the plastic deformation properties of the forming bolus change with time?; and what about the friction in the mouth? Apart from the composition of the foods, these mechanical properties also determine the release of water-soluble taste and volatile odorant components during oral processing, and finally lead to the desired sensations associated with indulgence (Chen and Engelen, 2012).

However, to understand these processes better, it must be noted that most of the sensory aspects have to be correlated with the hierarchical and multiscale (molecular) structure of the foods. When, for example, food materials break, the crack propagation depends on a number of molecular properties, the water activity, the resulting surface structure, and the molecular elasticity of proteins and carbohydrates, to name only a few. Even small differences in the physical properties yield distinguishable reactions. The most well-known examples are fresh and slightly stale potato crisps, where only a minimal uptake of humidity completely changes the fracture behaviour as well as taste and odour release. In addition, most processed foods are emulsions, which are sufficiently stable on long time scales. Well-known examples are liquid (e.g., mayonnaise) or solid (e.g., sausages, purees, and semi-solid foods) emulsions. In such emulsions, taste and odorant compounds can be exactly partitioned in their appropriate solvents to provide their controlled release during oral processing (Vilgis, 2014; Vilgis, 2015).

In this chapter, we concentrate mainly on the textural properties, which have a physical origin. One interesting and typical example is liver sausage, which is based on a mixture of pork fat, meat and liver, which forms a semi-solid emulsion. Its mouthfeel is dominated by a combination of plastic deformation and, when its temperature rises in the mouth, melting of the partially crystalline pork fat. Since such sensations are very much liked, it is also tempting to create fat-free, vegetarian or vegan alternatives, which might provide a similar sensation in the mouth. This is, however, a non-trivial challenge for soft matter physics.

Liver Sausages: The Role of Proteins and Fats
The mechanical deformation of liver sausage is dominated by its plasticity. Figure 87.1 shows a typical force–deformation curve of liver sausages when measured using a texture profile analyser. The increase at small deformation corresponds to elastic responses, mainly due to the protein matrix, whereas the constant force on increasing deformation corresponds to the plastic behaviour, when fat particles in the emulsion are pushed away from each other without any force required, which is easy to understand.

At room temperature, most of the fat is crystalline and becomes emulsified during processing inside a meat and liver matrix. In traditional liver sausages, the meat and fat are heated and cooled down, whereas the liver is not heated when processed in the cutter. In this case, the (native) liver proteins act as emulsifiers and enclose the still molten fat, which crystallises in the droplets during cooling. The water- (and salt-)soluble proteins of the liver (as well as mono- and diglycerides, free fatty acids and phospholipids present in the organ) act as highly effective surfactants and induce selective interactions between the fat particles and the emulsifiers in the sausage.

The surface activity (Lauridsen, 1976) of the proteins has its origin in the distribution of the hydrophilic and hydrophobic amino acid residues in the primary structure. When the sequence is sufficiently “blocky”, the proteins are highly surface-active, and stable emulsions can be formed easily during processing. Some proteins are denatured mechanically by the energy impact and distributed accordingly between fat and water phases. Such proteins emulsify oil droplets (Figure 87.2). Consequently, the liver sausage with its naturally high fat content forms an emulsion (Figure 87.3) in which the fat droplets form an irregular network.
The droplets interact with specific interactions between the emulsifying proteins.

In many practical cases, such networks may obey random fractal structures; i.e., the size of the cluster $R$ (part of a larger particle network) scales characteristically with the number of droplets, $N$; for example, $R^{d_f} \propto N$, where $d_f$ is the “fractal dimension” (Joshi et al., 2018). When clusters (Figure 87.3) are mechanically stressed, the force is transmitted only through the “simple connecting path” from the top droplet to the last droplet; the dangling parts of the clusters do not transmit forces. As long as this path resists the increasing force, the response is “elastic”. Once the connections between the droplets are broken, only local rearrangements of the droplets with no further increase of the force are necessary to change the form. The deformation is purely plastic, as can be seen in Figure 87.1, and makes a significant contribution to the texture and mouthfeel of liver sausages and patés.

It is tempting to reproduce this behaviour of particle gels in vegetarian and vegan products, and some consumers also demand strongly fat-reduced or even fat-free versions. Often, the fibre inulin, an extract from plant roots, is used as a “fat replacer” in such semi-solid foods, and indeed provides a very fat-like mouthfeel. Given the fact that inulin consists of fructose-based oligomers, between 2 and 120 fructose units, the question is why and how does it work?

Inulin: Selectively Crystallisable Molecules

Nutritionally, inulin is a dietary fibre, exhibiting low energetic content and prebiotic properties, and is suitable for diabetic nutrition. Inulin belongs to the family of the fructans, built up of $\beta$-(2,1) linked D-fructose units and, most often, a terminal D-glucose residue (Joshi et al., 2018; Beccard et al., 2019). Its physical strength is linked to its broad molecular weight distribution. Inulin consists of small units (of slightly sweet taste) and of long units (tasteless). When inulin is immersed in water, the chains dissolve at a given temperature that depends only on the chain length. Short units and chains dissolve first, while long chains do not dissolve at all. Only at temperatures close to 100 °C do all molecules dissolve in water. Consequently, when inulin becomes dissolved at some higher temperature, chains dissolve and crystallise again during cooling according to the chain length; the undissolved material acts as crystallisation nuclei. The result is again an irregular particle network (Bot et al., 2004), of very similar structure to that depicted in Figure 87.3, and, not surprisingly, the texture profile measured in a texture profile analyser appears very similar. The network of crystalline inulin particles is fractal at certain length scales, as shown by the red cluster at the left of Figure 87.4.

At the yield point of the force, the fractal network is destroyed, and the plastic regime takes over. Almost no force is needed apart from constant friction between the particles on increasing deformation. Apart from the details, inulin particle gels resemble very much the behaviour of pasty food like liver sausages.
Thus, inulin is a perfect candidate for building controlled textures that mimic plastic deformations in the mouth to give a “fatty” mouthfeel. However, the mouthfeel in foods containing fats such as pork, goose or duck fat, as well as coconut oil or cocoa butter, also contributes to the melting of the partially crystalline triacylglycerols (“fat molecules”), which cannot be provided by the inulin. The crystals need to be dissolved by the saliva, which has usually different time scales. Nevertheless, the fractal nature of inulin helps here as well.

**Fractal Particle Gels and “Simulated Melting”**

In the mouth, both mechanically induced sensations from compression and shear forces add up to the resulting physical texture. The resulting shear deformations between tongue and palate during oral processing consist of small deformations as well as large amplitudes. It is therefore necessary to study such particle gels under shear, especially in amplitude sweeps, which cover the linear response as well as the non-linear deformation regime of the gels. Whereas the gel supports linear elastic modes quite well (see Figure 87.4 in the low deformation regime), the gel becomes destroyed at larger amplitudes. Fractal particle gels under large deformation amplitudes show a very distinctive behaviour, which has features of melting under mechanical forces, as shown in Figures 87.5 and 87.6.

A typical experimental result is shown in Figure 87.5. The oscillation strain was applied in a rheometer, and the storage modulus was measured (at a frequency of 1 Hz). Small deformations show the linear elastic regime, where the modulus is practically constant, showing only a small decrease. At larger strains, above 20%, the modulus decreases, and the particle gel becomes weaker and weaker. Obviously, the gel structure breaks down. Interestingly, the modulus follows a scaling law, which has been developed in a completely different context, the physics of reinforced polymer materials such as rubbers (Vilgis and Huber, 1999). These experimental results allow for a more quantitative analysis and the development of a physical model for the breakdown and mouthfeel of particle gels in general.

The breakdown of the gel particles with increasing amplitude supports the fractal nature of the structure. As in liver sausages, the gel breaks successively at the weakest points. Since, on average, the broken pieces statistically resemble a similar structure to the large original network, a “scaling law” can be expected. The physical concept of this is shown in Figure 87.6, where the breakdown of the network is schematically illustrated. The large network breaks with increasing deformation into smaller and smaller self-similar pieces. This behaviour can be described mathematically, and the modulus can be predicted (Huber and Vilgis, 1999).

To do this, it is necessary to introduce several “fractal dimensions”. The first is, of course, the fractal dimension $d_f$ itself (see earlier), and the second is the “connectivity dimension” $C$, which describes the connectivity of the network and contains, for example, information on the “weakest path”, where the particle gel is likely to break. The connectivity dimension has two clear limits; the lower limit is 1, corresponding to a linear polymer, for example, while the upper limit is about 1.33 for randomly branched clusters, as in the present case.

After carrying out some mathematical “fractal calculus”, the shear modulus $G'$ for the experiment shown in Figure 87.5 can be described as

$$\frac{G' - G_0}{G_0 - G_\infty} = \frac{1}{1 + K^2 \gamma^{2m}}, \quad m = \frac{1}{C - d_f + 2}$$

This model for the storage modulus thus provides a simple scaling relation between the shear amplitude $\gamma$ and the structural parameters of the inulin particle gel. $K$ is a numerical constant, which reflects the local interactions between the particles forming the gel. When it is fitted to the experimental results, it is possible to extract the fractal dimension $d_f$ with a value of 2.3. The clusters are not space-filling in three dimensions.
Conclusions

The breakdown of the particle gels under oral processing by compression and shear determines to a great extent the physical contribution to the mouthfeel. In addition, the gradual lowering of the friction between tongue, the forming bolus and the palate enhances the pleasure simultaneously with the release of the flavour. Underlying the chemical senses triggered by the simultaneous release of taste and odorant compounds, a large part of the mouthfeel and the texture is driven by physical processes, which are ruled by quite fundamental physical properties. Simple model systems can help to indicate some underlying universal physical concepts. Clearly, the preparation of pure inulin gels and their investigations under ideal laboratory conditions are, indeed, a long way from packing inulin into vegetarian or vegan replacements for pasty sausages for spreading. However, even in complex matrices, the selective crystallisation of hydrophilic oligofructose molecules needs to take place in the water-rich domains, which leads to cluster-rich regions, which are responsible for the “fatty sensation”. Thus, this investigation leads to two basic conclusions. In many pasty and semi-solid foods, the interplay between the destruction of the structure seems to be the key issue for the mouthfeel. It is indeed coupled to many other processes, such as the increase of temperature of the food in the mouth associated with the dissolution of inulin droplets and the coupled accelerated release of flavour compounds. The structure of the particle gel, the size of the secondary particles and the properties of the food matrix define in the end the strength of the oral coating, too, and thus, the time span of the flavour sensation in the oral cavity.

REFERENCES


