



# Advances and challenges in soft tribology with applications to foods

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The last few decades have witnessed exciting progress in the understanding of soft material mechanics. Many of these advances have been inspired by, and have broad ramifications in the field of food science. One particular aim of food science is to get a better understanding of the physico-chemical mechanisms that are relevant in sensory perception and oral processing. It is recognized that not only rheological properties but also frictional properties are relevant in these processes. The frictional phenomena relevant for sensory perception can be understood by means of tribological measurements. The foods assessed are typically soft, hydrated and heterogeneous; measuring and understanding frictional properties of such materials is a challenge. Yet, also in the field of soft solid tribology, significant steps forward have been made, which now make it possible to do well controlled studies of even realistic food tribology scenarios. In this brief review, we provide a summary of recently developed experimental methods. We discuss challenges including the system dependence of a frictional measurement, and opportunities, such as mimicking in-mouth conditions by including human saliva and using tribo-pairs with similar properties to the oral surfaces. These advances lead to progress on the path towards a complete understanding of oral processing and sensory perception.

## Addresses

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## Introduction

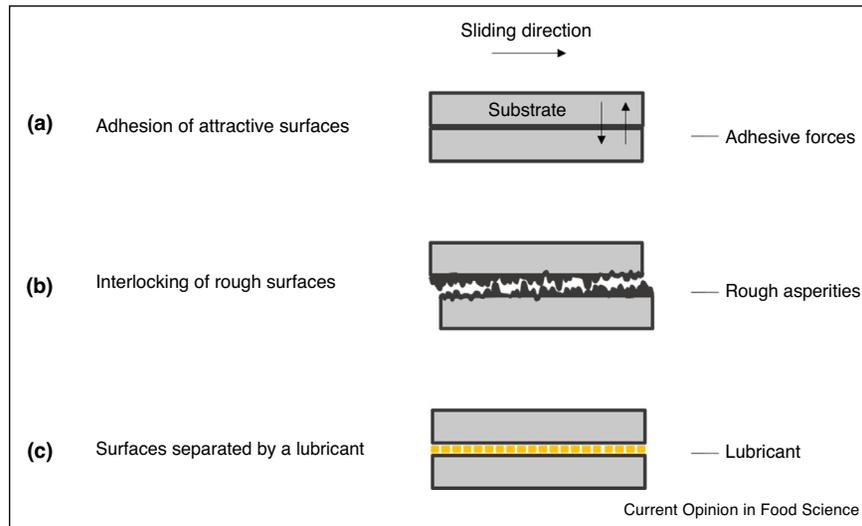
Friction is a well-known, yet rather complex physical process involving adhesion, fluid dynamics effects, surface deformation and wear of interacting surfaces

(Figure 1). The sliding force ( $F_f$ ) of two interfaces are characterized to be linearly proportional to the normal force ( $F_n$ ) resulting in the dimensionless friction coefficient  $\mu = F_f/F_n$ , a property of the characteristics of the interfaces. This straightforward perspective would only require us to characterize  $\mu$  for all material combinations to arrive at a complete understanding of friction. Unfortunately, understanding friction between touching surfaces, especially soft ones, requires more effort. Recently, advances have been made in soft solid tribology; one relevant finding is the often important role of molecular mechanisms in frictional dissipation. The field of tribology is now at a point where it can start to provide insights into more complex (food) systems that are often heterogeneous of nature. In this review, we aim to highlight the tribological methods that are currently used in food research. In particular, we will focus on the synthetic materials used to mimic biological systems and how to compare results across tribological devices. We end the review with an outlook on the exciting future of tribology research in general and food tribology in particular.

## Tribology of soft substrates

Traditional tribology involves hard surfaces and lubricants that display Newtonian behavior. This often translates into rolling ball bearings [1], oil lubricated systems [2] and gears [3]. Classic engineering materials have applications in machine engines or other (rotating) instruments. Recent developments in materials science have introduced new soft, deformable materials into the domain of engineering. These compliant, soft, lubricated, porous and rough materials introduce critical questions regarding the applicability of classic tribology know-how and bring the opportunity to uncover new insights into frictional laws. Investigating the frictional properties of elastomers [4,5], hydrogels [6,7] and natural cartilage [8,9] is of importance for the development of medical equipment, such as contact lenses, pacemakers or artificial organs to cure or treat diseases. As these materials are often designed to resemble soft tissue, they have also gained interest of food scientists, who use these materials to mimic the mostly soft and wet nature of the oral cavity. Soft surfaces, especially those belonging to the category of hydrogels, are often characterized by low frictional properties with friction coefficients, as low as  $\sim 10^{-3}$  [10–12]. These low friction coefficients have been attributed to loose hydrophilic polymer chains on the hydrogel surface, which can create a soft, hydrated polymer layer by entrapping water [10,13,14]. Friction coefficients in hydrogels can be increased by many system parameters,

Figure 1



Schematic representation of friction mechanisms between two surfaces. **(a)** Adhesion of surfaces increases the force needed to separate surfaces. **(b)** Interlocking of surface asperities immobilize interacting surfaces. **(c)** A lubricating film can be formed when space is created between surfaces. The schematics presented can represent a wide range of length scales, from nanometers up to centimetres.

such as the surface roughness [6], the material stiffness [11], and adhesive properties [15]. Such adhesive properties are a result of attractive forces between the surfaces. In addition, the interlocking of surface asperities and the ability of the lubricant to separate the surfaces in contact will also play a large role (Figure 1). Separation can also arise from repulsive forces between the surfaces. These phenomena are often present simultaneously, which makes frictional properties of soft systems complex to measure, analyze, compare, interpret, and understand. Unfortunately, this often also leads to poor reproducibility [16–18].

### Measuring friction

The many different materials encountered in soft solid tribology have inspired the development of a range of tribometers and tribological tools. In food science we often encounter tribometers produced by manufacturers such as Anton Paar, PCS instruments and Bruker. It is not uncommon for tribologists to design their own tribometers to answer specific experimental questions. Such customization requires the correct technical knowledge and specific equipment to assemble such an instrument. An overview of various existing tribometers with recent advances in the food and soft matter fields are displayed in Table 1. In the table, we include references to recent studies that used the respective devices.

As frictional measurements are system-dependent, all techniques described have their limitations. For example, many of these devices are limited to either reciprocating or rotating movements, along with constraints in applied

normal force and velocities. When aiming to measure realistic conditions for oral processing, these movements are not very representative for in-mouth conditions. However, the current techniques also provide new possibilities. For example, most tribometers offer the possibility of replacing the interacting surfaces with custom-made materials and can measure at speeds up to 3000 mm/s; low normal forces of less than 0.5 N can be applied. Specific tribometers can be programmed to move the probe in varying shapes, bringing food tribologists closer to simulating in mouth conditions [29]. Tribometers with such specific possibilities can be relatively expensive compared to other commercially available set-ups. Affordable tribo-tools are designed to be mounted on rheometers or texture analyzers that are often already present in food laboratories. Mimicking more specific conditions can be achieved by designing and building tribometers in-house using equipment able to apply a fixed load or force. This however requires specific knowledge and skills, and extracting reliable data may also be more challenging. The large variety of commercial and custom-built tribometers makes it difficult to compare obtained frictional data across different studies. In the next sections, we will discuss common strategies to measure and assess frictional data.

### Analyzing frictional data

Frictional measurement data are commonly assessed by plotting the friction coefficient as a function of the entrainment speed. Such a curve was first constructed by Stribeck in the early 1900's to describe the friction of

Table 1

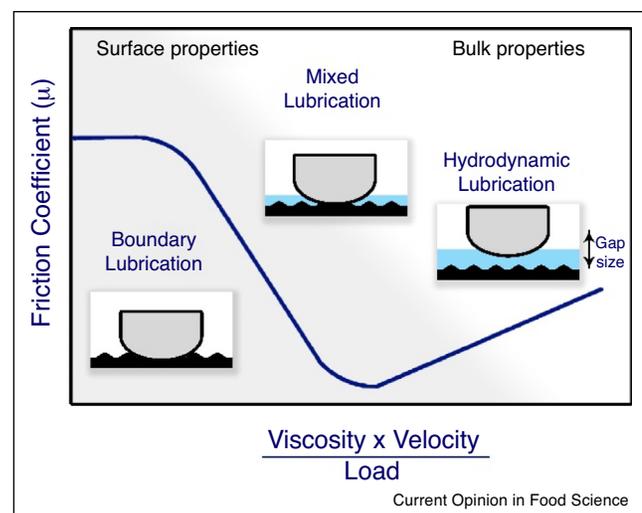
**Tribometers commonly found in soft matter/food literature with corresponding product features, recent findings and examples of systems that have been measured using specific tribometers**

Tribometer	Description	Recent developments	Measured foods/systems
Anton Paar MCR tribometer	Ball-on-3-pins rotational tribometer. Often used with a glass probe and elastomer pins in food literature. Different surfaces can however be used.	A Stribeck curve with a static and kinetic regime was obtained by measuring at extremely low speeds (Figure 3) [19].	(Double) emulsions [18] Chocolate spread, cheese sauce [19] Milk, yoghurt [20] Semi-solid model foods [21] Greases [22] Thermoplastic polymers [23]
PCS instruments mini traction machine	Ball-on-disc rotational tribometer with various surfaces available with rotational and rolling movements.	Specific laser textured surfaces that influence transition points between different frictional regimes were designed [24].	Emulsions [25] Milk, yoghurt, cream cheese [26] Chocolate [27] Oils with friction modifying additives [28]
Bruker UMT tribolab	Ball-on-plate tribometer with multiple (modular) drives available. Probes and substrates can be customized. Adjustable stroke lengths and sliding trajectories such as an ellipse or a 'figure 8' path.	Measurements at different sliding shapes. Friction coefficients are affected by selected sliding trajectory [29].	Whey protein model foods [29] Milk, rice starch dispersions [30] TiO <sub>2</sub> nanoparticles with polymer brushes [31] Hydrolyzed polyvinyl alcohol [32]
Customized texture analyzer	Three-ball-disc connected to a texture analyzer load cell with a water bath for temperature control. Surfaces are easily varied.	Using the tongue as a mold to create rough 'oral' surfaces [33].	Emulsions [34] Wines [35] Yoghurt [36]
Custom Pin-on-disk tribometers	Hemispherical hydrogel probes sliding against hydrogel disks.  Example 1: Rotating disk with stationary hydrogel probe (Sawyer group, Florida).  Example 2: Rheometer-driven hydrogel probe sliding against hydrogel disk (Dijksman group, Wageningen).	Custom-built devices on rheometer or other force sensing/strain inducing equipment. Friction coefficient were shown to correlate with Hertzian contact theories for gemini interfaces [37]. Sliding properties of hydrogels correlate with flow behavior of hydrogel particles [38].	Polyacrylamide hydrogels [37,12]  Gelatin and polyacrylamide hydrogels. [38]

metal surfaces lubricated by a layer of fluid, and is known as a Stribeck curve (Figure 2).

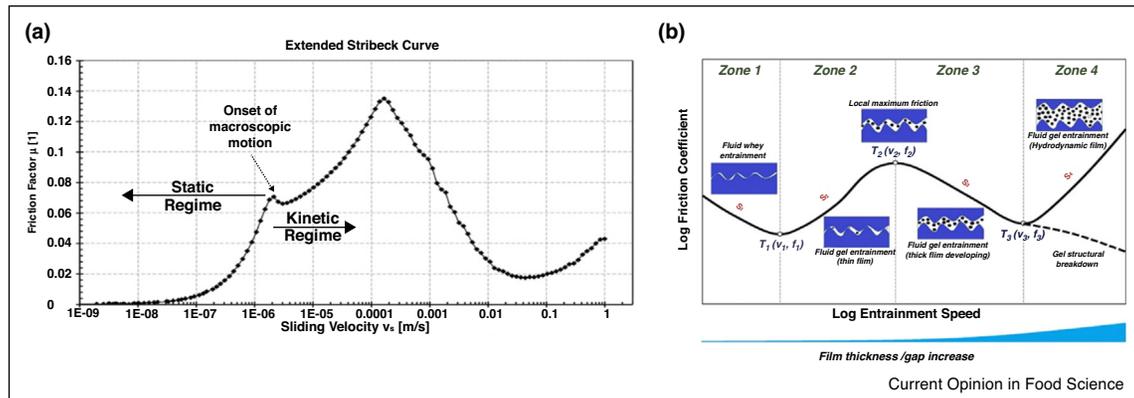
A Stribeck curve originally describes three regimes: 'boundary' regime, 'mixed' regime and 'hydrodynamic' regime [19,39–41]. The Stribeck curve has been discussed in detail in many (review) articles [19,42–46]. Typical Stribeck curves (Figure 2) have been obtained for sugar solutions and biopolymer mixtures [47], and for emulsions with differences in oil content [18,30]. However, due to the large variety of microstructures, many food materials can deviate from standard Stribeck behavior, as seen for measurements with yogurts and custards [48]. More extreme examples of non-standard Stribeck curves are given by Pondicherry *et al.* [19], and Nguyen *et al.* [49] for sunflower oil (Figure 3a) and model yoghurt samples (Figure 3b), respectively. The deviation from the standard Stribeck curve is due to different events during the measurements. The extended Stribeck curve presented in Figure 3a includes an additional static regime at velocities below  $10^{-6}$  m/s, next to the kinetic regime at higher velocities. The transition between these two

Figure 2



Schematic representation of the Stribeck curve. The boundary and mixed regime are dominated by surface properties while the hydrodynamic lubrication depends on the bulk properties of the lubricant.

Figure 3



Alternative Stribeck curves presented in literature. Curves are based on measurements with (a) sunflower oil [19] and (b) model yoghurt samples [49]. Figures have been reproduced with permission.

regimes is characterized by a peak, which is caused by the initiation of macroscopic sliding of the probe against the substrate. Another extension to the classic Stribeck curve is represented in Figure 3b, showing four frictional zones [49]. This model was obtained using a ring-on-plate tribometer with yoghurt samples on a hydrophobic surface (Transpore Surgical Tape). The four zones arise due to the heterogeneous nature of the yoghurt sample, as yogurt contains fat as the dispersed phase. In the first zone, only the fluid is considered to be able to enter the surfaces, and the fat droplets remain in the bulk phase. As the entrainment speed and the gap size increase, the fat droplets are also able to enter the gap and are now also present between the sliding surfaces. This initially increases the friction coefficient. As the speed increases, friction decreases again, and ultimately a hydrodynamic layer is formed, similar to the standard Stribeck curve. These alternative Stribeck curves show again that the measurements are very strongly system-dependent. Another interesting deviation from classic Stribeck behavior is shown with water-based lubricants that do not differ in viscosity but do show very different frictional behavior in the contact regime [29]. Such observations point towards the relevance of also molecular mechanisms in soft solid tribology and are of obvious relevance for oral processing, in which saliva plays an important role.

### Mimicking in-mouth mechanisms

The field of soft solid tribology is propelled by the desire to understand friction in food physics via other routes while inspiring the design of new tribology instruments. Food friction studies aim to understand sensory perception and oral lubrication during the different steps of oral processing. The question is then how to connect sensory perception concepts to frictional measurements, which are system-dependent. A possible route to make the link between sensory perception and tribology is to perform tribological experiments on mouth-mimicking substrates.

The artificial surfaces used should in that case ideally mimic the properties of the soft tongue, and the relatively hard palate. To perform such mouth-mimicking tribological experiments, a detailed knowledge of the role of substrate roughness, hardness and other surface characteristics is required.

Several studies have investigated the mechanical properties of the human tongue and values between 2.5 kPa and 150 kPa have been reported [50–54]. Movements of several groups of muscles in the tongue can give variations in stiffness of the organ. The tongue in dry form is hydrophobic. In the presence of saliva, the tongue exhibits more hydrophilic behavior as caused by adherence of amphiphilic proteins present in saliva [55–57]. Furthermore, the surface topology of the tongue is rather complex, as the surface of the tongue is decorated with papillae that have been found to reach up to 0.5 mm in height for fungiform (taste) papillae. Papilla diameters range from 0.5 to 1 mm depending on the age, size, sex of the person and the location on the tongue [58]. Papilla densities also vary between subjects and this has even been found to influence sensory perception [59,60].

To mimic the soft nature of the tongue, relatively soft deformable materials are used in frictional studies. This makes the surfaces subject to deformations which may influence the frictional behavior. In lubricated deformable polyacrylamide hydrogels this has been found to cause a decrease in friction coefficient with increasing normal forces [37]. The ratio between the stiffness of two interacting surfaces such as the relatively hard palate and the soft tongue should therefore also be considered. When pairing hard surfaces with softer surfaces, deformation may occur, leading to changes in the surface structure. For spherical contacts, Hertzian type deformation can be expected depending on the Young's moduli of the surfaces in contact [61,62].

Besides the relatively soft nature of the oral surfaces, also different normal forces and different sliding speeds and sliding directions are applied during oral processing. These pressures and specific movements should also be considered in tribological measurements when mimicking oral situations. As shown by the Stribeck curve (Figure 2), frictional behavior is strongly dependent on sliding velocity. Sliding speeds during oral lubrication will strongly depend on the food consumed and on the consumer, but have been estimated to be between 5–200 mm/s [29,63,64]. Most tribological tests are performed at a sliding speed of 0.01–2000 mm/s [65], covering indeed the estimated speeds during in-mouth friction. Many tribotests use a (hemi-)sphere against a flat substrate (Table 1) in either a rotating or sliding motion. It has recently been found, that the sliding shape or the trajectory of motion of the probe can change the lubrication behavior. Using model foods containing whey protein and saliva, it was shown that linear movements gave higher friction coefficients than elliptical movements at constant sliding velocity [29]. Therefore, also the sliding trajectory appears to be relevant when real mouth-mimicking situations are desired.

The same study also showed that saliva is an important factor to take into account. When saliva was mixed with the model food containing whey proteins, a large drop in friction coefficient was found [29]. Saliva generally makes masticating food easier by making it softer and more slippery while preparing the bolus to be swallowed [65,66]. Like many biological fluids, saliva is a complex mixture of ingredients, including proteins, and has excellent lubricant properties on its own. Friction coefficients as low as 0.029 have been measured for saliva [67]. The specific composition of saliva is person dependent [66], making it challenging to compare results of various studies. To simulate the environment of the mouth for tribological experiments, one can however take care to make a highly accurate representation of the mechanical environment, or at least learn to extract the individual factors relevant for sensory perception, which may work down to the molecular level [68,69]. Frictional studies on soft surfaces thus pose an interesting challenge to (food) tribologists. Most importantly, the tribological equipment should be capable of handling custom materials, surfaces and operate in both complete and partially wet conditions.

### Synthetic surfaces

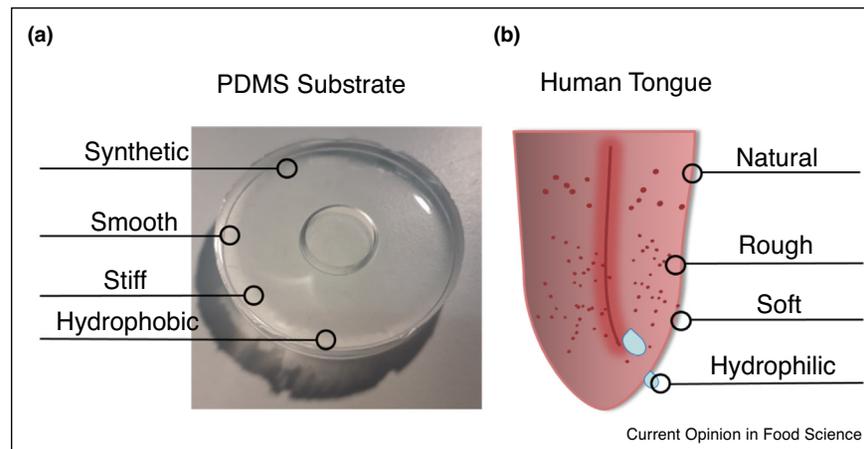
To mimic in-mouth conditions, it is necessary to carefully choose the materials used. As previously mentioned, simply changing the interacting surfaces can give large differences in friction coefficients [38,45,70,71], and therefore the material properties should be close to the properties of the oral surfaces. As mentioned in Table 1, different materials serve as potential analogues for the human tongue and palate (e.g. glass probe and rubber substrates). Interesting examples of mouth mimicking

conditions are frictional experiments done using a pig's tongue [56,72,73]. The resemblance between a pig's tongue and a human tongue is justified by the fact that the hydrophobicity of the pig's tongue is very similar to that of a human tongue, and the roughness of the tongues are comparable. Using animal organs post mortem is however discouraged due to the rapid post mortem biological changes occurring in the tissue [43]. Such changes will alter the surface structure, and thereby the frictional properties. The influence of the surface structure was already shown in a study using synthetic elastomeric surfaces covered with hemispherical asperities [74]. Under dry conditions, the friction coefficient decreased with increasing contact area. Not only in dry conditions, but also in wet conditions, the surface structure and changes in surface structure is of great relevance. Upon insertion of a lubricant between the surfaces, an increasing asperity height for example hinders fluid film formation and increases friction.

Most experimental tribology work uses synthetic materials as they allow for control of the material properties and are less affected by natural changes present in biological samples. The most prevailing synthetic material used to mimic the tongue is the elastomer polydimethylsiloxane (PDMS) (Figure 4).

PDMS is a type of organosilicon elastomer that can be found in various products and applications, ranging from contact lenses to food additives. Several manufacturers of (food) tribometers supply or recommend the usage of this elastomer (see overview in Table 1). The advantage over other tribomaterials are found in the fact that PDMS is easy to synthesize or shape via molds; it is also affordable costing around \$200 per kg. The Young's modulus of PDMS can be adapted over a reasonable range by varying the polymer concentrations or cross-linking densities during mixing [45,75]. Conveniently, PDMS does not swell in water and remains stable over a long period of time at a wide range of temperatures. Additionally, the material is inert and non-toxic [76]. Although PDMS stiffness can be easily adjusted by varying the ratio of the cross-linkers, it is important to realize that the stiffness will influence the frictional measurement [45]. In addition, PDMS has a tendency to leak polymers at the surface, which has been shown to increase the adhesive properties [77]. The Young's modulus of PDMS used in tribological research is often around 3 MPa [29,71], much higher than that of the human tongue, which is typically reported to be between 2 and 150 kPa [53,54]. PDMS is hydrophobic by nature but the surface can be hydrophilized using different techniques [78,79]. The relative smoothness of the PDMS surface can be altered by casting PDMS in a roughened mold. Using PDMS provides various opportunities to change the surface and mechanical properties with respect to softness, hydrophobicity and roughness.

Figure 4



Comparison between an (a) untreated PDMS surface and (b) the human tongue. Note that PDMS characteristics can be altered with specific treatments.

The intrinsic differences between synthetic, hydrophobic and smooth PDMS and the hydrophilic, rough human tongue may raise the question whether PDMS is indeed the holy grail when it comes to mouth-mimicking model systems. Pragmatically, using PDMS is certainly already better than using a commonly used very hard surface like steel. Moreover, by now PDMS has become a de facto reference system, which aids comparisons of studies, even if it does not mimic the tongue perfectly. Nevertheless, there is still ample space to explore novel soft substrates for food tribology in general and sensory perception studies in particular. The investigation of tongue-mimicking surfaces could be extended towards the category of biomaterial (hydro)gels. The material properties are easily adapted by varying polymer concentrations and Young's moduli are often in the kPa range. The surface topography can be altered by using patterned molds. Similar to saliva-covered tongues, hydrogels are already hydrophilic in nature, and maybe therefore be more representative. To avoid swelling of physically cross-linked hydrogels, a chemical cross-linker can be added, which also increases the stiffness and stability over time [70].

### Outlook and recommended considerations

Despite decades of work on soft solid tribology, there is still no clear consensus on how to accurately measure friction on soft, rough, lubricated solids, and how to obtain the desired correlations with *in-vivo* experiments. The sensitivity of tribological measurements to material and measuring parameters make it difficult to directly link the measured frictional data to sensory attributes perceived by human subjects in a sensory study. This sensitivity may even cause tribometers to measure differences between samples that participants in a sensory panel do not perceive as different. Nonetheless, many researchers are making progress and have even succeeded in

finding agreements between frictional measurements and sensory perception [20,26,36,57,80], justifying the growth in interest in soft solid tribology in food science.

There are many aspects that should be taken into consideration to make meaningful mouth-mimicking experiments. Important requirements of an oral-tribometer are affordability, versatility and reproducibility for complex food samples. When using tribometers with soft, synthetic surfaces to mimic biological processes, parameters such as surface roughness, stiffness and hydrophobicity need to be taken into account. To compare the lubrication behavior of food or different materials across different studies, it is important to realize that the results are system-dependent, and not only food-dependent. To increase resemblance to oral surfaces, the stiffness and roughness of the material should ideally be in the same range of the human tongue and palate. Molecular level surface and lubricant characterization is necessary to begin to understand non-Stribeck behavior. Tribological research in the field of food science will also greatly benefit from methods that allow various sliding movements, as food is not moved into a single direction in the mouth; interestingly, such anisotropic friction is also receiving interest in other fields [81]. Having better visual access to what happens in the boundary layer of the sliding surfaces will be essential to understand what happens during frictional sliding [82,83].

As the field of food tribology continues to expand, more realistic measuring systems are expected to rapidly surface. This opens the door towards a better understanding of tribology in relation to oral processing and sensory perception. Customization of tribological experiments will therefore be an important direction of future tribology work.

## Conflict of interest statement

Nothing declared.

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