Lubrication of soft oral surfaces

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Abstract

Oral lubrication deals with one of the most intricate examples of biotribology, where surfaces under sliding conditions span from the hardest enamel to soft oral tissues in human physiology. Complexity further arises with surfaces being covered by an endogenous biolubricant saliva before exogenous food particles can wet, stick, or slip at the surfaces. In this review, we present a description of soft oral surfaces, comparing them with the recent approaches that have been used to study oral lubrication using *in vitro* to *ex vivo* setups. Specifically, lubrication behaviors of saliva and soft microgels are discussed highlighting instances of hydration lubrication. We have structured this information creating a strong link between theoretical concepts and oral lubrication, which has thus far remained elusive in literature. Finally, we highlighted some of the several challenges remaining in this field and discussing how emerging technologies in material science might help overcoming them.

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Introduction

Oral lubrication involves mechanisms and processes in dissipation and manipulation of frictional forces arising from contact of two surfaces within the oral environment. Examples include teeth–teeth, tongue–palate, and tongue–mucosa interfaces, all of which are lubricated by an endogenous biolubricant 'saliva' or exogenously administered lubricants, such as food particles, oral drugs, mucosal coatings, and oral care products. Oral lubrication plays a vital role in effective functioning of oral tissues, eating, swallowing, speech, tactile perception, and bacterial adhesion that are fundamental to our life. Interestingly, vast majority of the initial studies in this field were mainly concerned with wear of the hardest interacting surface , that is, enamel in mouth involving innovations in dental restorations and implants [1–3]. However, in recent times, there has been a shift in research focal point towards understanding oral lubrication in soft sliding interfaces [4]. Quantifying friction coefficients (μ) in polymeric analogs of soft tongue sliding/rolling against palate surfaces is becoming increasingly recognized to approximate the mechanically complaint characteristic of biological contacts to establish correlations between oral perception and instrumentally characterized texture of food products. Besides strong interests of food colloid scientists, oral lubrication is also gaining momentum as a contributing discipline in biomedical and personal care areas involving orally administered liquid medicines [5], biofilms [6], dry mouth therapies [7], oral care products [8], and so on.

Eating, or in other words, oral processing, is a highly dynamic well-coordinated process that occurs across a range of length- and time-scales involving bulk rheology (flow behavior and deformation under shear, compression, and elongational fields), tribology (friction or lubrication, surface interactions), colloidal interactions (depletion, bridging, ion binding), temperature/pH change as well as material transfer of the food and/or saliva mixtures [9–12]. The work by Kokini et al [13], in 1977, is often cited as one of the seminal works of oral lubrication, where it was demonstrated that bulk properties, such as apparent viscosity (η) alone was not sufficient to predict mouthfeel sensations, such as 'smoothness' and 'slipperiness' in food colloids. These aforementioned sensory dimensions were rather related to interactions of food with tongue–palate surfaces and inversely correlated with μ. After 11 years of this experimental study, the term 'degree of lubrication' was first used by Hutchings and Lillford [14] as one of the three-dimensional axes in theoretical framework of oral breakdown of food. It was proposed that oral lubrication

depends on kinetics of saliva released in the mouth when exposed to different food.

Of note, in the last decade, there has been a renaissance in research efforts in oral lubrication, which has been outlined in a number of recent reviews [15–19]. Majority of this research trigger can be attributed to quantifying food textural perception that are now even more pertinent than before because of global obesity epidemic that increasingly demands for rational designing of low-calorie foods and satiety-enhancing foods [20] without losing their sensory and mouthfeel pleasure. Furthermore, lubrication measurements can be vital in designing foods with just-right oral textural properties, mucosal lubricants, and orally administered medicines for rising aging population, who have suboptimal oral processing capabilities and reduced quantity and quality of saliva [21–23]. Partly, the upsurge in oral lubrication studies is also linked to off-the-shelf availability of devices, such as ball-on-disc and pin-on-disc tribometers in recent times without the need for laboratory-specific designing of tribometers [24,25]. In the last two to three years, instrumental oral lubrication studies have shown interesting correlations with several complex mouthfeel dimensions, such as boundary lubrication failure to 'astringency' in wines [26], lower μ values to 'creamy' mouthfeel in fat-rich semisolid dairy colloids [27], and higher μ values to 'pasty' mouthfeel in hydrogels [28] in trained to untrained consumers.

Considering the importance and topical nature of oral lubrication research, this review aims to cover the latest advances in tribology research in soft oral contacts and theoretical developments relevant for this field. We briefly cover the surface roughness, modulus, wettability, and deformation of human soft oral architectures under shear fields, which influence oral lubrication. The review highlights the transition from rheology to tribology in oral dynamics and focuses on one endogenous (saliva) and one exogenous (food-based microgels/hydrogel particles) lubricant to capture recent knowledge gathered in how they reduce friction in sliding soft contacts. Specifically, we discuss these lubricants highlighting the relevance of their adsorption behaviors as well as and viscous properties under confinement contributing to lubrication in various regimes. For the reasons of space, oral lubrication and physicochemical interactions of other exogenous lubricants, such as emulsions and emulsion gels that have shown fascinating correlations to mouthfeel attributes, will not be discussed within this review, and interested readers may refer to previous reviews [10,19]. Although there have been recent reviews on oral tribology [15–19], the novelty of this review is that we focus on theoretical approximations used to understand the mechanisms behind lubrication of soft oral surfaces by endogenous/exogenous lubricants, ranging from load bearing abilities to drag force dependent entrainment. We discuss the

legacy of materials used for oral lubrication studies in the last decade covering latest experiments conducted mainly using polydimethylsiloxane (PDMS) tribopairs during *in vitro* and pig tongue surfaces in *ex vivo* setups that have not been systematically reviewed to date. For future, we highlight the window of opportunity offered by a range of polymeric surfaces together with the advent of 3D-printing technology that can be used to emulate oral surfaces with accurate roughness and modulus to enable colloid scientists to underpin principles that govern oral lubrication at colloidal scale.

Material properties of oral surfaces

In the last decade, there has been an extensive experimental research focusing in the tribological aspects of oral processing. However, surfaces used to mimic soft oral tissue contacts are mostly silicon-based materials with limited topological, chemical, or even, in most cases, mechanical resemblance to soft oral surfaces. Hence, we give a brief overview of the material physics and chemistry of real human oral surfaces to set the scene for understanding the relevance of synthetic analog surfaces used for doing oral tribology experiments. For instance, this will also enable to compare the stiffness and surface roughness of real human oral surfaces with synthetic analogs and question the biorelevance of the frictional forces measured using the polymeric analogs. Surveying the intricate features of biological surfaces in the oral cavity (Figure 1a), one can imagine oral lubrication involving soft surfaces can occur in hard–soft (hard palate–tongue), soft–soft (tongue–soft palate) surfaces using saliva, food particles, or other mucosal lubricants. Tongue surface is not smooth and has a palette of embedded papillae resulting in a multitude of surface roughness in different areas (Figure 1b). Almost front two-thirds of the human tongue's surface is covered by numerous filiform papillae that contain no taste buds and are believed to be contributing to friction and mechanosensation. Besides filiform papillae, taste bud–containing mushroom-shaped fungiform papillae and other papillae (foliate, circumvallate) also anchor to the tongue surface encoding taste perception [29] that have gathered most research attention in the literature. Spectrum of material physics properties, such as surface roughness generated by papillae structure [30,31] and modulus [32] of tongue surface, is quite unique. For instance, the human filiform papillae can contain 6–12 protruding hairs, each of which are 34–50 μm wide contributing to overall 420–500 μm diameter in the root [31] and height of 250 μm, whereas fungiform papillae almost doubling the diameter of the filiform ones (Figure 1b).

Noteworthy, filiform papillae of pig tongue are also in similar order being slightly longer than the human counterparts (height of 320 μm) and width of 120 μm in the root [24]. Recently, the R_a value of the tongue

Figure 1
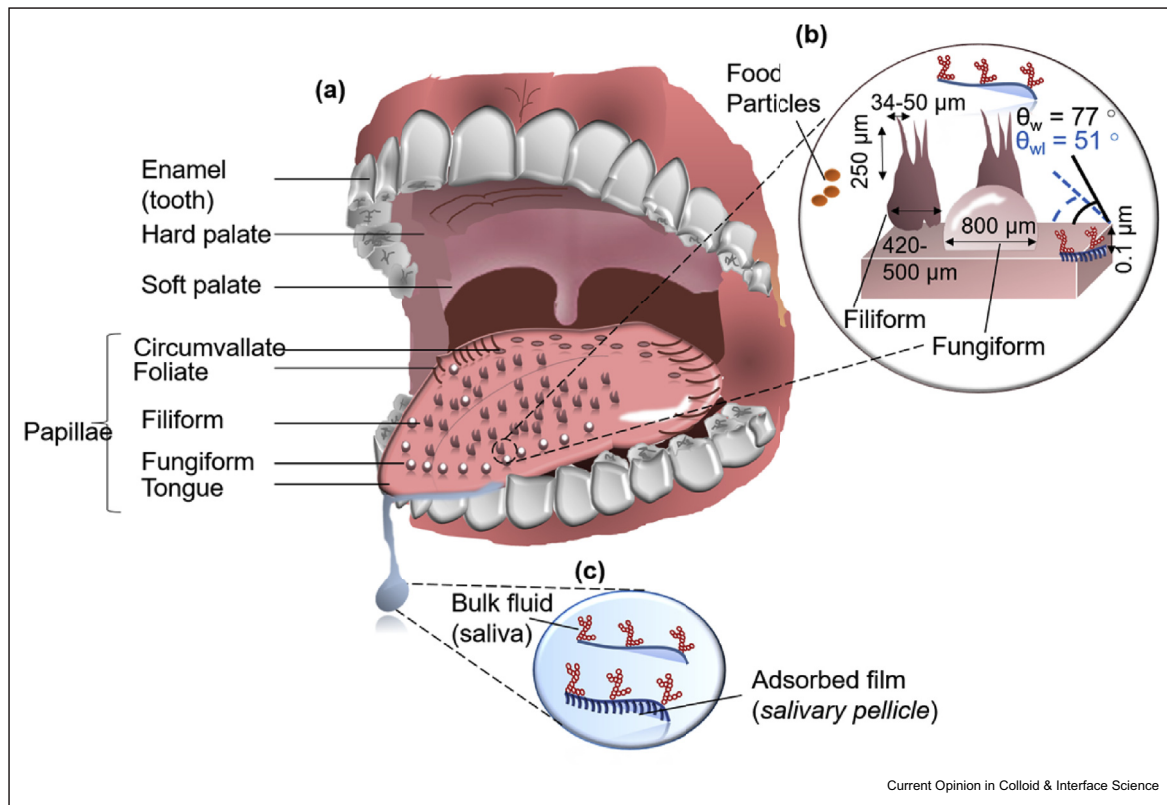
Building blocks of soft oral surfaces. **(a)** A schematic illustration of oral cavity highlighting the soft (tongue) and hard (tooth enamel) oral surfaces with the lubricant (saliva). **(b)** Building blocks of soft tongue surface (keratinized squamous epithelium) showing dimensions of filiform (containing no taste buds) and fungiform papillae at micron scale [33] and its change in wettability (θ_w is the water contact angle, θ_{wl} is the water contact angle upon adsorption of salivary film of nanometer scale) [34]. **(c)** Bulk saliva and adsorbed salivary pellicle, formation of latter is driven by the adsorption of salivary components, such as highly glycosylated mucins to the tongue surface [35].

obtained using lingual impression has been reported to be much smaller, that is, 65.0 µm (range, 42.5–101.4 µm) [36], which may be linked to squishing of papillae by the pressure used during taking the impression. Overall, all these surface topographic analyses point out that engineering of these filiform papillae on tongue surface by nature provides it a more coarse texture than even a 100-grit sandpaper. However, the tongue does not feel as coarse as represented by its roughness largely due its reduced "stiffness" (Young's modulus). Elastic modulus of healthy human tongue and soft palate has been reported to be nearly 2.5 kPa as measured by magnetic resonance elastography under *in vivo* situation [32], which is line with reduced contact modulus of pig tongue measured *ex vivo* using compression tests [24]. In oral conditions, voluntary tongue speeds can go up to 200 mm/s and can support bulk compressive pressure (tongue-palate) of one order of magnitude higher (30–70 kPa [37,38]) than its own modulus without plastic deformation, largely attributed to the vascularity of the tongue. Friction coefficients for the tongue against other biological tissues have been

postulated to be about 0.1–0.3 [39]; however, it may change considerably depending upon the type of lubricants being introduced in the tongue–tissue contacts.

From a material chemistry viewpoint, tongue is keratinized and can be considered intrinsically hydrophobic and weakly polar [40] (Figure 1b). However, tongue surface becomes hydrophilic with contact angle of 51° upon wetting by saliva. Saliva contains 99% water and <1% protein with material properties far from water and can be classified into a fluid-like bulk saliva as well as an adsorbed film, that is, salivary pellicle (Figure 1c). Salivary proteins adheres to the oral surfaces and helps to maintain an adsorbed salivary film, that is, saliva pellicle thickness of 30–100 nm [35,41,42], although this thickness may vary depending upon the pellicle's location in the mouth. That is why, in patients suffering from xerostomia (dry mouth originating from polymedication or Sjögren's syndrome), loss of saliva pellicle may result in poor surface hydration, reduced wettability, and, therefore, higher hydrophobicity of tongue surface. It has been also proven *via* ellipsometry in early 1990s that saliva

adsorbs better to hydrophobic than to hydrophilic surfaces (*ex vivo*) and the ions (up to 0.15 M) present in saliva also play a key role in enhancing such adsorption, largely attributed to electrostatic interactions [43]. It is worth considering that, although wettability is completely driven by salivary film prefood ingestion, wetting dynamics of tongue surface can be driven possibly by three kinds of adsorbed films during oral processing. These are (1) saliva-deficient or reformed saliva-dominant film, (2) saliva—food particle mixture—dominant film, or (3) food particle—dominant films, attributed to interactions of food with this salivary film at multiple lengths and time scales. However, it remains largely unknown which of these three mechanisms dominates, and this has an implication on mouthfeel that may range from 'astringency' to 'creamy' perceptions.

Mechanical approximations to oral lubrication: from rheology to tribology

Classical rheology and tribology are well-established techniques to study the forces arising from relative motion of solid surfaces in the presence of a fluid. From a phenomenological point of view, these techniques are concerned with different limits determined by the level of proximity among the surfaces in play. In the rheological limit, a thick film of fluid, typically in the order of hundreds of microns, separates surfaces with negligible role played by the solid—fluid interface [16]. In the case of complex fluids, distance between the surfaces can be at least one order of magnitude larger than the characteristic length of the single components, such as radius of gyration of the individual polymers in a solution or particle size of the microgels in a colloidal suspension. Under these ideal conditions, rheology is used to determine the bulk properties of materials such as linear elasticity, viscosity, and yield stress, for applications involving macroscopic flow in mass transfer phenomena. The capabilities of rheological techniques relies on subjecting the materials to simple flow conditions to facilitate a connection between the measured forces and macroscopic deformation. In addition, analysis of rheological data using appropriate constitutive equation or microscopic models enables to obtain information about the composition and structure of materials. For example, the well-established tube model is a powerful tool to determine the molecular weight distribution of polymer melts using the linear viscoelastic response of the material [44], otherwise difficult to quantify using gel permeation chromatography. Highly entangled polymers (among other complex fluids) represent also a challenge for rheological techniques because of the appearance of wall slip, which is caused by the failure of the surface to transmit the macroscopic deformation to the bulk material when the stress overcomes a certain value [45]. The threshold of wall slip is determined by the surface and material properties as well as the rheological geometry. An important characteristic of wall slip is the lowering of the stress thresholds by increasing the surface to volume ratio, for instance, by decreasing the thickness of the fluid film.

The role of the surfaces in fluid mechanics is more of a concern to the other end of the spectrum, that is, the tribological limit. Classical tribology deals with surfaces in close proximity defining three regimes spanning from direct to partial surface contact to surfaces fully separated by a continuous fluid film, traditionally described by Stribeck curves. Commonly, Stribeck curves describe the performance of lubricated contacts representing the friction coefficient as a function of the working conditions (load, surface speed) and lubricant bulk viscosity. Bulk mechanical properties of the lubricant are considered to be relevant in the tribological regimes involving a continuous fluid film separating the contact surfaces. However, the tribological limit involves thin fluid films, where the thickness of the fluid film approaches that of the surface roughness. Considering the basic definition of shear rate as the ratio of sliding speeds of the surfaces to the fluid film thickness, tribology explores the response of fluids at shear rates at least an order of magnitude higher than the maximum shear rate commonly reported in literature, that is, about 1000 s^{-1}. One might argue that the conventional plate—plate rheological geometry has been demonstrated to be capable of achieving shear rates as large as 10^5 s^{-1} when hydrodynamic flows that appear at small gap ($\sim 30 \text{ μm}$) are considered [46]. Nevertheless, in tribology, the thickness of the fluid film (hence, the shear rate) is determined by a balance between the load supported by the contact and the hydrodynamic pressure buildup in the fluid, which depends on fluid viscosity, surface speed, and contact geometry. Unlike rheology, tribology is concerned with how the presence or absence of fluid and its interaction with the contact surfaces determine the friction forces and their development with the working conditions.

Oral processing involves both rheological and tribological limits [17]. As depicted schematically in Figure 2 using a case of filiform papillae of tongue, rheological properties dominate the first stages of oral processing. At this stage, the bulk properties of food in continuum determine the forces appearing between oral surfaces. Sensory attributes, such as thickness, gumminess, and chewiness, are mainly related to bulk rheological properties in this early stage of oral processing. When oral processing progresses, the fluid film start to decrease in thickness due to swallowing and the tongue papillae are in close contact with the palate surface (Figure 2); here, mouthfeel perception, such as creaminess and pastiness, arises considering the importance of lubricant—surface interactions.

Lubrication science is the protagonist in the later stages of oral processing, which is not only a hypothesis but also

Figure 2

Current Opinion in Colloid & Interface Science
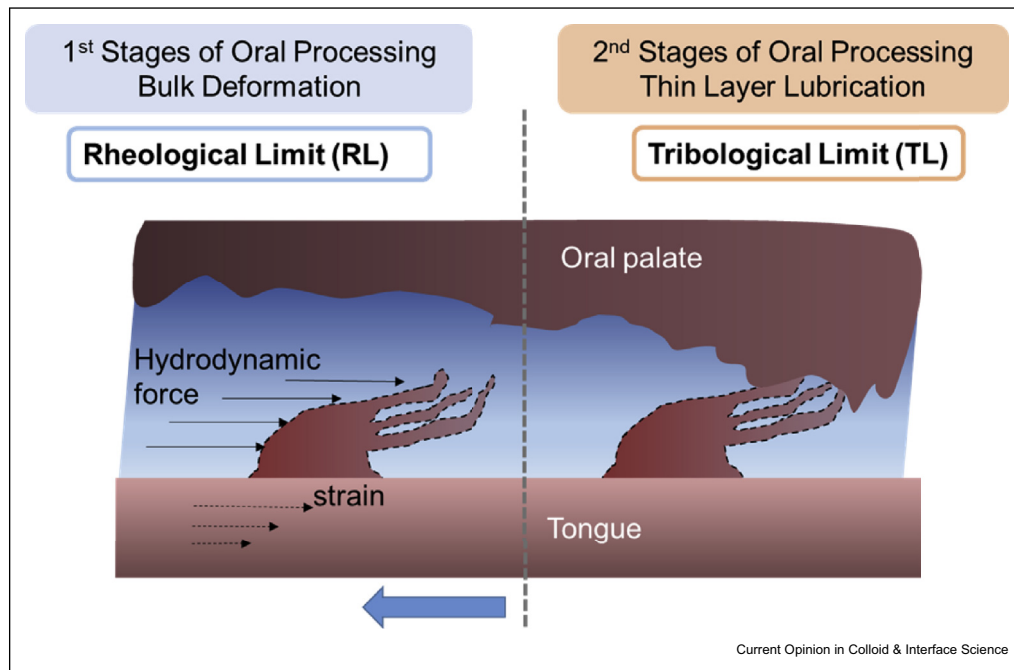
Rheological and tribological limits during oral processing. Schematic illustration of the soft tongue tissue (lower surface) and hard palate (upper surface) contact with flow of food and/or saliva shown in blue. On the tongue surface, only crown-shaped filiform papillae are shown. In the rheological limit, bending of filiform papillae due to hydrodynamic forces induces strain on the tongue substrate. In the tribological limit, some or all hairs of the papillae may be in asperity contact with the soft palate with the lubricant being confined with the moving papillae−palate surfaces, affecting frictional force and mouthfeel perception.

can be evidenced using recent experimental investigations. For example, a recent study in our laboratory by Krop et al. [28] has shown significant correlations between bulk properties of edible hydrogels made up of κ-carrageenan with or without structural homogeneities (introduced by adding other hydrocolloids, such as sodium alginate, locust bean gum, or calcium alginate beads [47]) and sensory perception. Apparent viscosity of the hydrogel boli (*i.e.* hydrogel + artificial saliva after simulated oral processing) showed strong correlation with sensory properties, such as 'firmness', 'elasticity', 'chewiness', and 'cohesiveness'. However, mouthfeel-related attributes that are experienced in later stages of oral processing, such as 'pastiness' and 'slipperiness', showed correlations with friction coefficients of hydrogel bolus filtrates, when large hydrogel bolus particles (>500 μm) were deliberately filtered out. This highlights the dominance of tribological limit in the later stages of oral processing over rheology, considering all the hydrogel boli [28] had similar apparent viscosities across the shear rates (from 0.001 to 100 s^{-1}) except for the ones containing sodium alginate. In similar fashion, Laguna et al. [27] demonstrated that 'creamy' mouthfeel discrimination was possible between isoviscous full and low/no-fat semisolid colloids using tribology experiments. In tribological regime, the surface properties

play a paramount role in the development of forces between the rubbing surfaces (Figure 2). Tribological experiments in combination with quartz crystal balance tests on polysaccharides solutions demonstrated that the boundary friction is essentially dominated by the amount of polymeric material absorbed onto the contact surfaces [48]. In addition, one should bear in mind that very thin films of complex fluids (submicron to nanometer scale thickness) in the tribological limit might have different mechanical properties compared with their bulk rheological properties [49]. For example, using particle-tracking microrheology, Haro-Perez et al. [50] demonstrated that polymer solutions would undergo a fluid−gel transition when confined within gaps comparable with the gyration radius of the polymer. Thus, it is expected that tribological performance of complex fluids is not explained simply by their macroscopic rheological behavior. It is important then to remark that the physical and chemical phenomena influencing the rheological and tribological limits are different and both frameworks are important to understand oral processing.

Mechanical forces developed during oral processing, either in rheological or tribological limit, are influenced by surface texture of the tongue containing papillae with

average size of few hundreds of microns as discussed previously. Based on fluid mechanics calculations, Lauga et al. [51] recently estimated that filiform papillae in our tongue can act as strain amplifiers. They considered a rheological scenario, where hard palate and tongue are separated by a simple Newtonian fluid (Figure 2). Filiform papillae were assumed to have cylindrical shape and similar Young modulus to the soft tongue tissue. Under this approximation, they estimated that the strain induced on the tongue substrate by the deformed papillae pillars was due to the rheological shear and squeeze flow. Expressions for shear γ and squeeze flow ε induced strain are given in Eqs. (1) and (2), respectively.

$$\gamma \sim \frac{UL^3}{a^3 EH} \tag{1}$$

$$\varepsilon \sim \frac{VDL^3}{a^3 EH^2} \tag{2}$$

Here, D is the characteristic length of the surfaces, H is the working distance between tongue and palate, U is the sliding speed, V is the approximation speed, and E, a, and L are the elastic modulus, radius, and length of the papillae pillars, respectively. Evaluating Eqs. (1) and (2) for average oral processing conditions, they found that the strain induced in presence of papillae pillars was at least one order of magnitude higher than in their absence. Thus, filiform papillae are of high importance in the mechanic perception during the oral processing. To date, there are no tribological experimental data sets available to confirm the role of papillae as strain amplifiers. Furthermore, a number of questions remain to be answered in future on how filiform papilla behave in the tribological limit and how such changed surface roughness due to deformation affects friction coefficients when lubricated by Newtonian as well as non-Newtonian lubricants.

Oral lubrication of soft surfaces by complex fluids

The oral cavity is a unique environment that is subjected to different niches of complex fluids. We will focus on two key complex fluids, namely (1) non-Newtonian saliva that is endogenous to the human mouth and (2) soft microgels (protein- or starch- or lipid-based) [52,53] that is exogenously used, latter representing soft food or oral medicines. We will discuss their corresponding interactions with the oral-mimetic surfaces; with compliance nature of soft tongue tissue been commonly mimicked using PDMS surfaces in the last decade.

Performance of lubricated contacts is commonly divided into three regimes in the Stribeck curve (Figure 3a) based on the response of friction coefficient on the changes of the working conditions (fluid viscosity, surface speed, load).

In the boundary lubrication regime (Figure 3a), occurring commonly at the lowest working speeds, the friction coefficient shows limited to no dependence on surface dynamics. Boundary lubrication dominates during periods of high load and low velocity, when the hydrodynamic forces are negligible [57]; thus, this regime is characterized by the absence of lubricant in the contact zone. At this stage, friction forces are determined by surfaces properties, such as viscoelasticity, roughness, and interactions of a surface-bound film rather than the rheology of a confined lubricating fluid. For instance, although Amonton's friction law states that dry friction coefficient of solid contacts is independent of the supported load (W), friction coefficient of soft compliant contacts changes with W following approximately the power low $\sim W^{-1/3}$. This dependence has been shown to correlate with changes in contact area that can be calculated using the classical Hertz contact theory. Mathematical expressions for the contact radius (a_H) and indentation (δ) for point contacts subjected to a load W are given in Eqs. (3) and (4):

$$a_H^3 = \frac{3}{4} \frac{WR^*}{E^*} \tag{3}$$

$$\delta = \frac{a_H^2}{R^*} \tag{4}$$

where $E^* = \left(\frac{1-v^2}{E'} + \frac{1-v^2}{E''} \right)^{-1}$ and $R^* = \left(\frac{1}{R'} + \frac{1}{R''} \right)^{-1}$ are the reduced Young's modulus and reduced radius of the contact, respectively. Here E', E'' are the elastic moduli and R', R'' the radius of the surfaces. Eqs. (3) and (4) are valid for small deformation limit where $\delta << R^*$. The maximum pressure at the geometric center of the contact is $P_{max} = 3W/2\pi a_H^2$. In the case of the popular PDMS ball on disc tribological set-up ($E^* \sim 2$ MPa, $R \sim 0.01$ m) with a typical load of 2 N, the maximum contact pressure is ~ 200 kPa. This pressure is about one order of magnitude higher than that measured in oral-palate contact of healthy adults (30–50 kPa). Mimicking oral conditions in the described setup would require using loads below 0.1 N or softer surfaces with elastic modulus closer to the real oral-palate contact.

When the contacting surfaces are coated by an amphiphilic macromolecule with polar head groups, a dramatic reduction of friction coefficient can be envisaged (Figure 3a). For example, charged polymers attached to the surfaces are known to trap and accommodate water molecules in hydrated layers. This phenomenon known as hydration lubrication has been a focus of attention because of its relevance in physiological processes [58–60]. Interestingly, proteins have been shown to act as aqueous boundary lubricants in physiology, such as lubricin (a mucinous glycoprotein) in articular joints

Figure 3
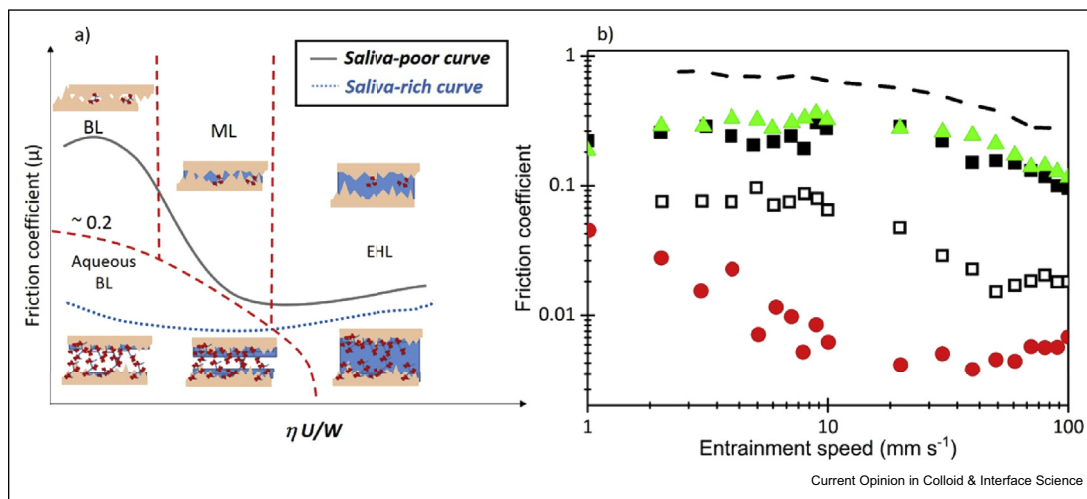
Lubrication performance in soft mechanical contact. **(a)** Stribeck curves and schematic representation of the lubrication performance of saliva in soft compliant contacts. Stribeck curves show the friction coefficient as a function of the main working conditions (load *W*, viscosity η, surface speed *U*) determining the ratio between hydrodynamic entrainment ($\sim\eta\,U$) and contact load. High molecular weight mucins contained in saliva form a brush-like layer entrapping water in a hydration layer, thus causing low values of friction coefficient (saliva reach curve). This regime is an example of aqueous boundary lubrication (aqueous BL) [54−56]. The saliva−poor curve illustrates the performance of contacts lubricated with saliva containing lower amounts of high-molecular-weight mucins and other lubricating salivary proteins. In this case, the brush-like structure is not formed resulting in high friction coefficients characterized by the conventional boundary lubrication (BL), followed by the mixed lubrication (ML) regime. At sufficiently large entrainment, the elastohydrodynamic lubrication (EHL) regime is observed. **(b)** Comparison of friction coefficients of a range of colloidal dispersions of whey protein microgels (80 vol%) (filled black squares) [52], starch microgels (60 vol%) (green triangle) [53], starch microgels containing emulsion droplets (60 vol%) (red circles) [53], and whole human saliva from one healthy adult (open squares) (Ethics Number: MEEC-16-046, University of Leeds, UK). A Stribeck curve for buffer is shown as a dashed line for comparison purposes [53].

[61] to saliva in oral cavity [62]. In similar fashion, food proteins can be predicted to provide aqueous boundary lubrication; however, there has been limited experimental data to date on boundary lubrication properties of food-based animal and plant proteins either as a monolayer within the asperity contacts or as few layers thick colloidal structures. In addition, complexes, co-acervates, and conjugates of protein−protein [63,64] and protein−polysaccharide [65,66] *via* electrostatic complexation or Maillard conjugation have been studied extensively for over half a century by colloid scientists. Several of these biopolymeric colloidal systems can potentially act as anchored boundary layers yet hold water molecules in the polysaccharide chains that can respond to shear in a fluid manner and contribute to hydration lubrication, which requires future attention.

The mixed lubrication regime is observed as a decrease in friction coefficient as the ratio between hydrodynamic forces to the contact load increases. As the described ratio increases, more lubricant is entrained into the contact area, decreasing the effective asperity contact, and thus the friction force (Figure 3a). Owing to the scarce presence and high confinement of the fluid film in the boundary and mixed regime, the performance of a complex lubricant does not relate to its rheological properties. For this reason, it is not strange that

lubrication-related sensory attributes present correlation with tribological experiments but not with rheological testing [28].

The lowest values of friction coefficients are commonly observed in the mixed regime at the border with the hydrodynamic regime, which marks the transition between partially (discontinuous fluid film) and fully separated surfaces by a fluid film. In the latter, a thin lubricant film supports the contact load in its totality, keeping the surfaces apart and from rubbing directly against each other. Hydrodynamic lubrication is well described by the continuous theory of fluid mechanics represented by the Navier−Stokes and continuity equations. Reynolds provided a simplification obtaining one single equation in the thin film approximation (Eq. (5)):

$$\frac{\partial}{\partial x}h^3\frac{\partial}{\partial x}\eta p(x,y) + \frac{\partial}{\partial y}h^3\frac{\partial}{\partial y}\eta p(x,y) = 12\eta\left(U_x\frac{\partial}{\partial x}h + U_y\frac{\partial}{\partial y}h\right)$$

(5)

Eq. (5) is an example of the Reynolds equation in Cartesian coordinates, representing surfaces sliding parallel to the x-y plane. Here, *p(x,y)* is the pressure, η is the lubricant Newtonian viscosity, *h* the film thickness, and

U the entrainment speed. This equation represents the equilibrium between the pressure gradients generated in the lubricant and the hydrodynamic forces due to relative sliding of the surfaces. It is noteworthy that, in case of parallel surfaces, the right hand side of Eq. (5) vanishes and hydrodynamic lubrication would not occur. Thus, surface topography plays a paramount role in the tribological performance of a contact. By numerically solving the Reynolds equation, de Vicente et al. [67] developed a simple algebraic expression to describe the friction coefficient in soft hydrodynamic lubrication performance of Newtonian fluids in elliptical rolling/sliding contacts, which is presented in Eq. (6).

$$\mu = 1.46\overline{U}^{0.65}\overline{W}^{-0.70} + SRR\Big(3.8\overline{U}^{0.71}\overline{W}^{-0.76} + 0.96\overline{U}^{0.36}\overline{W}^{-0.11}\Big) \quad (6)$$

Here, SRR is the sliding/rolling ratio, taking values from 0.0 in pure rolling conditions to 2.0 in the pure sliding conditions. Eq. (6) describes the hydrodynamic lubrication regime of Newtonian fluids, summarizing working conditions and lubricant (viscosity) and surface mechanical (Young modulus) properties using reduced expressions for speed and load given by $\overline{U} = \frac{U\eta}{E^*R'}$ and $\overline{W} = \frac{W}{E^*R'^2}$, respectively. Thus, using this expression, it is easy to predict friction coefficients at different conditions or estimating the lubricant viscosity required to achieve certain values of friction. This expression is of high relevance for biolubrication because most popular setups to mimic physiological conditions consist of compliant elliptic contacts. Lubrication forces described in Eq. (6) arise from macroscopically nonconforming surfaces and does not include any topographic detail at microscopic level, that is, asperities. Then, an unresolved research challenge is using Eq. (5) to recreate a scenario that includes the influence of papillae in the mixed and hydrodynamic lubrication regime, similarly to the rheological description provided by Eqs. (1) and (2). The application of the Reynolds equation in the mixed lubrication regime is not straightforward, because the lubricant film is a continuum but such continuity might be broken by the papillae/palate asperity contact in an oral surface contact model. Special boundary conditions are needed to describe the fluid discontinuity at the solid contact. Such a model would be useful to predict impact of viscosity in a realistic oral environment.

Complex fluids exhibit non-Newtonian behavior, and they are complicated to describe because they might present properties, such as yield stress, shear thinning, or shear thickening. Nevertheless, under the appropriate modification, generalized Reynolds equations are capable to describe non-Newtonian fluids [68]. Alternatively, finite element methods for fluid mechanics are excellent tools to simulate the flow of non-Newtonian fluids in general conditions. However, these calculations use the macroscopic rheological properties of complex fluids, which might not be representative under high degree of confinement as discussed previously for polymer solutions. Differences between Newtonian and complex fluids are evident when their lubrication performance is represented in the form of a Stribeck curve as those presented in Figure 3a and b. The mixed and hydrodynamic lubrication regimes of Newtonian fluids lubricating surfaces are represented by a single master curve when friction coefficient is plotted against the product of Newtonian viscosity and entrainment speed. This is due to hydrodynamic forces been controlled by the shear rate independent viscosity of the fluid. The boundary region might show subtle differences due to adhesion properties of the fluid, such in the case of corn syrup solutions [69]. However, in the case of non-Newtonian complex fluids, the scenario is slightly different. de Vicente et al. [67] studied the soft lubrication of xanthan gum and carbopol solutions. In the case of xanthan gum, a master Stribeck curve representing the mixed lubrication regimes was constructed using the high shear rheology viscosity of the material; nevertheless, the boundary lubrication friction was still dependent on polysaccharide concentration because this is controlled by hydrated layer absorbed into the surfaces [48]. Similar observation was obtained for other non-Newtonian hydrogels, such as κ-carrageenan with or without alginates or locust bean gums, where the master curve was not suitable to represent the boundary friction coefficient [28]. Bearing this in mind, the hydrodynamic lubrication regime of polysaccharide solutions such as xanthan gum can be described by Eq. (6) by using the high shear rate viscosity value as the viscosity constant. However, this approximation might not be valid for other lubricants such as particle suspensions [28]. To understand the lubrication of complex fluids, it is important to relate it to their microstructures because their thin film mechanical response might differ significantly from their macroscopic response.

Oral lubrication by saliva

Salivary lubrication has been extensively studied *ex vivo* in enamel contacts, and the effect of its components has also been assessed separately [54,70,71]. Between hard contact surfaces, human saliva has shown to reduce the friction coefficient by a factor of 20 [72], having friction coefficient of $\mu \approx 0.02$, that is, two orders of magnitude lower than that of water [62]. Vinke et al. [7] performed tribological experiments in the pig tongue/enamel contact finding that human saliva reduced the friction coefficient from 1.87 (dry contact) to 0.5 (after addition of saliva). Figure 3b presents the lubrication data of fresh saliva obtained from one healthy donor (Ethics MEEC-16-046) from our laboratory followed by centrifugation. Tribological experiments were performed in a

hydrophobic PDMS ball-on-disc setup at constant load and temperature of 2 N and 37 °C, respectively. In comparison to the lubrication provided by phosphate buffer (pH = 7.0) (Figure 3b), saliva reduces the boundary friction coefficient by one order of magnitude. A similar trend was obtained in the mixed lubrication regime. Although saliva plays such a crucial role, most oral tribological work do not consider saliva or salivary substitutes in the *in vitro* experimental setup until recently [27,28,53,73,74]. This is partly due to the unavailability of consensus on one standard artificial saliva formulation that replicates the lubrication behavior of saliva and partly due to the variability of lubrication behavior of real human saliva. For example, Bongaerts et al. [62] compared the tribological performance of fresh and air-dried whole human saliva in soft contacts. They found that dried saliva has boundary friction coefficients around 2.0, whereas fresh saliva lowered the friction of the soft contacts by an order of magnitude lower than that of the air dried counterpart. In addition, lubrication property of saliva can alter quite dramatically when it is stimulated (*e.g.* mechanical chewing or citric acid) versus when it is unstimulated, with high coefficients of friction in the former [75]. Hence, the conditions in which the saliva is donated and used for *ex vivo* lubrication experiments is extremely critical in understanding the results.

In the case of soft tongue surface, mucins, such as MUC5B, is mostly considered as the candidate molecule for the formation of the lubricating layer, whereas membrane-bound MUC1 enhances the anchoring of salivary MUC5B to the oral epithelial cells [76]. Adsorption of the macromolecules in saliva, such as glycosylated mucins [4,77−79], statherins [7], and acidic proline-rich protein 1 (PRP-1) [80] has also been often postulated to be responsible for creation of salivary pellicle contributing to aqueous boundary lubrication (Figure 3a). However, the true mechanism by which saliva lubricates is still under research as neither of these highly glycosylated mucin proteins alone or in simple combinations with other proteins have replicated the lubrication property of saliva to date. The role of hydration in the lubrication performance of saliva is also evident in the perception of 'astringency'. Poor lubrication perceived as a dry or 'astringent' mouthfeel when drinking tea, wine, or coffee is related to the failure of hydration lubrication due to the complexation of salivary mucins with polyphenol molecules in the beverages [15,26,81] and thus depletion of these hydrated mucins from the mucosal surfaces. Further studies are necessary to understand the structural mechanism of salivary lubrication and to use ideal protein−protein or protein−polysaccharide pairs to create salivary substitutes for *in vitro* oral tribological tests. Such knowledge is important not only to understand mouthfeel of food by quantifying food−saliva interactions but also to create therapies for dry mouth patients [21].

Oral lubrication by soft microgels

Soft microgels are swollen biopolymeric spherical particles with size ranging from 100 nm to 100 μm, which represent a viable model system to study lubrication properties of food particles. Depending upon the composition and structure of the microgels, they can swell or degrade when exposed to temperature, pH, ionic strength, and enzymes that may be encountered in oral cavity. The interest on microgel dispersions as biocompatible lubricants has increased dramatically in the last decade. This is because of their ability to generate higher degree of film thickness with complex flow behaviour that prevail during oral processing of food, potentially contributing to desirable mouthfeel and texture attributes [52,82−84]. Examples of soft lubrication of whey protein−based, starch-based, and emulsified lipid−based microgels are presented in Figure 3b. Recently, we studied the lubrication of whey protein microgels [52] in our laboratory using hydrophobic PDMS contact surfaces (water contact angle of 108°) demonstrating their capacity to reduce friction in the boundary regime. Particularly, such reduction in friction coefficients was evident only when the volume fraction (ϕ) of whey protein microgels exceeded 65 vol% reaching very low μ values (ca. 0.08) compared with μ ≈ 0.5 at lower ϕ (10−60 vol %). The friction reduction was attributed to an aqueous ball-bearing mechanism of submicron sized microgels (∼380 nm) that were entrained into the contact even at low working speeds. The PDMS surface had a R_a of 50 nm, and the particles were proposed to act as third body filling the gap between the asperities and result in a rolling motion, that is, acting as submicron scale "ball bearings", also previously reported for whey protein microparticles [85]. Similar results have been also shown in agar-based [82] and κ-carrageenan−based [83] microgels, where friction coefficients of the microgels in the boundary, mixed, and hydrodynamic regime were lower than that of the continuous phase, indicating that the entrainment of particles took place irrespective of the entrainment speeds. However, for agarose-based microgels, which were larger in size (83−106 μm) [84], the presence of particles did not reduce friction. This is because the entrainment of these micron-sized particles was restricted until the speed increased to give access to the particles to approach the gap between the two surfaces.

Recently, Torres et al. [53] studied the lubrication performance of starch microgels with or without containing emulsified lipid droplets in our laboratory. As it can be observed in Figure 3b, the Stribeck curve obtained for starch microgels showed an interesting resemblance to the performance of whey protein microgels, despite their difference in size and composition. Using Eqs. (3) And (4), we calculated the average force that microgels in the mechanical contact should bear to obtain the reduction of friction in the boundary

lubrication regime presented in Figure 3b. The indentation of particles relative to their size ($\frac{\delta}{R}$) under the mechanical load was calculated using a nonlinear model, as expressed in Eq. (7):

$$\frac{\delta}{R} = \left(\frac{a_H}{R}\right)^2 - \frac{4}{3\pi(1-v^2)}\left(\frac{a_H}{R}\right)^3 f\left(\frac{a_H}{R}\right) \quad (7)$$

where,

$$f\left(\frac{a_H}{R}\right) = \frac{2(1+v)}{\left(4+\left(\frac{a_H}{R}\right)^2\right)^{3/2}} + \frac{(1-v^2)}{\left(4+\left(\frac{a_H}{R}\right)^2\right)^{1/2}}, \quad (8)$$

and

$$\frac{a_H}{R} = \left(\frac{3W_L}{4\phi_p E^* a_{TP}^2}\right)^{1/3} \quad (9)$$

Here, W_L represents the load supported by all particles in the contact, R and a_H are the radius and contact radius of microgel particles, respectively, a_{TP} is the tribopair surfaces contact radius, and ϕ_P is the particle concentration. In addition, we calculated the hydrodynamic drag to estimate the size of particles that could be entrained into the contact by the hydrodynamic flow. Based on calculations of particle level elastic deformation and hydrodynamic forces, Torres et al. [53] attributed the lubrication of microgels to the rupture of these microgels releasing some of the emulsified lipid droplets under highly confined shear. Microgel particles were not capable to support the load and were almost lubricating like solid grease. On the other hand, higher elastic modulus of emulsion microgels ($\sim 20-30$ μm size) allowed these particles to be entrained inside the contact breaking them down to the level of emulsion droplet (0.08 μm size), supporting partially the contact load decreasing the friction coefficient to a higher extent (Figure 3b). Although the procedure described previously was developed to interpret experimental data obtained during *in vitro* tests, one might consider an intelligent modification in the theoretical calculation to extrapolate the lubrication performance of these microgels in real oral conditions. In such a case, the ratio a_H/R can be expressed in terms of the maximum Hertz pressure P_{max} as $\frac{a_H}{R} = \frac{\pi P_{max}}{2E^*}$. Considering the reduced elastic modulus of the tongue (2.5 kPa)/microgel (10 kPa) contact is about 4.5 kPa and it is subjected to a maximum pressure of 50 kPa, a value for a_H/R about 17.0 is obtained, and therefore, the calculated value for the dimensionless indentation δ/R is about 170.0. Because particles are relatively harder than the soft tissue, calculations predict a scenario where microgel particles would be potentially pushed into the tongue tissue incapable to sustain the load. However, Eq. (7) is mostly appropriate to describe a contact with surface roughness smaller than the average particle size. Noteworthy, tongue tissue containing filiform and fungiform papillae would produce nonuniform pressure distribution and gaps where microgel particle could accommodate without being squashed or immerse completely into the tissue. Following Eqs. (1) and (2), it is, therefore, important to develop new models to describe the role of papillae in the tribological limit and their interaction with exogenous lubricants, such as colloidal suspensions.

Future perspectives on synthetic materials to emulate oral surfaces

A key challenge to perform oral lubrication measurements is that the targeted oral modulus and surface chemistry offer a rather narrow window for the choice of soft elastomeric materials to effectively emulate tongue surfaces. We discuss few of the several challenges on tribopairs that needs to be addressed in oral lubrication research.

Material physics

Friction responses are highly system-dependent (both tribo-contact surfaces, load, lubricant used), and hence, there is absolute need for development of accurately tongue—oral palate mimicking soft contact surfaces with tailored topologies to do oral tribology experiments. Furthermore, these synthetic materials should be able to emulate time-dependent and shear—responsive properties of the oral surfaces so that they can interact with the oral environment (pH, ions, enzymes, temperature) in a similar fashion. A palette of materials from metal [2], rubber [86], ceramics [1], PDMS [52], pig oral tissue [34] to synthetic enamel [87] have been used to emulate oral surfaces to do oral lubrication measurements of soft surfaces. Unfortunately, such measurements suffer from obvious pitfalls as the aforementioned surfaces used in typical mechanical engineering context do not necessarily represent the biotribo surfaces (soft, slippery mucous-coated papillary surface of a human tongue). Naturally, one might consider the best substrate would be pig tongues, which are considered to have similar modulus and surface characteristics of the human tongue [24]. Few *ex vivo* tribological experiments performed using animal tissue to simulate the physical properties of soft oral contacts can be found in literature [7,24]. Recently, Vinke et al. [7] designed a setup using pig's tongue to study the efficiency of dry mouth—relieving agents. Their results showed that relevant correlations could be established using biological tissue instead of synthetic surfaces showing the need to improve the existing *in vitro* tribopairs. However, it is challenging to use it as a standard because of its variability and inhomogeneity, relative fast degradation and dehydration of the tissue, as well as having unknown surface chemistry.

Instead, PDMS) has been used as a gold-standard material for designing tribopairs for oral lubrication in today's tribological measurements, considering it is several orders of magnitude lower in modulus as compared with traditionally used steel-based hard mechanical contact alternative. Moreover, PDMS modulus offer three orders of magnitude range of tenability in modulus anywhere from 5 kPa up to 2 MPa just by varying the degree of cross-linking [88]. In addition, the reason for using PDMS is its optical clarity for doing microscopy of the tribologically sheared samples. However, PDMS surface used in literature so far has been mostly used with MPa range modulus and even with reduced degree of crosslinking (~ 0.65 MPa) that has two orders of magnitude higher modulus than that of the human tongue [24]. Although these state-of-the-art studies are advancing significantly, future studies should use PDMS of lower degree of cross-linking to emulate soft tongue surfaces better. Here, we summarize mechanical properties of a range of polymeric materials including hydrogel-based surfaces (Figure 4) that might see the light in oral lubrication domain in future [89–96]. In Figure 4, reduced Young modulus (x-axis) and reduced tensile strength (y-axis) represent dimensionless mechanical properties of materials obtained as the ratio of the absolute property values to a characteristic stress values related to the mechanical performance of

the oral cavity. On one hand, the reduced tensile strength is the ratio of the material elastic modulus to the maximum Kokini shear stress, that is, about 45 Pa. Reduced tensile strength is an indication of the capacity of the material to endure the stress generated during oral processing. On the other hand, the reduced Young modulus is calculated as the ratio between the material tensile strength and the maximum normal pressure, that is, about 50 kPa found in normal human oral conditions. The reduced Young modulus can be thus identified as an indicator of level of deformation of elastic deformation of the material under oral conditions. Hydrogels, such as collagen–polyethylene glycol, gelatin–alginate, gelatin–methacrylamide, alginate–polynipam, and alginate–cellulose, are currently being used in biological applications, such as scaffolding for cell culture due to their biological compatibility, hydrophilicity, and mechanical properties. Apart from alginate–cellulose composites, the rest of materials appear suitable for the oral working conditions. Reduced Young modulus of tongue is about 0.1, a value achievable also for some hydrogel composites such as gelatin–methacrylamide. Capacity of these materials to endure shear and compression can be also improved by external manipulation. For reduced Young modulus values beyond 10, the oral pressure might not be enough to cause significant deformation that could influence the lubrication. Pressure values commonly used in oral mimicking conditions with PDMS surfaces (cross-linked with MPa range modulus) are about five-times larger than the oral/palate pressure, imposing conditions on the lubricant that might cause deviations from its behavior during the normal oral processing. Another possible way to obtain mechanical properties to emulate oral conditions is by creating structures including feature that can be externally manipulated. For instance, the creation of a model tongue with air channels can be useful to manipulate shape, modulus, and capability to resist deformation without plastic behavior [97].

Another important feature, which is underestimated, is the vital role of surface roughness. The PDMS surface used currently has nanometer scale surface similar to steel, which is several orders of magnitude finer than human tongue surface as discussed before. Sandblasting has been also used to create PDMS surfaces with roughness (Root Mean Square [RMS] of 3.6 μm and peak valley heights of 17 μm) [107], which is still one order of magnitude lower than the human tongue. Interestingly, one approach has been the use of surgical tape with known surface roughness and well depth (R_a = 31.5 μm, well depth of 170 μm, respectively) to simulate human tongue roughness [108]. In future, colloid scientists have excellent opportunities to design surfaces using fast developing microfabrication strategies [109], such as 3D printing technology with targeted microasperities to effectively emulate oral surfaces. Lithographic and molding techniques can be applied on

Figure 4
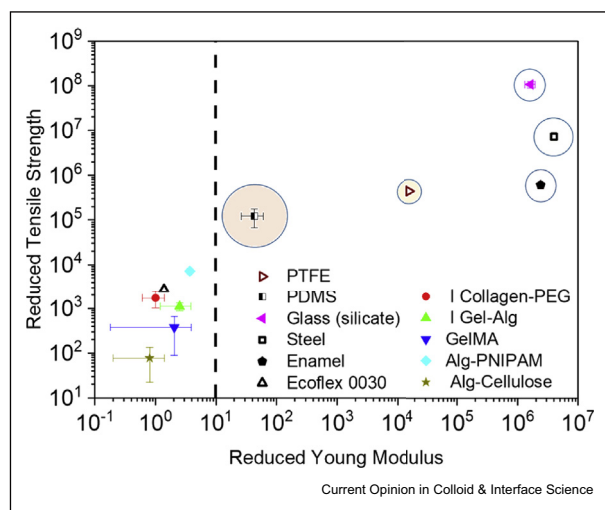
Mechanical properties of materials used in biotribological experiments and hydrogels used in other biological applications. The reduced tensile strength and the reduced Young modulus are dimensionless quantities obtained by normalizing absolute values of tensile strength and elastic using the maximum Kokini shear stress (45 Pa) and the maximum normal pressure (50 kPa) in human oral conditions, respectively. The left side of the line contains the materials that could approximate the mechanical response of the human tongue. Size of circles corresponds to the number of articles available in literature that use these materials [24,27,48,67,71,98–106] with PDMS being the main material used in biotribological studies currently.

these types of materials to create surfaces with the appropriate topography, such as papillae pillars.

However, in all of these traditional sandblasting and newer 3D printing technologies, a delicate interplay between Young's modulus, tensile properties (Figure 4), and surface roughness of the tribopairs is critical so that the tribopairs deform somewhat elastically similar to human tongue surfaces when subjected to normal loads. In addition, an average roughness parameter might not be representative of the mechanoreceptive properties of the tongue. Particular structures such as filiform (crown-like) and fungiform (mushroom-like) papillae shape need to be included to improve correlations between *in vitro* tribological tests and sensory studies. Such features might be obtainable using materials such as hydrogels using techniques developed for tissue engineering applications. Finally, conflation with robotics [110,111] can be particularly groundbreaking to effectively emulate locomotion of tongue and its consequences on frictional forces.

Surface chemistry

Another key issue that may be appreciated for most polymeric materials used for oral tribology experiments is their surface hydrophobicity that differs from hydrophilic saliva-coated tongue surfaces in reality. Functionalization of surfaces with hydrophilic brush polymers, oxygenated species, mucins, and salivary films [77,112,113] can be useful to generate appropriate wettability. For example, surface modifications of PDMS, such as O_2-plasma treatment, can be used to change the wettability of the tribopairs [52]. However, key shortcoming of these methods is the rather transient effect and rapid recovery of hydrophobicity unless the PDMS substrates are kept dispersed under distilled water [114]. One might also approach coating the polymeric surfaces with human saliva before doing tribological measurements. However, Bongaerts et al. [62] also pointed out that saliva-coated substrates are prone to degradation, dehydration, and have properties dependent on the preparation method and show large variability. Therefore, building a more appropriate artificial salivary pellicle is key to study the dynamics of oral tribology and consequently predict sensory mouthfeel, as discussed previously. One approach used recently in our laboratory was O_2-plasma treatment of PDMS surfaces followed by coating of PDMS with purified mucin, and it was demonstrated the hydrophilicity of the tribopairs was restored over weeks (water contact angle 47°) [52]. Although wettability of soft oral surfaces by salivary film is a key feature that might contribute to oral lubrication, experiments should be carried out both in the absence and presence of preadsorbed real or artificial saliva films. This will help to understand the mechanisms behind oral lubrication, whether it is driven by the food particle, saliva, or food particle–saliva mixture dominant film.

Conclusions

Understanding of oral lubrication is crucial to decode mechanosensing during eating and to underpin future design of biophysically informed food with tailored mouthfeel properties, mucosal lubricants, and pharmaceuticals. To build a robust machinery to test and design products with tailored lubricating properties in oral conditions, it is necessary to emulate the highly sophisticated oral mucosal surfaces engineered by the nature with delicate balance between modulus, tensile, and surface roughness of the tribo-testing materials together with appropriate working conditions. The use of soft surfaces, such as hydrophobic or hydrophilic PDMS, has commenced to demonstrate promising capabilities of tribological techniques and is already providing mechanistic evidence behind some of the textural attributes of fat and nonfat food products. Besides mechanical compliance, oral surfaces have topographic and chemical properties that influence the oral lubrication process. The role of filiform papillae as a mechanoreceptor has been theoretically explored recently and deserves more attention in future research. Surface wettability is of paramount importance to understand the interaction between endogenous saliva and/or exogenous lubricants and oral surfaces. A clear example is hydration lubrication, which might be the key mechanism for tribological performance of a range of food particles in the oral cavity that requires research attention. To explore these properties, systems must be designed with the right microstructure containing macromolecules to promote the formation of hydrated structures and/or provide a synergistic hydration effect in combination with salivary components. We believe that hydration lubrication using proteins, protein–polysaccharide with appropriate charge distribution, and polar head groups might capture more research attention in oral tribology and unlock fat reduction/replacement possibilities as well allow rational designing of dry mouth therapies. The experimental representation of different flow fields other than sliding, such as biaxial compression and uniaxial extensional flow, is a pending task. Subjecting complex fluids to different flow fields confined in-between bioinspired surfaces might empower experimental techniques to strengthen correlation between mechanical testing and oral perception. Finally, the development of theoretical tools to analyze oral mimicking tribological experiments is a pending and necessary task that cannot be underestimated. For instance, the behavior of complex fluids such as microgels in the thin film approximation and their interaction with saliva is still an open challenge. These research challenges might be addressed by combining micromechanical deformation models together with continuum fluid mechanics methods.

Conflicts of interest

The authors declare no competing financial interest.

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