REVIEW ARTICLE

A tribo-chemical view on astringency of plant-based food substances

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Abstract: Consumption of plant-based food products having high composition of polyphenols leads to the sensation of astringency. For sliding oral surfaces, friction is an essential property during the oral perception of roughness and dryness which are attributes associated with astringency. Different factors including the chemical composition of interacting layers, structure and operation of interfaces have an effect on the astringency development process. The manner of interactions occurring at oral interfaces suggest there is a system dependence of astringency and highlights the importance of adopting a tribosystems approach. Available measurement techniques have shown an existing relationship between salivary protein-polyphenol interaction and an astringent mouthfeel. Nevertheless, the tribo-chemistry involved in this multifaceted sensation remains largely unexplored in a comprehensive manner. In this review the underlying tribo-chemical processes useful in understanding the mechanism of astringency are highlighted and discussed considering current techniques employed to investigate astringency perception. Loss of lubrication on oral surfaces owing to the tribo-chemical interactions involving saliva and astringent plant proteins requires subsequent deformations of oral tissues which are significant enough to induce strains at mechanoreceptor locations, leading to the sensation of astringency. It is proposed that micro-scale contact modelling on the interaction of food particles/aggregates, boundary layers and oral surfaces shows potential in addressing the knowledge gap between tribo-chemical measurement techniques and panel tests, making it possible to attain a predictor for astringency.

Keywords: oral lubrication; astringency; tribo-chemistry; tribology

1 Introduction

As the European Union intensifies efforts at achieving a 55% minimum reduction in greenhouse gas emissions by 2030, food consumers are transitioning towards the consumption of plant based alternatives in place of animal based protein sources. Plant proteins have been investigated and introduced as alternative because they offer a higher protein content to CO_2 emissions ratio compared to animal proteins [1]. Several of these alternatives such as those based on pea or faba bean, however, contain polyphenols which when ingested, result in the sensation of oral astringency. Astringency describes an oral sensation involving dryness, shrinking and puckering of the oral epithelium resulting from exposure of the oral

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surfaces to 1) phenolic compounds, 2) organic and inorganic acids such as malic or hydrochloric acid, 3) dehydrating agents e.g. ethanol and acetone, 4) multivalent salts such as potassium ammonium sulfate, 5) proteins with a high isoelectric point, and 6) amino functionalized polymers carrying positive charges at physiological pH [2, 3]. At high intensities, this complex perception has been regarded as unpleasant to food consumers [4] which has resulted in the initiation of various studies aimed at understanding the oral perception of astringency.

The perception of astringency differs from person to person, given the differences in sensing thresholds amongst individuals and given the fact that its perceived intensity varies over time, taking up to 15 seconds to fully develop [5]. Attempts have been made to classify the oral perception of different food substances [6, 7], with some experiments specifically focused on the oral perception of astringency [8–12]. A clear coupling between plant-based protein food composition and the perception of astringency is however not available, indicating the need for a more thorough understanding of the controlling mechanisms of astringency.

There are available literature reviews highlighting various studies on perception in the mouth [13–17] and the mechanism of astringency at oral surfaces [18–22]. Several studies correlate astringency to a tactile sensation arising primarily as an effect of reduced salivary lubrication on oral surfaces [8, 21, 23–25], while others focus on the complexation of salivary mucins, precipitation of salivary proteins and reduction of saliva flow rate [9, 23, 26]. The combination of the two lines of research stresses the importance of tribology in understanding astringency, and more specifically in understanding the tribo-chemical aspects in relation to plant-based protein food substances. This review focuses on the tribo-chemical processes underlying the mechanism of astringency in plant-based protein foods. Experimental methods are reviewed in the light of assessing the effectiveness of the tribological interactions in support of the development of future plant based protein products. The tribo-chemistry involved in the astringency development process has received little attention and is yet to have been presented in a systematic way.

2 Analysis of the tribological system related to astringency

From current literature, including the excellent recent review on food oral tribology by Xu et al. [27], it shows that diverse factors affect the astringency sensation, including food composition and individual differences in saliva, tongue topography, and tongue motion pattern, hinting at a system dependency of the sensation of astringency, rather than at a food or oral property only. This observation is furthermore consistent with the described correlation of friction forces and astringency [8, 25], similar to the correlation of friction that is found with other sensory attributes of food such as creaminess [6, 28], fattiness [29], thickness, smoothness, and slipperiness [30]. These friction forces arise from the sliding interactions in the mechanical contact between the palate, the tongue and the plant-based food constituents in the presence of a lubricating layer based on saliva. The related tribological system [31] is schematically depicted in Fig. 1(a).

The oral palate, located at the roof of the mouth, acts as a barrier that separates the oral and nasal cavities. It is divided into two parts 1) the mobile posterior (fleshy) soft palate and 2) the stationary anterior (bony) hard palate (see Fig. 1). The bony structure of the hard palate is slightly rounded and lined by a layer of mucous membrane containing salivary glands. Also situated at the hard palate is a layer of ridges that aid with gripping food during chewing and eases the movement of food towards the rear of the mouth. Somatosensory innervation (i.e. neural network responsible for touch perception) of the hard palate is possible through Meissner corpuscles, glomerular endings and Merkel cells, giving it the ability to detect forces and vibrations, as well as the ability to distinguish between textures [32].

Also housed in the oral cavity is the tongue which is a soft tissue organ consisting of a complex mass of cross-striped muscles and serous glands, encased by a mucous membrane. The tongue is inherently hydrophobic, however the presence of a salivary layer is responsible for the hydrophilicity of the tongue surface [33, 34]. Figure 1(b) shows an exaggerated schematic of the anterior dorsal section of the tongue consisting of papillae which give the tongue a high
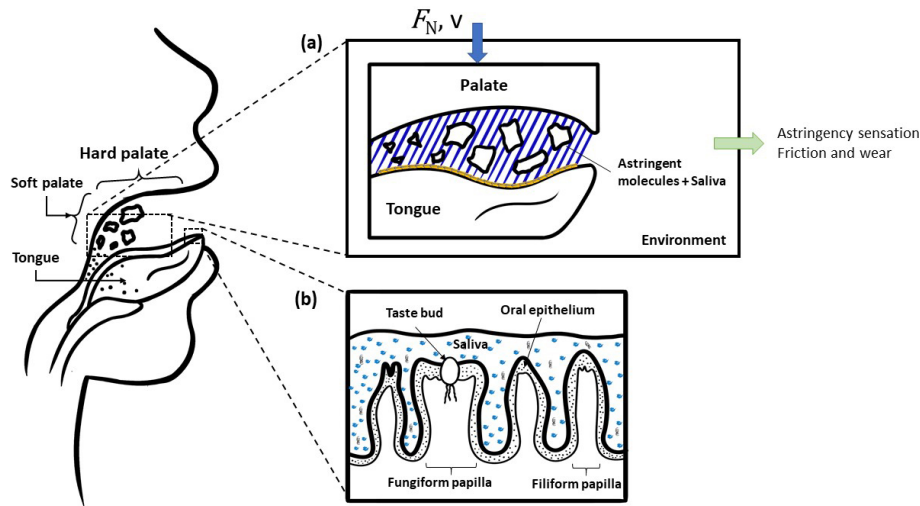
Fig. 1 Schematic representation of the human oral cavity showing (a) a tribosystem for the tongue–palate contact with sliding velocity v and normal load F_N and (b) the structural composition of the tongue surface.

surface roughness. The papillae are protuberances of the tongue epithelium and the lamina propria (connective tissue) which can be divided into four types; fungiform, foliate, circumvallate and filiform. The taste buds are located at all the papillae except the filiform and are sensitive to five different stimuli; sweet, sour, salty, bitter and umami [35, 36]. Despite the fact that the filiform papillae are the most abundant papillae in the mouth, they do not contain any taste buds. This is evident as mechanoreceptors embedded within and innervating the filiform papillae are not exposed to the tongue surface, implying a lack of chemical stimulation [22, 32]. Rather these receptors are sensitive to stresses due to tribomechanical loading of tongue tissues during contact with other oral components such as food, saliva, tooth enamel or palate [37]. The filiform papillae have a conical/cylindrical structure with an average height of 250 μm and a base width of 420–500 μm that extends to a hair-like structure at the papillae ends, whereas the fungiform papillae are dome shaped with a base width approximately twice that of the filiform papillae [38]. Measurements on the mechanical properties of the human tongue have reported a range of 2.5–150 kPa for the Young's modulus [39, 40]. The higher stiffness values may be as a result of the site dependence of measurements of the oral mucosa [41]. During in-vivo conditions, the tongue can move at a velocity between 5–200 mm/s and exerts a bulk compressive pressure of 2–70 kPa

on the palate [42, 43, 44]. Table 1 summarizes the values obtained from studies in the literature that have measured these parameters in the human oral cavity. Measurements of the typical motion patterns of the tongue at different locations and stages of mastication are also available in literature and serve as an important reference point for studies aiming to replicate tongue motion [45].

Current systems in oral lubrication studies use sliding speeds ranging from 1–100 mm/s and loads in the range of 0.34–5 N, to simulate the sliding contact between the tongue and palate [8, 26, 62–66]. The choice of tribo-pairs and testing conditions in these studies results in contact pressures that exceed the measured values presented in Table 1 [67]. This raises concerns about the relevance of the tribosystems used to investigate lubrication on oral surfaces.

Surfaces within the oral cavity are naturally covered by a thin film of saliva, which consists of approximately 99.5 wt% water, 0.3 wt% proteins and 0.2 wt% inorganic substances with an average pH of around 6.8 [19]. Salivary proteins and ions give the saliva

Table 1 Measured values for in-vivo conditions during oral assessment of food.

Parameter	Measured value	Ref.
Sliding velocity	5–200 mm/s	[44, 46]
Salivary film thickness	42–100 μm	[47–49]
Contact pressure	2–70 kPa	[42, 43, 50, 51]

enhanced physical properties. Specifically, mucins and glycosylated proline-rich protein content of saliva constitute the boundary film layer on epithelial surfaces and are responsible for the enhanced lubricating properties of saliva [22]. The main salivary proteins can be classified based on their structure and characteristics as follows: mucins, α-amylase, histatins, P–B peptide, proline-rich proteins (PRP's), cystatins, and statherin [52]. All these are present in human saliva at different concentrations. The non-homogeneity of salivary proteins has been linked to the resulting differences in perception of astringency among individuals which suggests that lubrication regimes have a role in astringency perception [44, 53]. This is due to the fact that variations in saliva composition may result in more or less binding of astringent food compounds to proteins in saliva and a consequent alteration in the quality of oral lubrication experienced by individuals.

Mucins present in saliva constitute about 20% of salivary proteins and are associated with lubrication, maintaining viscoelasticity of secretions, hydration and protection of the oral cavity [54]. Mucins are anionic glycosylated proteins with a pH in the range of 6.2–7.4, an isoelectric point between 2 and 3 and a high molecular weight ranging between 0.2 and 40 MDa [55–57]. The mucosa layer has a variable viscosity which is dependent on changes in environmental factors such as pH and ionic strength [58]. This variation in viscosity indicates that mucins are significant in determining the lubrication regimes during the oral sensation of astringency.

The family of PRP's have been broadly explored in relationship to the role of salivary proteins in astringency development. Salivary PRP's have a high binding affinity with tannins (another class of polyphenolic biomolecules with the characteristic ability to bind and precipitate salivary proteins), readily forming insoluble complexes that could be responsible for the roughness feeling on the tongue associated with astringency perception [59]. The extended structure of PRP's and their high content of proline residues provide a preferential site for the binding of multiple astringent molecules, such as tannins and catechins [60]. It is therefore expected that the structure of the precipitates formed will differ according to the astringent molecules bound. Studies found that precipitation of salivary PRP's is increased when there is an interaction with larger and more complex polyphenols such as the high molecular weight tannins containing freely rotating interflavan bonds and gallolyl groups [61]. These tannins have more binding sites available to interact with the proline residues and the ensuing Tannin-PRP's complexes are insoluble [53].

3 Polyphenol–Salivary proteins interactions

Plant-based protein food products contain small volumes of polyphenols which are natural organic compounds possessing strong antioxidant properties and are the most common astringent molecules present. Polyphenols have a chemical structure characterized by a benzene ring attached to a hydroxyl group (–OH) and can be classified as either non-flavonoids or flavonoids. The former consists subclasses of phenolic acids, lignans and stilbenes, and the latter consists of flavones, flavanols, flavan-3-ols, anthocyanins, flavanones, and isoflavones.

Phenolic acids such as caffeic acid (CA), chlorogenic acid (CGA) and gallic acid (GA) are commonly found in green tea leaves and wine. Sensory studies on these phenolic acids showed that there was a relationship between increasing concentrations and the perceived astringency [68]. Flavanols such as quercetin, catechin, and epicatechin exhibited a similar characteristic increase in perceived astringency at higher concentrations due to increased interactions with salivary proteins [69, 70].

The molecular basis for the interaction of salivary proteins and polyphenols is associated with the precipitation of proteins in saliva which may lead to formation of aggregates. Various studies have investigated the interaction of polyphenols with salivary proteins to be dependent on their concentrations, chemical structures, pH and molecular charge [71, 72]. The cross-linking of proteins and polyphenols are considered to occur through van der Waals interactions, hydrogen bonds, ionic bonds or covalent bonds. Van der Waals interactions facilitate hydrophobic interactions between the benzene rings of polyphenols and non-polar amino acid side chains whereas

hydrogen bonds facilitate polar interactions of hydroxyl groups of phenolic compounds and carbonyl with amino groups of proteins [73]. The dominant tannin-protein interactions are considered to be the hydrogen bonds and hydrophobic interactions [74]. Nevertheless, it is also possible that covalent bonds form between nucleophilic groups or proteins and quinone forms of phenolic compounds. Ionic bonds between cationic sites of proteins and phenolate anions are also possible. Salivary protein–polyphenol interactions via these bonds are considered to be occurring in three main processes: i) formation of the smaller protein-phenol aggregates; ii) self-association of the small aggregates via a cross-linking process to form complex aggregates; and iii) precipitation of the large complex aggregates [54].

It is clear that there are changes to the boundary film as a result of the tribo-chemical processes taking place during the interaction of polyphenols and salivary proteins on oral surfaces. However, what is still missing is a well-founded connection between the tribo-chemical changes at the boundary film and the physiological sensation of astringency. The primary trajectories involved in establishing this connection are based on the formation of protein-polyphenol aggregates, breakdown of the salivary film and exposure of the oral epithelium.

Analysis of the microstructure of mixtures containing saliva and astringent compounds shown in Figs. 2(a)–2(c) revealed an increased salivary protein binding, precipitation, and formation of insoluble complexes [8, 26, 59, 75]. De Wijk and Prinz [76] investigated
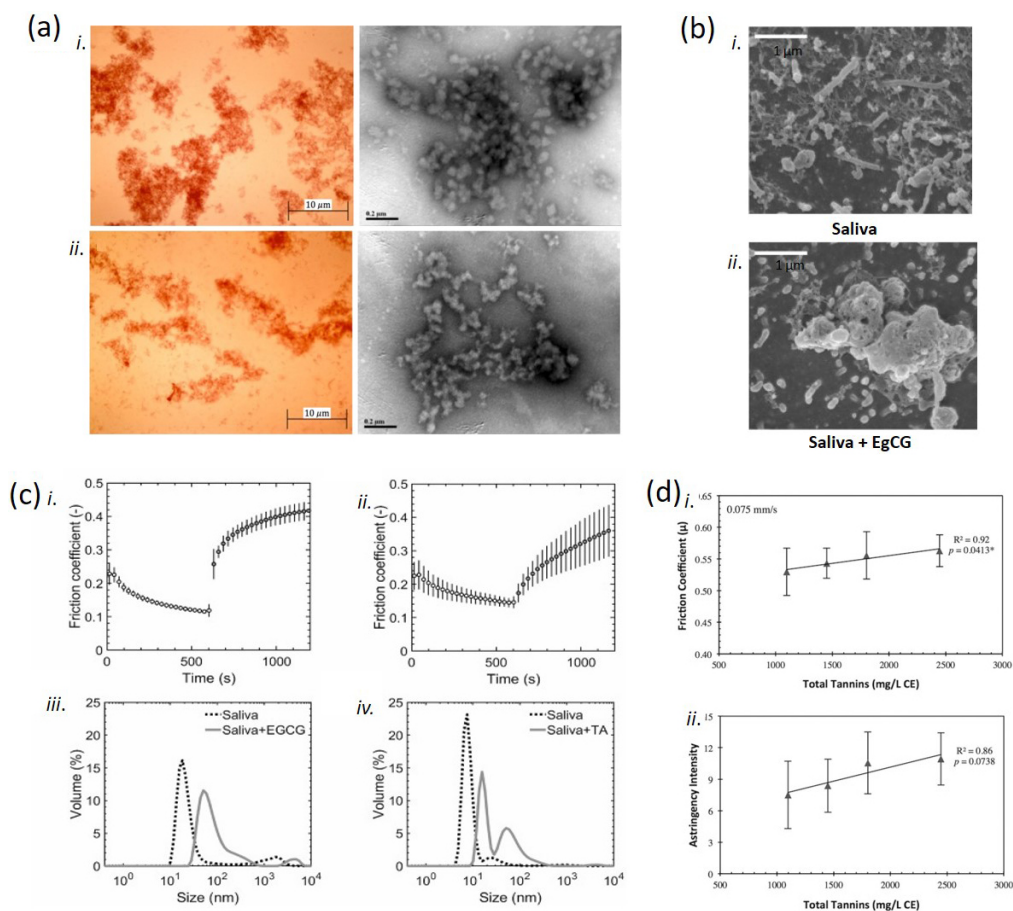
Fig. 2 Effect of the addition of astringents to saliva showing (a) images of saliva-tannin(red wine) aggregates using a light microscope (left) and transmission electron microscope (right). Reproduced with permission from Ref. [8] © John Wiley and Sons 2016. (b) Scanning electron microscope images for saliva and saliva-epigallocatechin gallate (EgCG) mixture. Reproduced with permission from [75] © Elsevier 2018. (c) Coefficient of friction for (i) EgCG and (ii) tannic acid, and changing particle sizes for (iii) EgCG and (iv) tannic acid. Reproduced with permission from Ref. [26] © Elsevier 2021. (d) (i) Friction coefficient and (ii) astringency intensity for increasing concentrations of tannins in red wine. Reproduced with permission from Ref. [8] © John Wiley and Sons 2016.

the aggregation of debris shed from epithelial cells following the addition of polyphenolic compounds to saliva. These mucin and PRP aggregates were considered to be particles capable of inducing a roughness sensation and increasing friction at oral surfaces (see Fig. 2(d)) [8, 23]. Rudge et al. [26] showed a correspondence between the increasing particle sizes when astringents were added to saliva, and an increase in the measured coefficient of friction (see Fig. 2(c)). However, the same study observed aggregation of salivary proteins without any changes to friction, suggesting different interactions at play at the interface depending on the structure of the binding polyphenol [26].
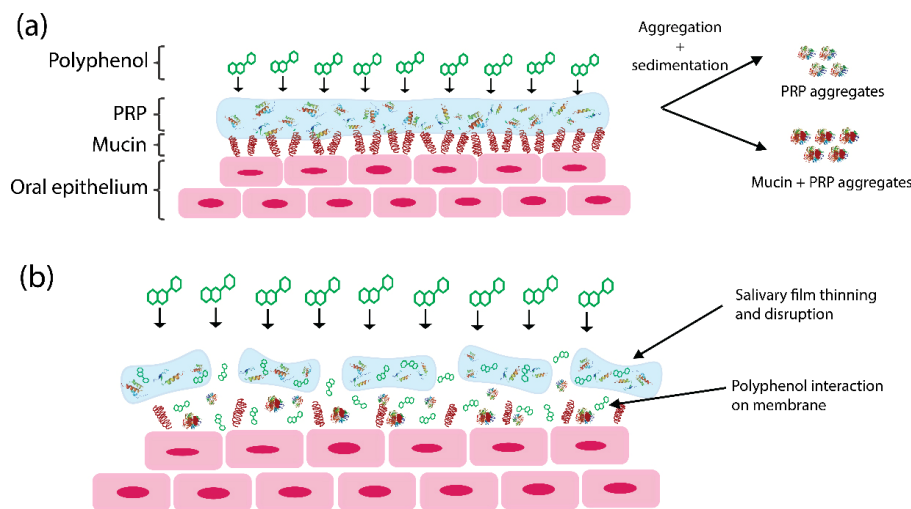
The binding of salivary proteins and polyphenols also results in depletion/disruption of the salivary proteins/mucosal film and exposure of the oral epithelium. This loss of salivary proteins and breakdown of the salivary film means saliva is no longer able to carry out its lubricating function and has been associated to the dryness and roughness sensation attributed to astringency [77, 78]. A combination of protein-polyphenol aggregation and disruption of the salivary film as shown in the schematic of Fig. 3, allows for direct interaction of soluble tannin-protein aggregates and other oral constituents with the oral epithelium [21, 79]. This could lead to increased stimulation of receptors embedded within oral tissues which are responsible for transmitting signals to the brain, possibly eliciting an astringent sensation. Tactile activation of oral mechanoreceptors has in the past been attributed to playing a major role in oral textural perception [20, 80].
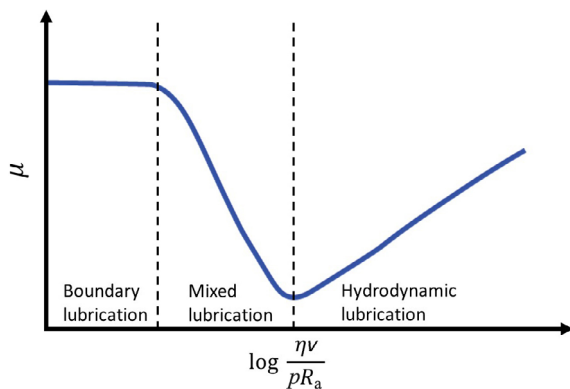
4 Oral lubrication

The friction characteristics of lubricated surfaces is defined across the three lubrication regimes, referred to as hydrodynamic, mixed and boundary lubrication. Typically, each regime can be identified and predicted by the characteristic response of the friction coefficient (μ) to contact pressure (p), sliding velocity (v), center line average surface roughness (R_a) and viscosity (η). This is represented by a Stribeck curve, as shown in Fig. 4, for conventional tribological contacts, such as those found in machine elements with hard, elastically deforming surfaces [81, 82].

The (elasto) hydrodynamic lubrication ((E)HL) regime is distinguished by sliding surfaces completely separated by a continuous fluid film acting as a lubricating layer. Compared to surfaces in a dry contact, the presence of a fully lubricating salivary layer is expected to significantly reduce the friction coefficient as illustrated by the Stribeck curve. However, there remains a friction response which is dependent on the viscosity of lubricant [83]. Sensory perception of thickness of food such as fattiness, smoothness, and creaminess have been attributed to this regime where bulk rheological properties of foods are dominating [17]. During astringency, interactions

Fig. 3 Schematic representation of the oral epithelium showing possible mode of astringency due to (a) aggregation of salivary proteins and (b) thinning and breakdown of salivary film layer due to polyphenol–saliva interaction.

Fig. 4 Stribeck curve highlighting regions of the hydrodynamic, mixed, and boundary lubrication regimes.

between food and salivary proteins alter the lubricating property of saliva. A decrease in saliva viscosity may result in a transition towards the mixed lubrication regime assuming surface roughness, sliding velocities and contact pressures remain relatively constant.

The mixed lubrication regime contains regions separated by a fluid film and regions where asperity peaks are in contact. This implies friction in the mixed lubrication regime is dependent on the tribo-chemical properties of oral constituents as it involves regions partially separated by saliva-food mixtures and regions where asperities on the tongue and other oral components come into contact [6]. Disruption of the salivary film during the astringency development process results in oral lubrication losses, suggesting that the friction mechanism of the mixed lubrication regime plays a key role in astringency perception. The total friction force in this regime is a combined effect of the shear resistance of the saliva-food mixture and the force needed to overcome/deform contacting asperities.

On the left side of the Stribeck curve is the boundary lubrication regime which is dominated by contacting asperity peaks coated by a boundary layer. Bulk rheological properties at the interface are of little importance in boundary lubrication, rather physico-chemical interactions govern the friction behavior in this regime. Properties such as surface roughness, hydrophobicity of the tongue, and adhesion dominate the friction characteristics within the boundary lubrication regime. Loss of proteins in saliva and the resulting disruption of the salivary film during astringency suggest that the lubricating layer is no

longer capable of keeping oral surfaces apart and consequently a transition from the right side of the Stribeck curve towards the boundary lubrication regime. This has already been described previously as the lubrication regime experienced on oral surfaces during the tactile sensation of roughness and dryness, which are attributes linked to astringency perception [8, 14]. A compelling argument in favor of this was presented by Vlădescu et al. [84], demonstrating that interfacial shear stresses play a crucial role in the detachment of saliva proteins from oral surfaces. Figure 5 illustrates this shear-induced loss of lubrication observed when astringent food proteins are added to salivary proteins on oral surfaces.

Establishing which one of the lubrication regimes is predominant during food consumption is an ongoing debate, particularly as regards to which properties of food influence the friction response at each regime. Currently, Stribeck curve transition points in oral lubrication systems are difficult to predict as the influence of different parameters on friction is largely unknown. Lubrication models can be used to determine the coefficient of friction and film thickness for fully or partially lubricated systems. Pressure within the fluid film is calculated using the Reynolds equation and the film thickness is updated with a surface elastic deformation equation [85]. Properties of the lubricant can be incorporated through its viscosity which is reflected in the pressure and shear stress terms, both of which define the load carrying capacity and friction respectively. Furthermore viscoelastic property of saliva can be represented using an appropriate viscoelastic fluid model such as the Maxwell, Oldroyd-B or PTT models [86–90]. Parameters within such lubrication models can then be varied to understand how each influence the Stribeck curve transition points.

5   Methods for astringency assessment

Analysis of salivary protein–polyphenol interactions from a tribo-chemical perspective indicate that binding affinities and formation of precipitates/aggregation play significant roles in the friction mechanism during the astringency development process. It is therefore useful to extract test protocols that are capable of
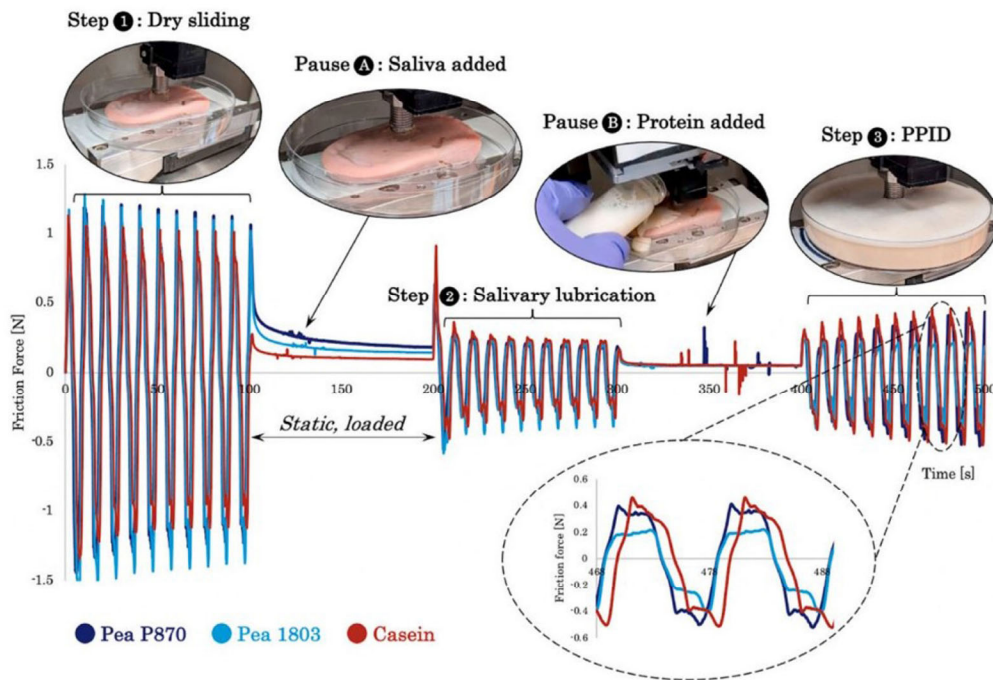
Fig. 5 Comparison of friction forces for different astringent proteins in a 3-step process involving dry sliding, saliva lubrication, and protein induced delubrication of the salivary film. Reproduced with permission from Ref. [84], © Elsevier 2022.

investigating the tribo-chemistry associated with astringency. In this section, methods used in studying the astringency development process are discussed with particular focus on tribological measures in oral lubrication.

The traditional assessment of astringency in food product innovation and development is conducted using a trained sensory panel [90, 91]. Although various sensory techniques are in use, most commonly used is the quantitative description analysis (QDA). In QDA, the attributes on which the panelists score are first generated by the panel and discussed until consensus of the description. The products are scored for all attributes on a scale of 0–100 and the obtained data is statistically analyzed to identify significant differences.

Given the fact that panel tests are time consuming, less reliable at high polyphenol concentrations, costly and can only serve as a subjective judgment of perception, alternative testing methods are under development. The goal is to find objective measures that can serve as predictors for the subjective sensation of astringency. Clearly, panel tests are always needed to verify improvements that are predicted based on objective measures.

Alternative techniques from the perspective of objective measures are currently in use to determine the boundary layer properties of saliva in astringency studies. Table 2 summarizes some of these measurement techniques. These methods are effective in studying how several factors such as pH, temperature, and saliva composition, regulate the interaction of salivary proteins and astringent compounds. Given that a compelling relation between protein aggregation/ lubrication loss and the sensation of astringency is lacking, it is essential to develop rigorous tribo-chemical test protocols focused on understanding the highlighted astringency mechanisms (see Section 3) on oral surfaces. This also creates a pathway for validating multiple theories using sensory panel tests.

Some attempts at establishing this connection between changes at the saliva–tongue interface and the roughness/dryness sensations linked to astringency made investigations using tribometers. In its basic mode of operation, a tribometer uses a controlled relative motion over a range of applied loads to obtain friction measurements of interacting surfaces. Depending on the system under investigation, several types of tribometers are available for specific testing conditions. One of the pioneering works in oral

Table 2 Measurement techniques in astringency studies from a tribo-chemistry viewpoint.

Measurement technique	Principle of operation	References
Fluorescent microscopy	Imaging technique based on the excitation of fluorophores in labelled samples and the detection of the fluorescence signal using optical microscopes. This technique has been employed to detect labelled saliva proteins at the contact zone between sliding surfaces. Astringency studies employing fluorescent microscopy image the interaction between salivary and plant proteins by replicating the conditions encountered within the oral cavity. They measure size distribution and changes in fluorescence intensity of protein-phenol aggregates.	[75, 84, 92–95]
Electron microscopy	Uses beams of accelerated electrons to obtain high resolution images by either transmitting electrons through or scanning the surface of a specimen. This technique has been used to observe microstructure features of astringents and saliva. Also towards the morphology and surface characterization of aggregates.	[8, 78, 96, 97]
Dynamic light scattering	Measures particle sizes via the autocorrelation of the intensity of reflected light passing through the sample. Mostly used in astringency studies to measure sizes of protein-polyphenol aggregates.	[26, 69, 93]
Size exclusion chromatography	Measures size distribution based on the elution times of molecules. Previously applied towards astringency by quantifying binding parameters of polyphenols to proteins.	[2, 98]
Atomic force microscopy	High-resolution topography imaging, force spectroscopy, and friction measurements of samples in dry and humid environments via a scanning cantilever probe. Applications in nanotribology through topography and friction images of tannins interacting with salivary proteins.	[91, 97, 99–101]
Quartz crystal microbalance with dissipation	Measures adsorption of molecules using an oscillating piezoelectric quartz crystal sensor. Typically applied to observe changes in the structural properties of the salivary layer from interactions with polyphenols.	[100, 102]
Zeta potentials	Quantifies electrostatic interaction of protein molecules. Commonly applied towards understanding the effect of pH on the zeta potential and the role of protein charge in astringency perception.	[78, 103, 104]
Nuclear magnetic resonance	Measures interaction of nuclear spins of materials in a magnetic field for molecular structure analysis. Specifically, chemical shifts are used to determine conformations of proteins.	[105–107]

tribology by Kokini et al. [108] used a friction tester which was one of the simplest methods available to measure the friction induced at oral surfaces by food. After many years, a friction tester consisting of a spherical ball rotating on a rubber band was used by De Wijk, and Prinz [76] who investigated the textural characteristics of food and established a correlation between roughness/astringency and higher friction as opposed to creaminess/fattiness and lower friction.

The mini traction machine (MTM) is another device commonly used to measure the frictional properties at contact interfaces across a range of sliding and rolling conditions. This device consists of a steel ball loaded against a flat disk immersed in a fluid (lubricant) at a controlled temperature [34, 90]. Rossetti et al. [25] used a mini traction machine with a PDMS coating on the ball and disk to measure the frictional properties of human whole saliva in the presence of astringent tea catechins.

Scientists have also modified Texture analyzers for

tribological studies because of their capability for measuring friction forces at controlled temperatures, sliding speeds, and surface loads. This modified device is also able to measure friction/lubrication properties across the different lubrication regimes and has been adapted for studies relating friction and perceived astringency intensity [8, 78].

Although much progress has been recorded in friction studies of saliva-food mixtures at oral surfaces using tribometers, the tribosystems in these tests differ significantly from in-vivo conditions [33, 76, 109–112]. Research has already shown that variations in surface chemistry and material stiffness have a significant impact on both boundary friction and thickness of the elastohydrodynamic lubrication (EHL) film at different sliding velocities during oral lubrication [94]. System dependence of friction makes the choice of components, relations, and functions of tongue models critical towards studying the tribo-chemistry involved in the sensory perception of

astringency. Therefore, improvements to the highlighted tribometer setups should consider tribosystems that are equivalent to oral components.

On the other hand, computational methods such as molecular modeling provide a robust tool for investigating the molecular basis of polyphenol-protein binding and the shear stresses at the boundary layer. Another promising trajectory is the use of contact models that are able to capture how structural changes at the oral interface are transformed into microscopic strains at receptor locations for sensing.

6   Summarizing conclusions

Plant proteins have become a popular diet alternative as they offer a higher protein content to carbon emission ratio compared to animal based proteins. These plant proteins containing polyphenols are known to be astringent, leading to a dry/rough oral sensation which is unpleasant to most consumers. Researches on astringency aim to minimize these unwanted taste sensations elicited by plant-based protein products as a means of fostering the protein transition which is essential for sustainability.

Oral perception involves mastication, mechanical and chemical stimulation, receptor sensing, signal propagation, cognition before finally establishing sensory perception. In the available literature, much work has been carried out in studying the interaction of salivary proteins with polyphenols at the mastication stage. This has led to interesting findings on the role played by aggregation of salivary proteins in lubrication losses experienced on oral surfaces. What is missing, however, is determining the influence of tribo-chemical changes of the saliva based lubricating layer on the physiological sensation of astringency. Closing this gap requires enhanced studies into the tribo-chemistry involved in the astringency development process. Examining the results obtained from studies exploring the tribo-chemical basis for astringency perception reveals some insights for future development efforts:

1) **Predictive modeling of the Stribeck curve:** Tribological analysis of oral contacts have introduced tribopairs notably different from what has been extensively studied for conventional machine elements.

Assumptions such as the Hertzian approximation are no longer valid in soft contacts. For this reason, much of the substantial achievements on predicting transition points of the Stribeck curve which were largely known for machine elements remain unknown in the current contact of interest. This raises the question on how the Stribeck curve can be predicted in soft contacts. A solution to this problem requires a contact model that incorporates the behavior of all components within the tribosystem, analyzed over a broad range of operating conditions. Viscosity in the hydrodynamic regime has already been identified as a main controlling parameter. Similarly, parameters such as the interfacial shear strength can reflect the tribo-chemistry at the boundary lubrication regime and be utilized to predict transition points of the Stribeck curve.

2) **Effect of saliva-astringent complexes on the boundary layer:** Dryness/roughness sensations elicited during astringency is ascribed to the chemical interaction of salivary proteins and polyphenols, forming protein-phenol aggregates which can act as third-body particles during sliding motion of the tongue. The reported variations of tactile sensation between structurally different polyphenols suggest that different characteristics of protein-phenol aggregates influence friction at the boundary layer. This highlights the need for a tribo-chemical approach in the study of astringency. The aspect of astringency research investigating properties of protein-phenol aggregates capable of inducing dryness/roughness sensations is yet to be largely explored. Properties of aggregates such as their roughness, stiffness/hardness, size and shapes could be valuable pointers towards the observed perception differences related to astringency.

3) **From tribo-chemical interactions to panel test via multiscale modeling:** A main goal of future tribo-chemical studies on astringency should be towards providing insights on the relation between chemical reactions, oral friction, and human cognition, making it possible to define predictors of astringency. Attempts at establishing a psychophysical interface has already been made by studies relating sensory panel results to friction at the macroscale. Contact at the microscale on the other hand, which could

have a huge potential in shedding more light on the interactions of food particles with oral surfaces, remains largely unexplored. This can be particularly interesting for studying how different properties of saliva-astringent complexes influence contact conditions with oral components resulting in mechanical stresses at possible mechanoreceptor locations. Also providing insights as to what conditions could potentially trigger a higher/lower stimulation of the receptors.

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Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article. The author Emile van der HEIDE is the Editorial Board Member of this journal.

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