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ABSTRACT

Plant based meat surrogates attract increasing interest. Modern methods of biotechnology, food chemistry/technology, and process engineering allow for surrogates with high optical similarity. Nowadays, targeted molecular-sensory methods taste and smell to be largely approximated to the original products. Nevertheless, the products appear completely different on a molecular scale, which is clearly noticeable in texture, oral processing, friction, and bolus formation. A consequent physical consideration of the function and effects of the proteins of different origin reveals the strengths of the respective products and offers suggestions how sensory weak points can be understood better and avoided. This is illustrated here by means of exemplary examples and experiments joined with underpinned by molecular models. Meat sausages, vegetarian, and vegan surrogates are microscopically investigated by rheology, tribology, and tensile experiments. The interpretation of the results is illustrated and supported by simple models.

I. INTRODUCTION

Vegetarian and vegan meat substitutes, such as plant milk instead of cow's milk, vegan cheeses made from nuts or legumes, have become popular. Their significantly lower contribution to harmful climate gases, attention to animal welfare, and ethical reasons, are in the foreground. However, economic interest is also a strong argument. The stock market gains of companies of plant based burgers, meat, and milk product surrogates are considerable. With advanced techniques, it appears possible to create nearly similar products, which resemble meat products. Consumer acceptance increases. Nevertheless, on a molecular scale, plant based surrogates do not have much in common with comparable meat products. Deficiencies in texture, flavor release, and perception become noticeable. This is not surprising, since the biological functions and structures of plant proteins are very different from muscle proteins.[1,2]

Nevertheless, the solution seems to be very simple: proteins are extracted from plants, mixed together, textured, accordingly shaped. Depending on the requirements, oils, salt, and spices are added and the plant based batter is used to produce cooked sausages, minced meat, or vegan roasts. Industrial food technology provides various processes for this purpose,[3,4] to provide alternatives with high consumer acceptance[5] and to create products that are offered on supermarket shelves for specific target groups.[6,7]

Concerning the raw materials, the differences between the original and the plant based imitations are striking: meat consists largely of protein, fat, and water, which are present in a natural food matrix. The fibrous proteins are organized at the molecular level in muscle cells, and soluble proteins are found in the plasma, the meat juice. Each fibril and muscle fiber is encased in a connective tissue, with intramuscular fat between fibers.[8] Plants, on the other hand, are composed of less protein, less fat, but much more cellular material. The molecular organization of these components in protein-rich legumes, seeds, or potatoes differs markedly from that in meat. In seeds and legumes, proteins serve as a storage material to provide a sufficient amount of amino acids and energy during germination until the seeds have sprouted and the cotyledons had penetrated the soil. This observation alone shows that fundamental problems, dictated by the protein function, have to be solved apart from technological possibilities. However, this also shows the impossibility of producing exactly identical meat products from plant proteins if all molecular length and timescales were to be taken into account. Compromises on molecular levels are, therefore, unavoidable.

The use of appropriate protein mixtures, in combination with long chain thickening and gelling agents, try to adopt viscoelastic properties, fracture mechanics, and water binding of the sausages during oral processing. At low deformation speeds or frequencies, the long chain molecules with typical timescales of milliseconds (see, e.g., Mohorovic *et al.*[9] for polymer melts) are able to follow the relative deformation. At high elongation rates, the samples break due to topological restrictions by entanglements in the melts.[10,11]

Such viscoelastic properties are well known in many food system,[8] too, albeit at different times scales, which are determined by molecular properties, architecture, and interactions. A well known example is bread dough. If pulled slowly, the protein chains that span the network can easily follow, but if it is pulled quickly, long chains can no longer disentangle, and the dough reacts by cracking and breaking.[12] The dough is, therefore, viscoelastic, at high deformation speeds the required pulling force is high, becomes higher and higher until it breaks, whereas at low deformation speeds, the dough behaves more like a liquid. Such remarks hold similarly for high viscose honeys,[13] which do not contain long chain molecules, and hence, show little elasticity, but strongly hydrogen bounded natural sugar compounds and water molecules, responsible for highly viscous systems. A spoon inserted in highly viscous honey sinks slowly without much resistance in the jar under its own weight. When trying to push the spoon in quickly, the force required to do so exceeds many times the weight of the spoon, as the sugar molecules, which are strongly bonded by water, first have to be moved out of the way to make room for the spoon. These intuitive examples show and illustrate two almost extreme cases in the flow behavior of foods.

It is, therefore, hardly surprising if many current products always lack the last bit in flavor release, texture but also mouthfeel, especially the friction of the food pulp between tongue and palate, or during the swallowing process. The oral processing and the associated flavor release are noticeably different. Direct comparisons between the original products and the surrogates are difficult, because a whole range of product characteristics depends on the multitude of possible recipes and the different manufacturing processes. In order to be able to better classify the differences between animal, vegetarian, and purely plant-based sausage products, there is no way around looking at the molecular interaction of the various components.

In this paper, the focus is put on scalded sausages, which are another culinary examples of viscoelastic materials. The findings not only provide initial information about the interaction of the proteins and biopolymers, the so-called hydrocolloids in the sausages, but also about the mouthfeel. Chewing, biting, and all subsequent processes during oral processing are determined in the mechanical-rheological investigations on the viscoelastic properties of the sausages.

The nonlinear flow properties (rheology) and fracture mechanics are investigated in more detail using tensile testing. These methods provide objective data that can be used to measure and explore the interaction of the molecular components. The physical properties of the sausages are investigated in rheological and stress–strain–fracture measurements. These mechanical results are complemented by tribology.

Tribology gained during the last decade importance for understanding the physical sensory aspect during oral processing.[14] When solid foods, for example, the sausages, are put into the mouth, it becomes destroyed by mastication and chewing. This process addresses the elasticity, brittleness, and the fracture properties during the first bites. The pieces become smaller and smaller. At the same time, the fragments are salivated, and the bolus starts to form. The food in the mouth becomes pasty, and friction always plays a greater role. The oral processing enters the tribological regime. As the fragments become even smaller and the bolus becomes swallowable, the flow processes are determined by both rheological and tribological processes.[15,16]

The combined results from rheology, stress–strain-experiments, and tribology will provide a deeper insight into the molecular and physical aspects of mouthfeel and perception of the different sausages. Moreover, they allow predictions for alternative plant based proteins, their function, and their role on nano-scales in plant based food preparations.

II. MATERIALS AND METHODS

The German company "Rügenwalder Mühle" (Rügenwalder Mühle Carl Müller GmbH, Bad Zwischenahn, Germany) produces scalded meat sausages, vegetarian, and vegan sausages. For vegetarian analogues, chicken egg white powder, binding and gelling agents are used to form the structure. For vegan surrogates extruded proteins, protein powder can be used together with the hydrocolloids and other additives in combined mixing and cutter processes. The company provides them in packed slices and are suitable for systematic comparison. These versions have been bought in local supermarkets for the current investigations. Table I shows which types of molecules act at different technological and sensory levels. This breakdown shows the idea of how surrogates have to be constructed in order to sufficiently approximate the properties of scalded sausages. Meat already brings sufficient

TABLE I. Ingredients in the various sausages types according to the manufacturer.

Type of ingredient[a]	Meat	Vegetarian	Vegan
Functional proteins	Pork meat	Egg white	Isolated pea and potato proteins
Functional polymers (carbohydrates/hydrocolloids)	...	Tara gum, locust bean gum, Xanthan gum, inulin	Carrageenan, konjac gum, Locust bean gum
Fat/oil	Pork fat	Canola oil	Canola oil
Aids, flavor (aroma)	Glucose sirup, dextrose, Salt, diphosphates	Salt	Distilled vinegar, glucose sirup, Salt, processed Eucheuma seaweed

[a]Spices and other nonfunctional ingredients are not listed.

functional properties, no further structure-giving carbohydrates, such as starch, biopolymers (hydrocolloids), or fibers, are necessary. Only the ingredients required for cutter aids (mainly phosphates), humectants (for example glucose sirup, glucose), and salt for flavor and the adjustment of ionic strength are sufficient to scald sausages suitable for consumption that approximate traditional, artisan techniques. The total composition of the ingredients is found in Appendix for convenience.

A. Rheology

Rheological measurements were performed with a Bohlin Instruments Gemini 200 rheometer (Malvern Panalytical Ltd., Malvern, UK) equipped with a 25 mm parallel plate geometry. The sausage slices are placed between the plates of a rheometer. Afterward, the upper plate is set into sinusoidal oscillatory motions with a frequency of 1 Hz, systematically increasing the deformation amplitude. To prevent sample slipping, both plates were covered with 80 grit sandpaper. Before placing under the geometry, the sample was cut out using a round stencil (ø 25 mm). The gap size was stepwise adjusted to the thickness of the slices until an increase in the normal force was detected leading to gap sizes between 950 and 1200 μm. Oscillatory amplitude sweeps were performed from 0.01% to 200% strain with a frequency of 1 Hz at 5 °C, while recording the storage (elastic) modulus, G', and loss (viscous) modulus, G''. The temperature ramp measurements were performed with 0.2% strain and 1 Hz starting at 5 to 40 °C with a rate of 1 K/min. A equilibrium time of 5 min at 5 °C and 10 min at 40 °C was set before and after the ramp, respectively, was performed. Storage (elastic) modulus G', and loss (viscous) modulus G'', were recorded. Each sample was at measured at least in triplets.

B. Tensile tests

The tensile test were performed with a ZwickRoell Universal Testing Machine AllroundLine Z005 (ZwickRoell GmbH, Ulm, Germany). From the individual sausage slices, bone shaped samples were cut (Probe area $32 \times 4\,\text{mm}^2$). The samples were clamped carefully straight into the sample holder. The samples were stretched with 1 mm/s until the samples ruptured. All measurements were performed at room temperature with samples cooled to 5 °C, while recording strain and force. To obtain the values of the Young moduli E, the data points above a threshold of 0.01 % until 0.2 % strain were fitted by a linear regression. Each sample was measured at least three times.

C. Tribology

The friction of the different samples was measured using a Discovery HR 3 rheometer (TA Instruments, New Castle, USA) equipped with a tribology test setup. Three balls on a plate geometry was used, consisting of three balls (ø 8 mm) in the upper part arranged on a round plate (ø 30 mm) and a stationary lower plate attached with a silicone rubber (ø 40 and 1 mm thick). This leads to a point contact between a stainless steel and rubber surface. To provide a uniform solid-solid contact and axial forces during rotation an aluminum spring beam coupling was used (specified by Trios manual 2019). The tribology measurements were performed at 25 °C with a normal force of 1 N. At this normal force, the contact area was calculated 6.5 mm^2 using approximations[17] based on the hertzian contact theory[18,19] for a sphere with a modulus of about 200 GPa and a silicone rubber surface with a modulus of about 0.5 MPa.

The sliding speed was conducted from 100 to 100.000 μm/s. The friction coefficient μ was calculated as the ratio of the measured frictional force to normal force and is given as a function of the sliding speed (μm/s) for each testing. For the measurements of the intact sausage slices, round specimens (ø 30 mm) were cut with stencil. For the measurement of the bolus like samples, 10 g of each sausage was purred with 50 wt. % demineralized water in an IKA ULTRA-TURRAX (IKA- Werke GmbH & Co KG, Staufen, Germany) equipped with a 20 ml grinding tube. Tribology tests were carried out in triplicate.

III. RESULTS

A. Rheology of meat, vegetarian, and vegan sausages

The viscoelastic properties are characterized by two parameters that are measured during the sinusoidal deformation, the elastic modulus G' (storage modulus), and the viscous modulus G'' (loss modulus) for the scalded meat sausages show typical curves, which are shown in Fig. 1. In general, a surprisingly broad and clear consensus between all three versions can be seen. For small deformations between 0.01% and 1%, the linear viscoelastic regime (LVR) is shown. The storage and loss moduli, G' and G'', do not depend on the deformation. Moreover, in this deformation range, the storage moduli G' are larger than the loss moduli G'' for all three sausages up to 100% of deformation, expressing the dominance of elasticity over viscous flow proprieties. All sausages are soft solids. Within the LVR, the sausages behave rubber-like and recover from the small deformations. Above 1% deformation, the moduli decrease. At about 100% deformation, the storage and loss modulus become equal. The viscous properties of the sausages play a greater role when structural changes become significant. The values for G', G'' within the LVR and the LVR limit are summarized in Table II.

The agreement between the different types of sausages in G' is indeed remarkable, given be the different composition and structure of

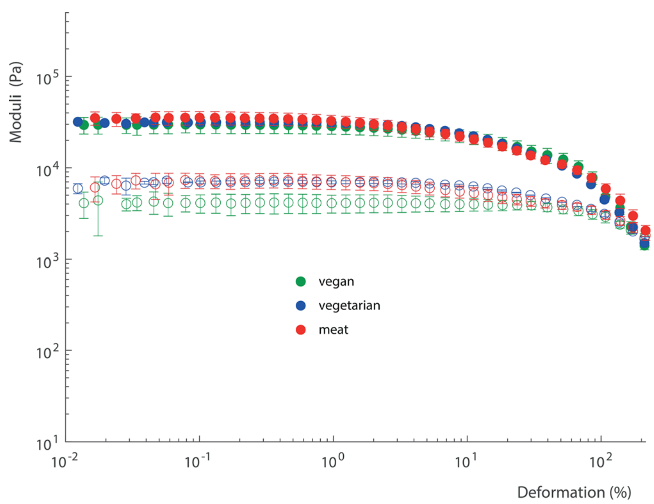

FIG. 1. Amplitude sweep of meat (red), vegetarian (blue), and vegan (green) sausages. The filled samples correspond to the storage, or shear modulus G', the open symbols to the loss modulus G''.

TABLE II. Summary of the mechanical properties of the different sausage types, Young modulus from tensile experiments, storage, loss modulus, and linear visco-elastic range from rheology.

	Meat	Vegetarian	Vegan
E (mN/mm^2)	7.2 ± 0.7	5.4 ± 0.1	4.3 ± 0.1
G'_0 (Pa)	35 180.0 ± 5653.7	31 286.0 ± 866.7	29 652.4 ± 6123.3
G''_0 (Pa)	6863.8 ± 1639.0	6781.7 ± 548.1	4157.2 ± 1212.4
LVR (%)	1.60 %	4.12 %	3.25 %

the samples. Only slight deviations in the LVR and in the non-linear deformation regime can be observed. Only the data for the loss modulus G'' show more pronounced differences between the samples, especially for the vegan preparation. The values for G'' are with 4157.2 ± 1212.4 Pa significantly lower compared to the vegetarian and the meat sausages (6781.7 ± 548.1 Pa, respectively, 6863.8 ± 1639.0 Pa).

This observation is supported by the plot of the loss factor tanδ, shown in Fig. 2. The loss factor is significantly lower for the vegan version compared to the vegetarian and meat sausages up to 100% of deformation.

Additionally, temperature ramps under oscillatory shear have been performed, which show the change in the mechanical properties in the limit of small amplitudes (0.2% in the linear regime). This experiments allow some insight in the behavior during oral processing, when the temperature of the sausages increases. Figure 3 shows the results of the different versions of the sausages.

This experiment gives an indication of the behavior of the different sausages under the temperature increase in the mouth. Normally they are eaten chilled, but the temperature rises during oral processing. In the meat version, the fat first becomes waxy at oral temperatures; then, it melts. Consequently, the meat sausage shows the most significant decrease in G' and G''. In the vegetarian and vegan versions, the vegetable oil, canola oil in both cases, always remains liquid in these temperature ranges, and its viscosity only decreases slightly. The influence of the oils on the moduli is, therefore, significantly lower. Surprisingly, the moduli change least in the vegan version. These points will be discussed again below.

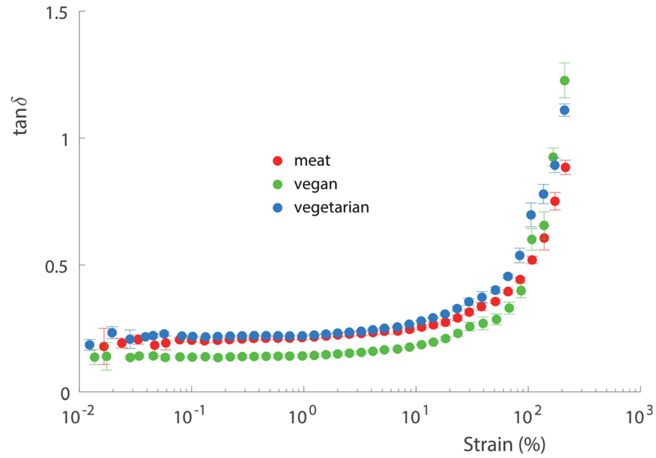

FIG. 2. The loss factor tan $\delta = G''/G'$ for meat (red), vegetarian (blue), and vegan (green) sausages for the results in Fig. 1.

B. Tensile tests: Stress–strain–fracture experiments

To understand the mechanical behavior better, further experimental methods need to be consulted. For this case, tensile tests were chosen, which provide in addition to fracture stress and fracture elongation more information on the deformation behavior during elongation. The form of the progression of the different stress–strain curves allows conclusions about the elastic behavior and the influence of the

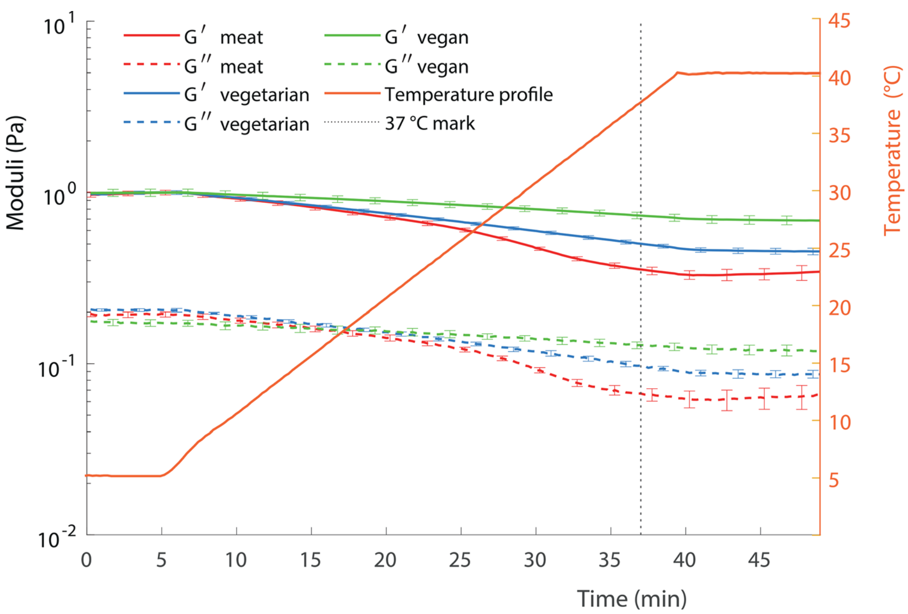

FIG. 3. The normalized moduli G' and G'' for the different sausages as a function of time and increasing temperature from 5 to 40 °C. The vertical dashed line corresponds to the average mouth temperature 37 °C. Measurements have been carried out in triplets. Note that G' of the different samples have been normalized to 1 Pa, to illustrate the differences during heating explicitly.

different materials compositions. However, tensile tests are hardly common for food, unlike in materials research. Nevertheless, in this case, they allow a very special insight into the physics of sausages and their surrogates. In tensile tests, specimen (in standardized) shapes are punched out of the materials, clamped in tensile machines, and stretched at a constant speed until they break.

Force-deformation curves are measured, from which the elastic modulus, maximum fracture stress, and fracture strain can be determined. These can then be compared very well with molecular models. Thus, some information about the shape changes and molecular interactions of the structure-giving proteins and hydrocolloids during deformation can be concluded, as well-known from rubber-like materials.[20–22]

The results are summarized in Fig. 4. The experiments were carried out in multiple determinations, since all sausages preparations contain pieces of vegetables and small air inclusions, predetermined breaking points occur at these interfaces, which may influence the elongation at break. For the sake of simplicity, only mean value of the results is shown, which displays the trend of all tested samples. For the determination of the initial moduli (Young moduli) E, the slopes at small deformations (up to 0.2%) have been fitted by a straight line following Hooke's law. The fitted values for the initial moduli from the tensile experiments E-module and rheology, and G', G'', and LVR are also summarized in Table II.

Compared to the surrogates, the meat sausages show as expected the highest modulus in the tensile experiments. This suggests a highly cross-linked protein network, but also a contribution by the solid pork fat particles. However, the shape of the force-strain curves also provides further information. These reveal significant larger differences, in contrast to the rheological experiments. The meat specimens showed a rapid increase in force, which even under 2.5%–3% deformation still becomes stronger with increasing elongation, indicating a significant strain-hardening. The fracture strain (or ultimate deformation) corresponds to the maximum force and its sharp drop. For the meat sausage, this happens already after 11%–12% elongation. The

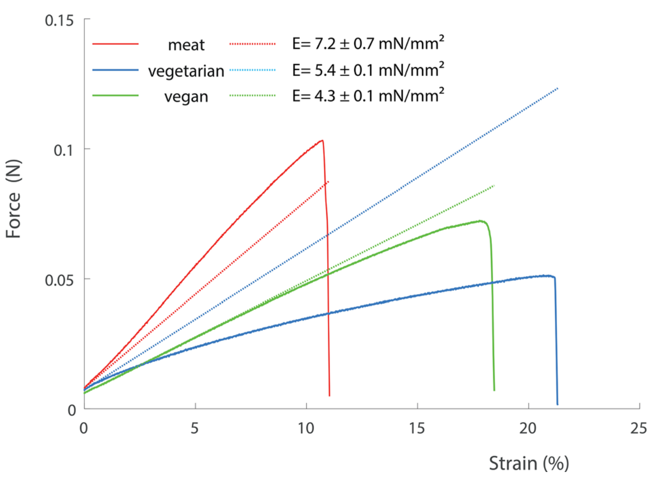

FIG. 4. Stress–strain curves of meat (red), vegetarian (blue), and vegan (green) sausages. Typical results of the tensile test show clear differences in deformation behavior. The dashed lines correspond to the linear fits (initial Young modulus (E in mN/mm^2). Meat sausages show a significant strain hardening, vegan sausages a weak, less pronounced strain hardening, vegetarian sausages a significant strain softening.

ultimate force of the meat sausage shows the highest value between all the types. This indicates already a very tightly meshed complex protein and fat filled network. The vegan version shows a rather linearly increasing curve almost within the entire deformation range. The ultimate force of the vegetarian version is significantly lower, the breaking strain considerably larger. The vegan version shows a less pronounced strain hardening for deformations before it gets softer before its fracture. The vegan version based on hydrocolloids, pea, and potato proteins shows lower forces than the meat version, but higher force values than the vegetarian sample. Both the breaking strength and the elongation at break of the vegan version lie between those of the meat sausage and the vegetarian version. Compared to the meat sausage, the vegetarian, egg white/hydrocolloid based vegetarian version shows a much softer behavior. The force– extension curve shows a typical softening behavior, which is also well-known from rubber-like solids.[23]

C. Tribology

Tribology experiments are performed to quantify the mouthfeel sensation during oral processing, mastication, and bolus formation on physical grounds. The method of tribology is more suitable for this, because the friction that food creates between the tongue and the palate when crushed in the mouth determines part of the mouthfeel, in addition to texture.[15] These effects are well known. For example, when a low-fat cheese is masticated in the mouth, a dry sensation occurs in the mouth that does not improve significantly during further chewing and despite cumulative salivation. High-fat cheese, on the other hand, releases the emulsified fat during chewing, deposits itself on the tongue and palate and acts there like a lubricant. Friction is reduced and oral processing becomes more pleasurable. The food pulp becomes significantly softer, as the free fat also surrounds the particles produced during chewing and reduces friction within the bolus. The food particles forming the bolus slide off each other more easily, and the bolus becomes significantly more plastic and is easier to swallow.

To quantify these effects, the friction coefficients as a function of the sliding speed are measured. In the simplest case, these result from the ratio of the force required to pull a body over a surface and its weight (i.e., the normal force with which the body presses or is pressed onto the surface).

Two different experiments for the sausages turn out to be useful. On the one hand, the friction can be determined directly on the intact sausage slices, which corresponds to the first touch mouthfeel, on the other hand, in a puree, which resembles an already formed bolus. For this purpose, the friction tool, in this case with the "three-ball geometry", is placed on the samples with the force of 1 N and rotated over them with increasing rotational speed. The speed-dependent coefficient of friction is determined by measuring the resistance force.

The friction on the intact slices at low sliding speeds determines the initial attrition properties of the sausage until the three ball tool abrades particles from the surface as the speed of rotation increases, measuring not only the property directly on the surface but also the layers deeper and the resistance offered by the resulting particles. In the second case, the sausage slices of each sample were pureed with demineralized water (50% of sample weight) with a ULTRA-TURRAX lab blender and the friction of the purees was measured. This corresponds approximately to an already chewed bolus, with the restriction that the influence of salivary proteins is then not taken into account. The results are shown in Fig. 5.

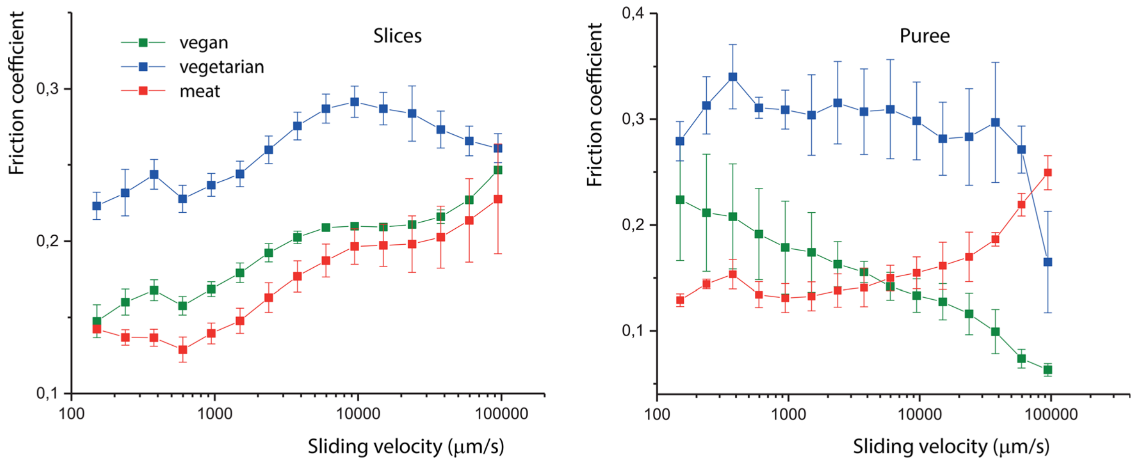

FIG. 5. The friction coefficients of the different sausages as a function of sliding speed are clearly distinguishable, measured both on the slices (left) and on the puree (right). The slices are the original from the package, cut to the tribometer plate, the puree has been processed by an ultra turrax.

IV. DISCUSSION

A. General remarks

Despite the apparently good sensory agreement between the different sausages, it is noticeable that the loss modulus of the vegan version systematically lies in the linear viscoelastic range below the loss moduli of the meat and the vegetarian version. The viscous parts of the deformation are therefore lower and therefore less viscous, which affects the mouthfeel. The vegan version is slightly pulpier when squashed between tongue and palate. A closer look into the molecular structure and interactions is useful to understand these remarkable differences better. In the following models for the sausages with different composition will be developed.

Common to all sausages are the basic steps of preparation. The different components are put into a cutter and blended under certain speeds.[24] The resulting emulsion will be heated (scalded) to cross-link the proteins, and the emulsion becomes a soft solid. Oil and fat droplets will be embedded in a protein, or protein-biopolymer-matrix.

However, the differences can be understood by having closer looks into the different composition, the molecular properties of the compounds, and their interactions on molecular scales. Meat sausages are composed of meat, fat, and water. Vegetarian versions of cooked sausages make use of egg whites. However, egg white consists by far fewer proteins than meat, which show globular structure, whereas most meat proteins are fibrous. Therefore, the consistency is supported by biopolymers and hydrocolloids. Good candidates for foods are xanthan gum, locust bean gum, and tara gum. They are also known for molecular synergies.[25] The combination of the latter two with xanthan gum provides a separate network after scalding. Also oligofructose (inulin) is added, to support a "fatty" mouthfeel.[26–28]

B. Scalded meat sausages

For meat sausages, muscle meat, fat, and chopped and grounded ice will be placed in the cutter. The knifes grind the meat and the fat to smaller and smaller pieces until a fine batter emerges.[29] The cutter speeds (and thus, the shear rates) primarily determine the resulting particle size and its distribution. Sausages with a fine, smooth batter require high shear rates and longer times for good homogenization. At temperatures kept low by finely ground ice ("snow") cooling (at temperatures between 10 and 12 °C), fat remains quasi-solid, meat is broken down, and muscle proteins are released and can contribute to the emulsifying effect. The meat proteins must emulsify both the added water (as ice) and the fat in a homogeneous manner, which ensures stability, necessary to fix the structure during cooking, which defines the texture by the physical and sensory properties during oral processing.[30–34] Meat sausages contain naturally a broad composition of proteins. The most important ones are the proteins of the muscle motor, actin and myosin, water soluble, globular proteins (and enzymes) dissolved in the sarcoplasm, and collagen from connective tissue. Due to *rigor mortis*, myosin and actin are bound to a fixed complex by ionic binding via oppositely charged amino acids. During cutting and by aid of adding a phosphates and monovalent salts,[35] a fraction of the myosin becomes dissolved from the actin-myosin-complex,[36] which is essential for the structure of the final emulsions. Myosin in skeletal muscles is composed of head, neck, and tail domains. The heads are able to bind to the filamentous actin by using the hydrolysis of fourfold negatively charged adenosintriphosphate (ATP) and contracts the myofilaments for muscle motion. The heads are bound to the neck domain and acts as a linker and as a lever for transducing force. The tail domain joins two dimers by a helix into the myosin filaments.[37] Fibrillar actin itself is a linear helix bundle of globular actin sub-units. Another main protein fraction consists of a large concentration of water soluble, globular sarcoplasmic proteins, such as myoglobin, heat-shock-proteins, repair enzymes, and many other types. The exact composition of the sarcoplasmatic proteins is not known in detail. All these very differently structured proteins have different physical functions during emulsification and cooking.

During cutting and mixing under low temperature, the different components become arranged between the fat particles. Proteins adsorb preferably with their hydrophobic parts at the surfaces of the solid fat particles. Especially free myosin with their largely hydrophobic parts of their primary sequence.

To understand the structure and the mechanical properties of scalded meat sausages better, it is useful to remind of the important structural and thermal properties of the most important muscle proteins.[38] Therefore, to illustrate and understand the function of the function of the proteins, these results are summarized in Fig. 6. The denaturing of the different muscle proteins and their structural changes has been extensively described in.[8] The denaturation of the myosin heads in pork starts already about 45 °C, whereas the helical tails denature between 50 and 55 °C.[38] The globular sarcoplasmic proteins change their structure within the broad peak in Fig. 6 between 58 and 63 °C, whereas the collagen (connective tissue) starts to denature between 62 and 68 °C. Fibrillar actin, the most stable muscle protein, denatures only at temperatures larger than 70 °C.[39,40] The different thermal properties of fats and proteins suggest a scalding in different temperature steps. A first scalding step at between 48 and 50 °C melts the pork fat and denatures simultaneously the myosin heads. The second step around 60 °C denatures the sarcoplasmic proteins. The globules unfold partly form loose networks and contribute strongly to the binding of the water inside the network.[41] Similarly the helices of the myosin start to denature and contribute to the protein network formation. Connective tissue consists mainly of collagen fibers composed of triple helices.[42,43] Their unwinding requires longer timescales.[44] The final temperature step at about 72 °C fixes the network by establishing cross-links by disulfide linkage of cysteine of different proteins.[45–47] At such temperatures, the viscoelastic meat batter solidifies, water is bound in the process, and a strong gel is formed. A simple model is illustrated in Fig. 7.

The complex network proposed in Fig. 7 in meat-based cooked sausage is very tightly meshed. Its degree of cross-linking is very high due to the large number of free cysteine amino acids available in myosin and the sarcoplasmatic proteins. At the same time, a high number of stiffening elements are found in the scalded sausage network, such as the muscle fibers, free fibrillar actin, partially cross-linked collagen and gelatin networks. The solid fat particles and destroyed myofibrils contribute as reinforcing filler components, which enhance the shear modulus as well as to the loss modulus in similar ways as in reinforced

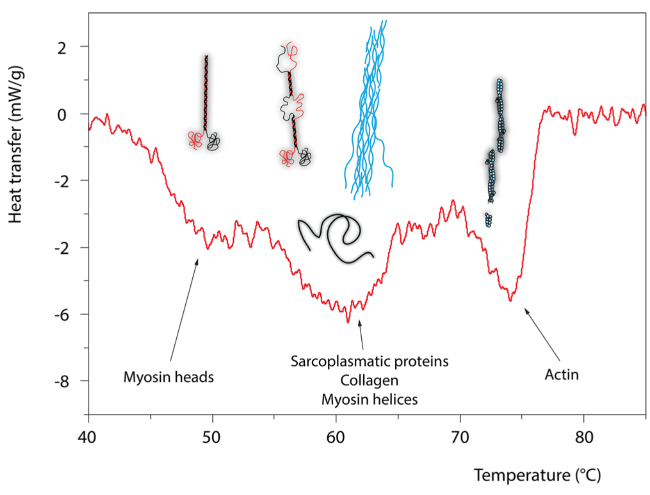

FIG. 6. Typical DSC of pork muscle meat. The denaturation and structural models of the relevant proteins are indicated. The relevant proteins are shown on top from left to right: myosin bundle (homo-2-mer) containing two chains and two heads, drawn in red and black. Myosin bundle at higher temperatures, the tails start to denature, collagen bundles (in blue), sarcoplasmatic protein (black), and actin.

network systems.[48] Consequently, the modulus is relatively high and the ultimate deformation is low, as can be seen in the rapid increase in force and the small maximum elongation in Fig. 4. The meat sausage is highly elastic and has, therefore, a convincing "crack" by the first bite and the induced crack propagation throughout the solid meat emulsion.[49]

C. Vegetarian sausages

The solid matrix of the vegetarian version is mainly carried by the egg white. Egg white is a perfectly cross-linkable system, as known from egg cooking. Egg white roughly resembles the sarcoplasm of meat, as it contains water and dissolved globular albumins that can unfold and cross-link when heated above 72 °C.[50] The main representative protein in egg white, ovalbumin, proves to provide sufficiently good network forming abilities with promising results. Ovalbumin is with 386 amino acids sufficiently long and contains unsaturated, non-bound cysteine for rapid cross-link formation, as illustrated in Fig. 8. Additionally unbound cysteine is found toward both the N- and the C-Terminal of the chain. The ovalbumin network fraction contains wider meshes; its extensibility will be higher. The ultimate deformation of the vegetarian version can be expected to be larger, as seen in the experiments shown in Fig. 4.

Most of the physical and functional properties of proteins are already hidden in their primary structure, as can be recognized, for example, from the ovalbumin in Fig. 8. In contrast to natural meat proteins, the structure and function of ovalbumin are quite different. However, their use in meat replacing batters requires the adoption of two tasks. On the one hand, the emulsifying ability must be ensured, and on the other hand, a network formation resembles a sausage as much as possible. The surface activity for ovalbumin is well known from its foaming and emulsification properties.[51] Thus, the cross-linking and properties require more attention here. Apart from the water solubility of the amino acids, the primary structure offers also information about the locations of cysteine and disulfide bonds in the native state, as shown explicitly in Fig. 8. As indicated there, ovalbumin contains cysteine with reactive groups close to the termini of the protein and one internal disulfide bond.

The elastically "wasted loop" in the middle of the ovalbumin chain, via the closed disulfide bridge, plays only a subordinate role,[52] because the denatured protein chains already have sufficient functionality. Moreover, such disulfide bridges can open with a certain probability at a pH value around 7 and higher,[53] so that further cross-linkable cysteine is available. At the same time, ovalbumin is an excellent emulsifier and can bind the oil added to the mixing process. Another advantage of the egg-white proteins is their excellent emulsifying properties. The arraignment of hydrophobic and hydrophilic amino acids in their primary structure ensures stable emulsions[54,55] also for culinary applications.[8,56] Other proteins of the egg white, such as conalbumin and ovotransferrin, are highly intramolecularly cross-linked and occur in smaller concentration compared to ovalbumin. Therefore, they are neglected for the structure formation in a first approximation.

The added hydrocolloids, xanthan gum (XG) and locust bean gum (LBG), play a more important role, for the structure and the resulting mouthfeel. Pure xanthan solutions quickly form a ketchup-like strongly shear-thinning solution after mixing via an "orientation glass,"[57,58] while LBG and tara gum (TG) show strong thickening

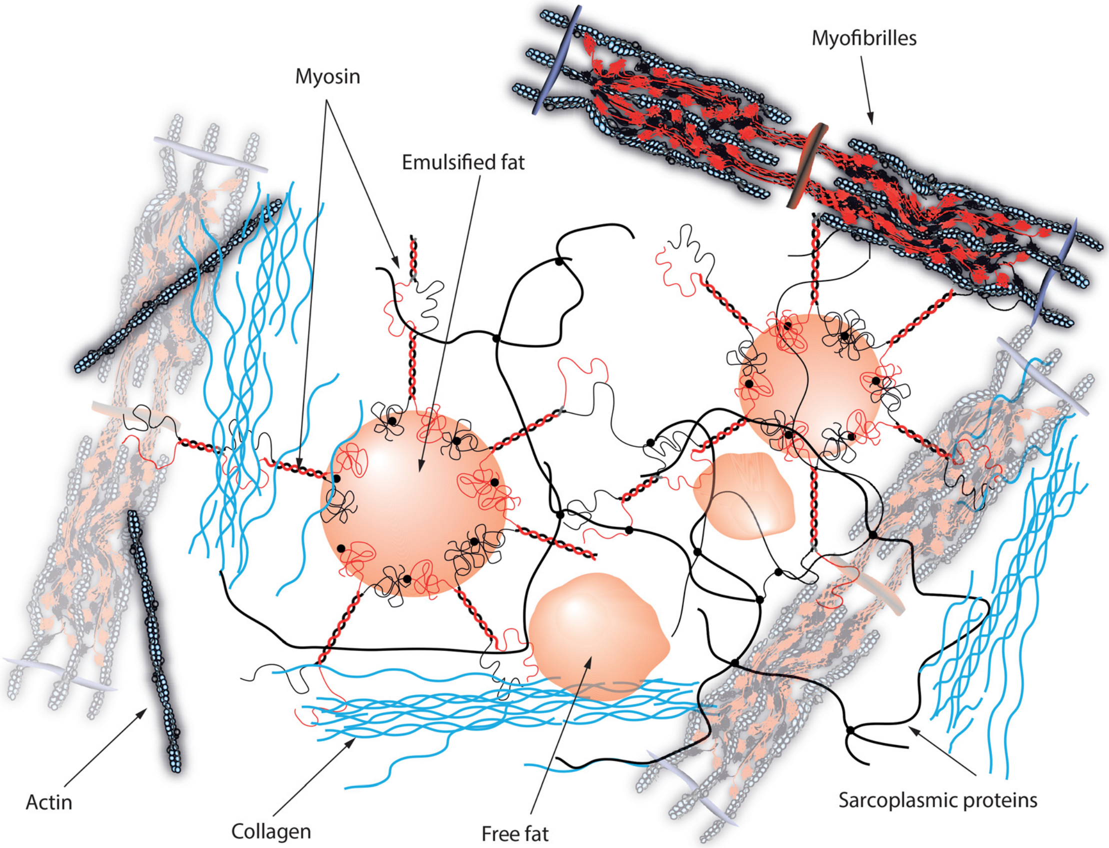

FIG. 7. A simple model for the micro structure of a boiled (scaled) and cooled sausage, ready to eat. The cysteine cross-links (disulfide bridges) are indicated by the black dots. The mean distance between two cross-links defines a mean mesh size of the network. The pork fat is mostly solid at room temperatures and below.

effects, and the viscosity increases.[59] However, when XG and LBG are mixed and heated, both act synergistic and form a gel,[60] which competes with the egg white gel. At this point, the kinetics of network formation comes into play. The free cysteine of ovalbumin is closed by chemical reactions within a few minutes during heating, whereas XG and LBG require considerably longer times for network formation. The gel sets only under cooling.[61]

Consequently, the ovalbumin cross-links first and fixes a network under heating. Due to high viscosity, excluded volume repulsion, and restricted (phase) space, the interpenetration of XG or LBG chains into the protein network is low. Therefore, the hydrocolloids need to form a separate and independent network for kinetic reasons. Thus, it is reasonable to assume that protein and hydrocolloid networks merely interlock and provide high plasticity and larger deformations. These considerations lead to the structural model proposed in Fig. 9. In contrast to the pork fat in meat sausages, plant oils remain liquid at temperatures between 0 °C and higher, i.e., process, storages, and eating temperatures.

The orange circles in Fig. 9 indicate closed disulfide bridges, whereas the small beads at the ends of the ovalbumin still open cysteine groups. The free ends have in the dense, highly viscous environment, only little chance to come so close that they are able to react. The network remains relatively soft and is less tightly cross-linked. The two separated networks allow for larger deformations compared to meat, which is shown by the low modulus and the larger extensions in Fig. 4 before fracture. It is worthwhile to notice that the canola oil droplets are preferably located in the protein-rich parts of the network, since the hydrocolloid regions are largely hydrophobic.

The matrix of the vegetarian sausage batter is highly viscous after mixing and cutting, so that balancing processes are hardly triggered. XG is a relatively stiff, almost rod-like shaped molecule that carries a high negative charge. Therefore, the molecules repel each other strongly. The electrically uncharged but highly polar molecules of LBG and TG act synergistic with XG after scalding. They form networks with the rods via hydrogen bridges. Due to the restrictions of the already formed ovalbumin gel, the XG-LBG-TG gel cannot spread throughout the space. Therefore, they act as soft filler gels, which mainly contribute to the elastic modulus via their volume, i.e., hydrodynamic network reinforcement.[48] The entire network is largely spanned up loosely by ovalbumin, which is cross-linked because of the positioning of the cysteine at the ends. However, not all ends are close enough to be cross-linked. The oil and the hydrocolloid filler material keep them separate and uncross-linked, since the ovalbumin proteins are too short in their total length. At the same time, they still act as emulsifiers on the oil droplets. There, a part of the hydrophobic amino acids is held, and the freedom of

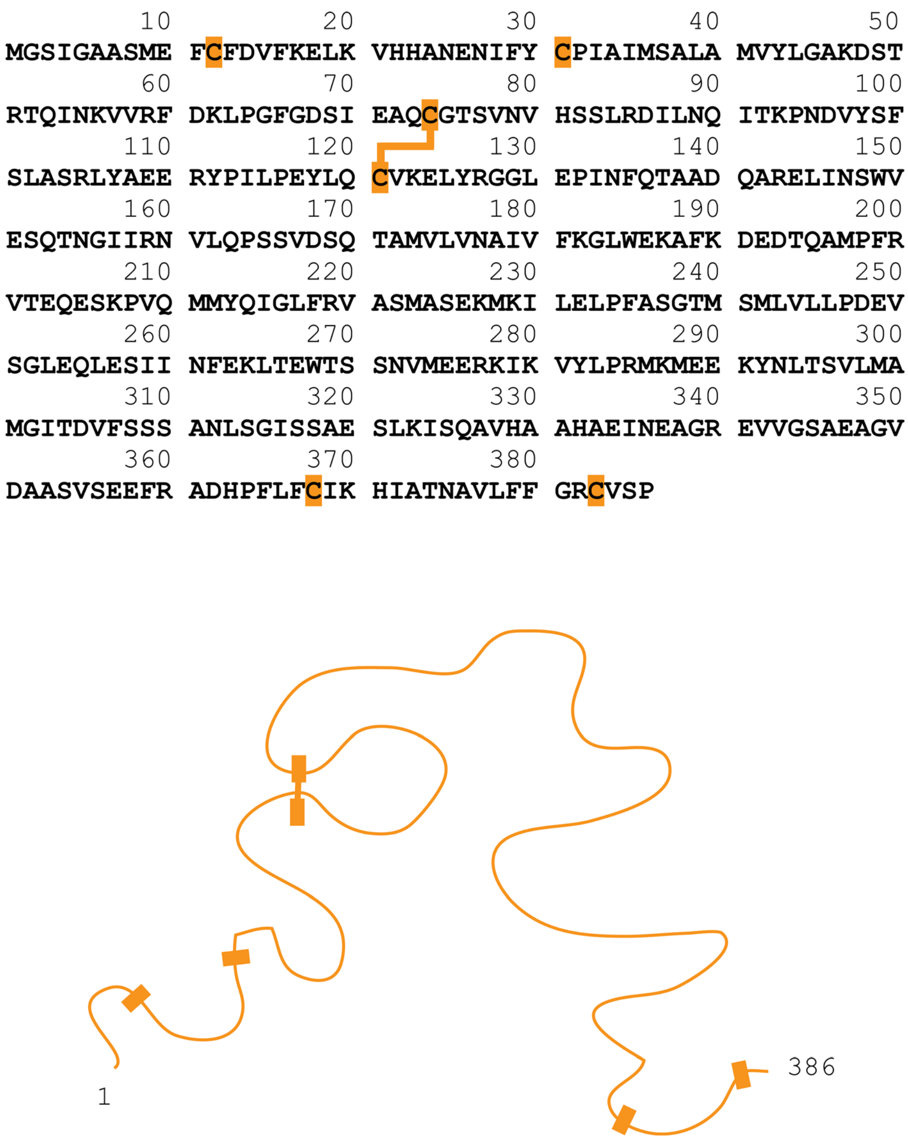

FIG. 8. The primary structure of albumin in the single-letter code shows free cysteines C (highlighted in orange) and two cysteine form a disulfide bridges (orange line). In a partially denatured state, the protein chain contains a disulfide bridge still intact forming a elastically "wasted loop." (Primary sequence taken from Uniprot: P01012 OVAL-CHICK).

movement of the free ends of the ovalbumin is additionally restricted. Already some connections remain open, i.e., the network becomes more wide-meshed than that of the meat sausage. The storage modulus is lower in comparison, but the elongation at break is significantly higher, as can be seen in the tensile test in Fig. 4.

The model shown in Fig. 9 is oversimplified and assumes that the ovalbumin denatures completely by losing all secondary structures, such as helices and β-sheets.[62] However, heating of ovalbumin increases the amount of the β-sheets in the protein.[63] These are hydrophobic and contribute as emulsifiers when these part locate at the surface of the oil droplets.

D. Vegan sausage

The matrix of the vegan variant of the cooked sausage uses a protein mixture of pea protein isolate and potato protein, combination with LBG and konjac gum (KG). The two protein fractions have significantly very different cross-linking properties, as can be seen from the primary structures. The pea protein has two main functional components, albumins and legumins. Legumins in peas are largely similar and have protein chains with over 500 amino acids, as can be seen in the two representative examples in Fig. 10.

To gain a deeper understanding of the function of the pea proteins in the surrogates, it is useful to take a more detailed look at the primary structure. The minimum requirements of plant based sausages for a pleasant mouthfeel are again sufficient surface activity for stable emulsions and tight cross-linking of the proteins. Figure 10 shows the amino acids in one letter code. Hydrophobic amino acids are shaded in blue color. Hydrophilic amino acids are either positively charged (shaded in green), negatively charged (shaded in red), or polar (non-shaded). The charged amino acids are highly water soluble and

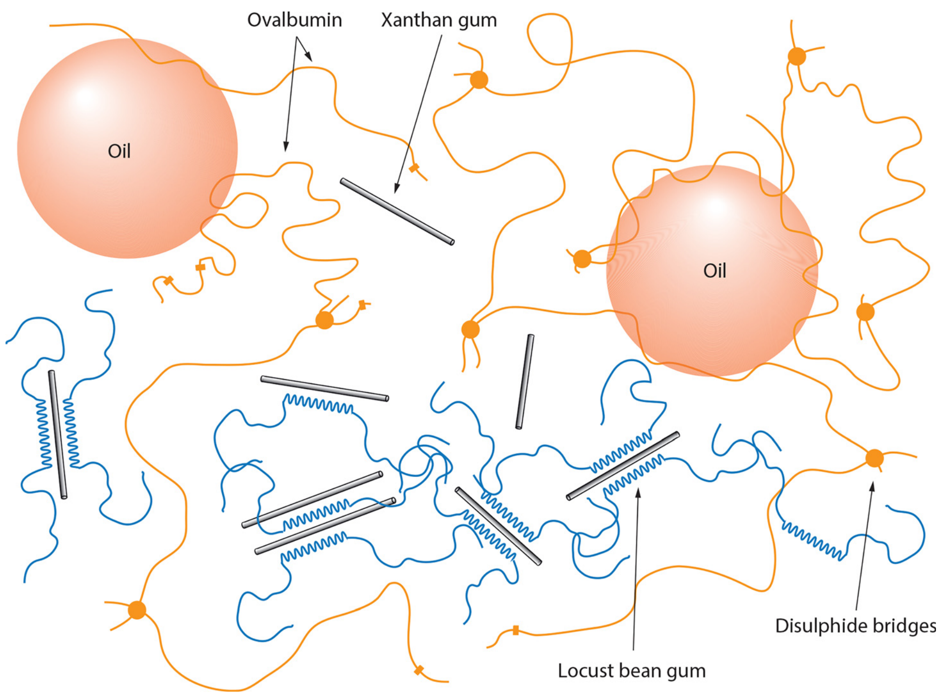

FIG. 9. Simple model of the vegetarian sausage after scalding and cooling. Oil droplets are emulsified by egg white proteins, mainly ovalbumin (drawn in orange). These proteins cross-link (orange circles) partially amongst each other and form a loose network. The XG and LBG chains form a separate hydrocolloid network. The model for XG-LBG-interaction is adopted from Zhan *et al.*[61]

are sufficiently separated by hydrophobic or polar amino acids. Hence, the protein in fully denatured form resembles a amphiphilic "block copolymer" with a sufficient surface activity to emulsify the added oil. Pea proteins act as emulsifiers. Additionally, the protein shows internal disulfide bonds. The connected cysteines are labeled in orange. As in ovalbumin, these indicate internal loops in their connectivity. Nevertheless show the legumins free, unbound cysteines in the "chains" in both termini, shown by the orange frames around the cysteines, which provide the formation of cross-links to provide the elastic an d fracture properties required for a pleasant mouthfeel of the vegan surrogates.

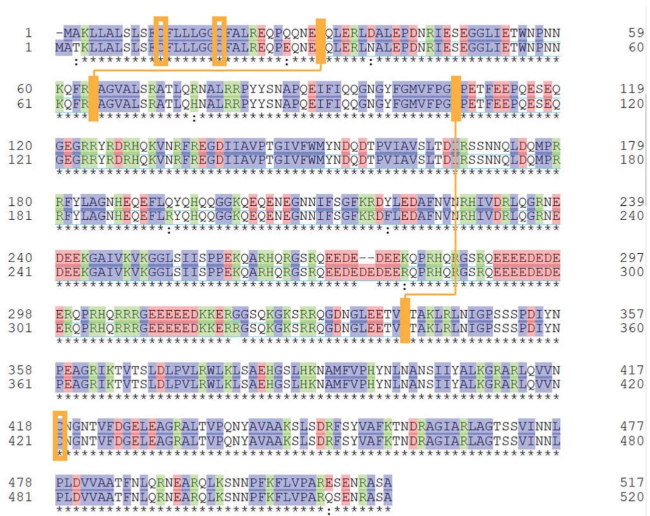

FIG. 10. Direct comparison of two representative primary structures (top and bottom row) from pea legumins with 517 and 520 amino acids by alignment. Blue: hydrophobic AS, red: negatively charged AS and green positively charged amino acids. Legumins are internally cross-linked with disulfide bridges (orange rectangles), but have free cysteine at the ends (open orange rectangles). Primary structures taken from Uniprot, top row: P02857 LEGA-PEA, bottom row: P15838, LEGA2-PEA.

The legumin fraction of the pea protein shows from its primary structure reasonable good properties for network formation. Despite two wasted loops in the native structure, the chains are relative long and provide cysteine at the end of the chains for cross-linking under heating. The albumin fraction of pea protein isolates contains two types, albumin-2 with 231 amino acids and albumin-1 with 130. The latter is highly self-cross-linked, and despite three free cysteine at one end, it appears to short and inflexible to contribute significantly to the network formation. Albumin-2 contains three free cysteines and might serves as a short cross-linker. The functional properties of the pea protein fraction are shown in Fig. 11. Given the free cysteines, albumin-2 can serve as an intermediate linker to support tighter cross-linking to enhance the mouthfeel during oral processing.

Given the primary structure, albumin-2 also acts as a highly effective emulsifier. It shows a pronounced block-like structure in the hydrophilic-hydrophobic amino acids sequences. Thus, the protein helps to bind the oil into the matrix, and cross-links may fix the molecule and, thus, the emulsifiers via the free cysteines. Moreover, the three free cysteines are able to cross-link with the matrix. Thus, albumin-2 can fix the oil droplets in the matrix, but less strong compared to myosin (see Fig. 7). The network scaffold from the pea proteins is more tightly meshed compared to egg white, so a higher modulus and, thus, steeper increase in tensile strength is expected in Fig. 4. However, a surrogate sausage made of pea protein only would

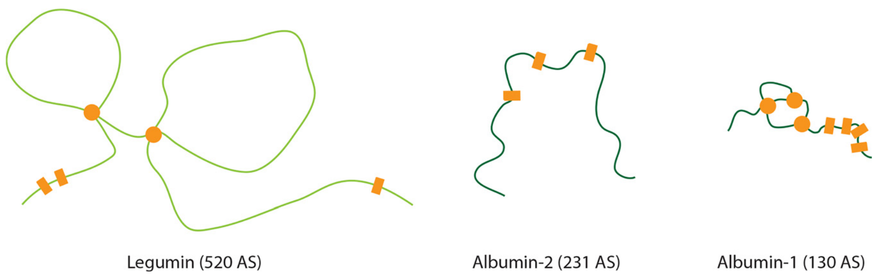

FIG. 11. Legumin and albumins of pea proteins contribute to different cross-linking properties. The circles are closed disulfide bridges, the rectangles represent –SH groups, which can be further cross-linked.

have too low breaking elongation, leaving the sausage with a more brittle mouthfeel.

To support emulsification, structure formation, and a better texture, potato protein is added to the vegan matrix, which contains two major groups, which are of similar length, 374 and 376 amino acids. Patatins show also an amphiphilic block structure that underlines its surface activity. Their lengths are comparable to ovalbumin and contain much less amino acids as legumins. Both patatin fractions are very similar in their primary sequence, but with one important difference. One type contains two free cysteines and the other only one, as can be seen in Fig. 12.

Apart from excellent emulsification properties, patatin contributes to the network formation only marginally. Nevertheless the short pea albumins-2 contains three free cysteines, which are able to form cross-links. Hence, it serves as a perfect linker and stabilizes the network structure. Its main strength lies in its high surface activity as can be read off its primary sequence. Moreover, patatin is not internally cross-linked and can in a denatured form develop strong emulsifying properties without strongly competing with the cross-linking process of the legumin.

The main network is spanned up by the pea proteins, the very long legumin, and shorter albumin-2. It appears, thus, as a bimodel protein network.[64] The shorter chains induce higher moduli during elongation and less stretchable networks. It is expected that the tensile modulus of the vegan network is larger compared to the vegetarian version, but still lower than for the meat sausages. Additionally, the smaller and shorter albumins allow for the smaller ultimate strains as the vegetarian version. The results of the temperature ramp experiments, see Fig. 3, support this model, too. The bi-modality of the legumin-albumin network indicates the more tightly meshed networks, compared to the vegetarian, egg-white network. The mechanical properties of these systems are less dependent on temperature than entropy-dominated, wide-meshed networks.

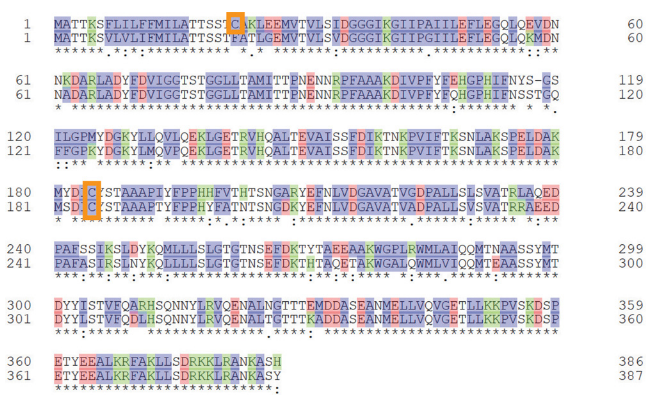

FIG. 12. Typical representatives of patatin are very similar in their primary structure, are excellent emulsifiers, and differ in their free cysteine (orange rectangles).

Carrageenan form soft gels by the formation of helix structures, reinforced by LBG and KG. As for the vegetarian version, discussed in Sec. IV C, the gels are unlikely to interpenetrate with the protein gels for thermodynamic reasons, such as incompatibility and strong repulsion.[65] During mixing, proteins and hydrocolloids are mixed, the oil droplets are emulsified mainly by the highly mobile, non-self cross-linked patatins. As shown in Fig. 12, the proteins contain unbound cysteine, which is able to cross-link occasionally with albumins and legumins from the pea protein fraction. This is an advantage of the vegan version. In the vegetarian, the hen egg albumin acts simultaneously as cross-linker and emulsifier; hence, a strong competition between these different requirements determines the resulting structure.

As in all other versions, the heating induces cross-linking between the unbound cysteines under forming disulfide bridges of the proteins, hydrocolloid chains will become expelled partly from the protein network. Carrageenan, LBG, and KG build up slowly separated gels distributed in the sausages during cooling after the scalding (cooking process) and contributes strongly to the water holding capacity and the juicy mouthfeel of the sausages.

Therefore, highly elastic networks, which are cohesive over larger length scales, can only form in this vegan scalded sausage with the help of the carrageenan. Although the elastic modulus in the stretching curves is higher for the vegetarian version, the cohesion remains looser than in the meat sausage and is expressed, for example, via the significantly low loss modulus in the rheological experiments shown in Fig. 1, as well as in the lower values for tan δ shown in Figs. 1 and 2.

E. Structural differences and friction

The remaining question is, how do these different structures influence the friction in the mouth? Indeed, the tribological processes are relevant to understand the physical part of sensory properties and mouthfeel in a better way.[66,67]

In the experiments starting with intact slices, it is noticeable that the friction coefficient of the meat sausage lies below the vegan and vegetarian versions, over the entire range of applied sliding speeds. This is clearly attributable to the free fat fraction, which is less incorporated in the meat sausage (see Fig. 7). Also, the fat content of the meat version is likely to be higher. In particular, the coefficient of friction decreases for the meat sausage at low velocities, while friction increases for the alternatives. This is also likely to be a consequence of the free fat, which is also present on the surface of the meat sausage. The uniform kink in the friction coefficient (at about 600 μm per second) in the slices is a consequence of the abrasion process starting under increasing speed. At sliding speeds around 10 000 μm per second, the friction of the vegetarian version decreases. There the egg white network is likely to be largely torn, so that the emulsifying effect is no longer complete. Oil is released and acts as a lubricant. In the vegan surrogate, the oil seems to remain strongly emulsified, which can be attributed to the patatin from the potato and the albumins from the pea, as earlier noted.

Vegetarian versions of cooked sausages make use of egg whites. However, egg white consists of far fewer proteins than meat. In addition, egg white proteins are also largely globular in the structure, whereas meat has globular sarcoplasmic proteins in the meat juice as well as the fibrous muscle proteins. Therefore, the consistency of hen egg white-based cooked sausages needs to be supported with biopolymers, such as xanthan gum (stiff, negatively electrically charged), locust bean gum (flexible, polar), and tara gum (flexible, polar), which is similar to locust bean gum, via molecular synergies.[25] The combination of the latter two with XG provides a network after scalding. The oligofructose inulin (compare Table I) produces a mouthfeel reminiscent of fat. The physical behavior of inulin and a comparison to fats have been already discussed earlier.[27,28]

For the sausage purees, the friction behavior is very different, compared to the slices. At sliding velocities up to about 5000 μm/s, the coefficient of friction of the meat sausage is, as expected, below the surrogates. The reason is again the free, not completely emulsified fat in cold meat sausages (see Fig. 7). For the vegan variant, the coefficient of friction decreases steadily (almost linearly). This is in line with the remarks with the already indicated high water binding of the hydrocolloids and the poorer cohesion of the network. Moreover, the carrageenan-hydrocolloid gels are largely separated from the protein networks for thermodynamic reasons,[65] or only weakly entangled, as the many hydrophobic blocks of the proteins literally repel the hydrophilic monomers of the hydrocolloids. The particles formed during pureeing disintegrate and separate further under the increasing sliding velocity. Oil becomes released, displacing the water, as well as the water-soluble hydrocolloids; the friction becomes lower. In the vegetarian egg white variant, the particles formed during pureeing are even firmer, as egg gels are characterized by high stability, the oil remains incorporated between pieces of ovalbumin gels. The XG-LBG networks oppose higher resistance to the sliding. Therefore, the vegetarian purees show the highest coefficient of friction of all three broths. Furthermore, the friction still increases slightly at very low sliding velocities and remains practically constant until further disintegration and forced water release from the hydrocolloid gels occur at very high sliding velocities. It is also interesting that the comparatively high friction in this product cannot be lowered by the vegetable fat substitute inulin.[21,68] Apparently, the (separated) particles of the egg white and hydrocolloid networks were the main contributors to the coefficient of friction.

V. CONCLUSIONS AND OUTLOOK

On macroscopic scales, it seems to be simple to construct and produce plant based meat analogs which resemble original meat products. Their optical impressions, their smell, and even the taste can be trimmed close to the originals. However, sensory deficits for most commercial products show up during oral processing. Shortcomings show up in the mouthfeel, different flavor release, deficiencies in the dynamics and sufficient cohesive bolus formation before swallowing. Considerations on molecular scales help to understand these shortcoming and show ways to improve the sensory properties. Apart from positive reports about taste, texture and aroma release by consumers, the physical properties of the materials are of interest, as they all contribute strongly to texture, fracture behavior, and bolus formation.

Consequently, plant based surrogates require more multi-scale physical understanding and input. It is not enough to look at the macroscopic properties. The interaction of proteins, defined solely by their properties and original function, must be taken into account. Only a closer look at the molecular scales reveals the strengths and shortcomings of these products. Although the vegan variant works well at first glance and first bite, it suffers, for example, from the still weak network formation of the pea and potato proteins, compared to meat. For an improved network formation, for example, an admixture of gluten (especially of glutenin) can to be considered. Gluten is known to provide doughs their perfect flow properties, and perfectly tight networks after heating.[65] A systematic data screening of proteins, especially their primary structure, provides suggestions and predictions for their functional properties. This shows, for example, the storage proteins of sunflower seeds, whose globulins could show a better network formation.

However, the precise network formation properties of plant protein isolates depend strongly on the extraction process of the proteins.[69] The extraction properties require strong changes in the pH values, which open native disulfide bonds and increase their functionality. By contrast, it remains unclear to what extension the subsequent spray drying process changes again the molecular properties.

The physical properties of the used fats and oils contribute to the texture and mouthfeel, too. Pork fat contains at room temperature a significant solid fat fraction, i.e., parts of the fat is crystallized, which affects the rheological properties of emulsion,[70] hence, the oral processing properties. Most used vegetable oils are and remain liquid during technological and oral processing. Pork fat melts at mouth temperatures, yielding an extra perception, which is missing in vegetarian and vegan surrogates. Improvements along these issues can be achieved by using various types of oleogels.

These ideas are supported by the observation shown in Fig. 3. The clear softening for meat sausages can be drawn back to the fat melting. However, the temperature ramp experiments offer also insight in the interplay between the separated protein and hydrocolloid networks postulated in Figs. 9 and 13. For perfect cross-linked, entropy dominated rubber networks, it is well known that the moduli increase with the increasing temperature, a clear sign for entropy elasticity.[71,72] Such a behavior cannot be expected, since the protein and hydrocolloid networks do not interact with each other and keep separated for kinetic reasons, as explained above. The stronger softening of the moduli of the vegetarian version, compared to the vegan system, supports the idea that the hydrocolloid network prevents the ovalbumin to cross-link fully. In the vegan sausage, a stronger cross-linking is likely due to the presence of the albumin-2. The hydrocolloid “islands” consisting of carrageenan, konjac gum, and locust bean gum are likely to be smaller and show lesser influence on the temperature dependent elasticity.

Despite all progress and increasing markets,[73] it must be clearly stated: no substitute sausage, no surrogate has a naturally occurring food matrix with all the naturally occurring macro- and micro-nutrients

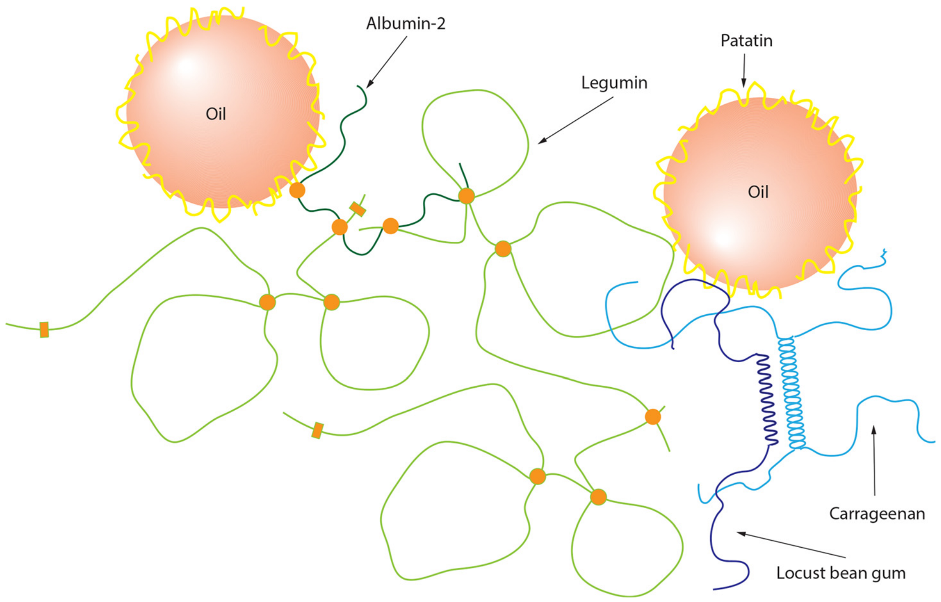

FIG. 13. An oversimplified sketch of the most important elements in the vegan version after scalding and cooling. The main network is formed by legumins (bright green), and albumin-2 (dark green) acts as short cross-linker, which is able to form cross-links with patatins (yellow). Carrageenan and LBG gums form separated small networks. Konjac gum shows similar properties as LBG and is not drawn explicitly.

contained therein that are essential for nutrition and enjoyment. Every surrogate must be upgraded and trimmed to this end in the future, even if the taste, aroma, and texture are already perfectly matched.

However, the approaches and methods of the ideas used in this paper can also help to show how biophysical concepts can contribute to improve structure and mouthfeel of plant based foods in future.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

M.G. contributed to rheology, tribology, writing, and editing. M.B. helped with rheology, tensile experiments, writing, and editing. T.A.V. involved in project idea, interpretation, model development, supervision, writing, and manuscript review.

DATA AVAILABILITY

The data that support the findings of this study are available within the article.

APPENDIX: NUTRITIONAL CONTENT AND VALUES FOR THE SAUSAGES

For convenience, the concentration of fat, proteins, carbohydrates, starch, fibers, and water are summarized in Table III.

TABLE III. Ingredients according to the nutritional label provided by manufacturer.

Ingredients	Meat (%)	Vegetarian (%)	Vegan (%)
Fat	18.0	13.0	12.0
Carbohydrates	3.0	3.9	3.7
Starch	1.7	2.4	...
Proteins[a]	11.0	7.2	2.2
Fibers	...	2.9	6.5
Salt	2.0	2.2	2.3
Water (approx.)	64.3	70.8	73.3

[a]Individual composition not provided by the producers.

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