



Research Paper

Measurement of molten chocolate friction under simulated tongue-palate kinematics: Effect of cocoa solids content and aeration



Georgios Samaras^{a,*}, Dimitrios Bikos^a, Josélio Vieira^b, Christoph Hartmann^c, Maria Charalambides^a, Yannis Hardalupas^a, Marc Masen^a, Philippa Cann^a

^a Department of Mechanical Engineering, Imperial College London, London SW7 2AZ, United Kingdom

^b Nestlé Product Technology Centre York, Nestlé, York, United Kingdom

^c Nestlé Research Centre, Vers Chez Les Blancs, CH-1000 Lausanne 26, Switzerland

ARTICLE INFO

Keywords:

Friction
Food oral processing
Artificial saliva
Soft tribology
Friction coefficient

ABSTRACT

The perception of some food attributes is related to mechanical stimulation and friction experienced in the tongue-palate contact during mastication. This paper reports a new bench test to measure friction in the simulated tongue-palate contact. The test consists of a flat PDMS disk, representing the tongue loaded and reciprocating against a stationary lower glass surface representing the palate. The test was applied to molten chocolate samples with and without artificial saliva. Friction was measured over the first few rubbing cycles, simulating mechanical degradation of chocolate in the tongue-palate region. The effects of chocolate composition (cocoa solids content ranging between 28 wt% and 85 wt%) and structure (micro-aeration/non-aeration 0–15 vol%) were studied. The bench test clearly differentiates between the various chocolate samples. The coefficient of friction increases with cocoa solids percentage and decreases with increasing micro-aeration level. The presence of artificial saliva in the contact reduced the friction for all chocolate samples, however the relative ranking remained the same.

1. Introduction

Food mastication is a complex dynamic procedure that involves multiple oral operations resulting in the breakdown of food in the mouth until swallowing. Throughout the food consumption, various interactions of the oral cavity, including the tooth-tooth and the tongue-palate contacts take part in the process. During mastication the structure of the foods changes and different components contribute to the texture and taste perception (Liu et al., 2017). Texture perception, which includes aspects such as smoothness and slipperiness, is usually related to mechanical stimulation and hence friction experienced in the mouth (Stri-
bić et al., 2020).

Food texture is considered a multidimensional tactile sensation that is influenced by the composition, structure, rheology and surface properties of the food as it is sheared between the tongue and palate. Extensive research has already been conducted into the bulk properties of foods and the relationship to oral texture, mainly through rheological studies (Janssen et al., 2007), (de Wijk et al., 2003). However, this method does not quantify the mouth-feel sensation, which is mainly correlated with the thin film formation in the tongue palate contact

zone (Malone et al., 2003). According to Dresselhuys et al., 2008a,b) ‘an aspect that is not measured by rheology, but is probably sensorial relevant, is the process of rubbing and squeezing the product between tongue and palate during which the tongue and palate surfaces are in partial contact. In this process a frictional force is generated between palate and tongue, with the semi-solid food acting as a lubricant’. Moreover, de Wijk et al. (De Wijk et al., 2011) suggested a link between the type of sensation and the time point processing in mouth, stating there is a transition from rheological to tribological properties depending on the oral processing time and the length scale of the food particles in the mouth.

Tribology, which is the science of wear, friction and lubrication, can be applied to food science and especially in the study of thin film formation between the tongue and palate. A review of oral tribology by Chen and Engelen (Chen and Engelen, 2012) provides valuable background on tribology fundamentals in food science. Chen and Stokes (Chen and Stokes, 2012) suggest tribology to be a contributing discipline for understanding oral processing, texture and mouthfeel of food. In the study of Pradal and Stokes (Pradal and Stokes, 2016) the importance of soft tribology as a characterization tool for sensory attributes is also highlighted. Sarkar and Krop (Sarkar and Krop, 2019) review the applications of oral tribology in

* Corresponding author.

E-mail address: georgios.samaras17@imperial.ac.uk (G. Samaras).

modelling food structures through correlating the coefficient of friction with sensory attributes. Thus, over the past few years, there has been increased research in the correlation between tribology and sensory perception. There is an increased interest from the food and beverage industry regarding tribology applications, since it is believed that the consumer's experience is influenced by the thin layer formation on the mouth surfaces. [Prakash et al. \(2013\)](#) review progress made in food tribology with a detailed presentation on how tribometers are modified and used to characterise the textural sensory properties of foods.

In this work, chocolate was studied as an example of a food where the consumer experience is influenced by the oral perception of creaminess and smoothness ([Afoakwa et al., 2007](#)). The global market for chocolate was valued at USD 103.28 billion in 2017 and is expected to grow 7% in revenue by 2024. However, there is considerable interest in the food industry in developing foods with reduced energy density, in the case of chocolate meaning less fat and sugar, but with the same mouthfeel. There are very few published papers reporting chocolate tribology ([Lee et al., 2002](#)), ([Lee et al., 2004](#)), ([Masen and Cann, 2018](#)), ([Rodrigues et al., 2017](#)), ([Breslin, 2013](#)), ([van Aken et al., 2007](#)) and even fewer relating friction properties to sensory perception ([Lee et al., 2004](#)), ([Masen and Cann, 2018](#)). A range of tribology test methods have been used including ball-on-flat ([Lee et al., 2002](#)), ([Breslin, 2013](#)), ([van Aken et al., 2007](#)) and modified rotational viscometer ([Lee et al., 2004](#)). The studies usually compare friction/speed response for different chocolate manufacturing methods ([Lee et al., 2004](#)) or composition ([Lee et al., 2004](#)), ([Masen and Cann, 2018](#)). In consumer sensory tests of chocolate texture, the smoothness is usually identified as a key characteristic ([Lee et al., 2004](#)) and might be linked to reduced friction in the low speed boundary regime. Friction coefficient varies with sliding speed ([Lee et al., 2002](#)), ([Lee et al., 2004](#)), ([Masen and Cann, 2018](#)), ([van Aken et al., 2007](#)) and is in the range $\mu = 0.1$ – 0.6 depending on chocolate composition. Masen and Cann ([Masen and Cann, 2018](#)) used a reciprocating contact to measure friction with time of shear-degraded chocolate and reported friction coefficient range of $\mu = 0.18$ – 0.32 at 30 mm s^{-1} depending on composition. In tests where human saliva was present friction drops to $\mu = 0.06$ – 0.09 ([Rodrigues et al., 2017](#)).

The cocoa solids content is thought to play a significant role in the oral friction experienced during mastication and this is studied in the paper. One possible way of maintaining or even improving mouthfeel may be the inclusion of micro-sized bubbles. In this paper, micro-aerated chocolate will be tested and compared with commercial non-aerated chocolate samples. The development of a validated tongue-palate simulated test to measure the friction response of rubbed food samples could possibly replace the panel tests, which are currently used by the food industry. As will be presented below, the design of such a test is difficult when based on traditional tribology machines.

One of the aims of this paper is to develop a new approach to designing tribology tests to measure friction in a simulated tongue-palate contact. This requires consideration of the “engineering” conditions in the mouth and the artificial models for biological tissue (tongue, palate, saliva), which will be used as test specimens.

The tongue palate environment is crucial for the perception of taste and texture. The palate is a relatively smooth and hard surface that does not have taste receptors and is used mainly to spread the already partially processed food from the teeth to the larger area in the mouth ([Breslin, 2013](#)). From there the food reaches the tongue where the receptors are placed that are responsible for the perception of different tastes and textures. The human tongue is a rough and flexible muscle and its surface is covered by conical papillae of order $100 \text{ }\mu\text{m}$ in height ([van Aken et al., 2007](#)). Almost two-thirds of the human tongue is covered by filiform papillae that do not contribute to the taste perception but are sensitive to friction. Taste receptors are located at the fungiform and foliate papillae ([Sarkar et al., 2019a,b](#)).

The tongue-palate contact is lubricated by saliva and according to Humphrey and Williamson ([Humphrey and Williamson, 2001](#)) this plays a crucial role in the following processes: (1) lubrication and protection,

(2) buffering action and clearance, (3) maintenance of tooth integrity, (4) antibacterial activity, and (5) taste and digestion. Saliva is a complex biological fluid which is mainly water ($\sim 99\%$) but also contains a mixture of mucins, glyco-proteins, electrolytes and enzymes with a pH of 6–7 so that it is slightly acidic ([Humphrey and Williamson, 2001](#)). Lubrication is provided through the adsorption of saliva proteins, such as mucin, which account for 16% of the total proteins ([Rayment et al., 2000](#)), onto the mouth surfaces ([Shi and Caldwell, 2000](#)), ([Hahn Berg et al., 2003](#)).

Saliva is involved in the perception of taste flavour and texture of foods and serves as a lubricant reducing the coefficient of friction and the shear stress on the surface of the tongue. Recent work by Upadhyay and Chen ([Upadhyay and Chen, 2019](#)) highlights the importance of the addition of saliva in tribology characterizations. [He et al. \(2018\)](#) investigated the reduction of friction due to the presence of saliva, studying chocolate boluses composed of chocolate and human whole saliva. In order to have consistent results, in this study a simple artificial saliva recipe was used ([Ionta & et al., 2014](#)).

Eating starts with the first bite, where the food is mechanically broken down into smaller pieces. After a few chew cycles, while heat transfer may control the melting of fragments, the food enters the tongue-palate area, where it is sheared and forms a bolus, before swallowing. When designing simulation tests the mechanical conditions occurring during the oral process must be considered. These include contact pressures, kinematics and the appropriate time-scale for friction measurements. The mechanical properties of the tongue change depending on the orientation of the muscle fibres ([Napadow et al., 1999](#)), resulting in varying contact pressures for different relative tongue-palate positions. [Nishinari et al. \(2020\)](#) measured the average maximum isometric tongue pressure, during food oral breaking, to be $50 \pm 14 \text{ kPa}$ which agrees with the findings published in the study of Alsanei et al. ([Alsanei and Chen, 2014](#)) who reported maximum isometric pressure ranging between 10 kPa to 70 kPa .

The relative speed between tongue and palate is another important physical factor related to mechanical processing. Hiimae and Palmer ([Hiimae and Palmer, 2003](#)) suggested a speed range between 2 and 35 mm s^{-1} depending on parameters, such as the stage of food oral processing, different speeds correspond to biting and to swallowing. The shear rate in the tongue palate region during mastication is a parameter that should be taken into consideration. Several studies have attempted to quantify the shear rates that occur in the tongue-palate interaction, and reported values range between 10 s^{-1} ([Cutler et al., 1983](#)) to 50 s^{-1} ([Akhtar et al., 2005](#)).

In mouth temperature is also important for the accurate design of the experiment. The values used in the literature vary depending on the stage of processing and the what is consumed. The reported values range between room temperature ([Dresselhuys et al., 2008a,b](#)) and $40 \text{ }^{\circ}\text{C}$ ([Lee et al., 2004](#)). In this study the lower specimen is heated at $35 \text{ }^{\circ}\text{C}$ and the chocolate is allowed to melt before the start of the test. This value is believed to be representative as we expect to be less than $36.6 \text{ }^{\circ}\text{C}$, but certainly higher than the room temperature. A summary of the mechanical conditions in the tongue-palate contact during food processing is provided in [Table 1](#).

Several types of equipment have been used to measure friction in applications related to oral processing. [Prakash et al. \(2013\)](#) provided a detailed overview of the various tribological instruments that have been used in food studies. Configurations used include pin-on-flat,

Table 1
Summary of the mechanical conditions in the tongue-palate contact.

Parameter	Range Value	Test settings
Contact Pressure	3–36 kPa	35 kPa
Speed range	2–35 mm s^{-1}	120 mm s^{-1}
Time scale	5 s	5 s
Temperature	35 $^{\circ}\text{C}$	35 $^{\circ}\text{C}$

ball-on-flat and flat-on-flat (Prakash et al., 2013), (Chen et al., 2014), (Joyner Melito et al., 2014). The most common technique used both in tribological and food science is the MTM tribometer (Shewan et al., 2019), which consists of a ball in combined rolling and sliding motion against a rotating disk. The MTM test is usually employed to measure friction as a function of the changing speed (increasing or decreasing) to generate a Stribeck-type curve (Sarkar and Krop, 2019). It measures a single friction value, averaged over several seconds, which is a relatively long time-scale compared to mastication times, which for chocolate range between 15 and 20 s (Carvalho-Da-Silva et al., 2013). Thus, these tests cannot be used to follow friction changes occurring during the first few seconds of mechanical breakdown in the tongue-palate region. In addition, the traditional test design requires a continued supply of fluid which is entrained through the inlet of the contact. Under these conditions, fluids experience shear rates of the order 10^5 – 10^7 s⁻¹ in the inlet and contact regions, which is significantly higher than those reported for the tongue-palate contact region (~ 50 – 100 s⁻¹) (Akhtar et al., 2005), and thus complex fluids may shear-degrade or suffer phase-changes. Thus, the composition of the film in the contact zone might differ from the average bulk (Myant et al., 2012). To measure the mechanical breakdown of food over time, a single sample must be used which is subject to repeated rubbing, while reducing the effects of inlet shear.

Traditional tribometers that use a steel sample pair are not compatible with oral processing studies, since the pressure between the two surfaces is much higher than desired (Scholten, 2017). For this reason, tribological set-ups for food research use a combination of soft and hard surfaces in order to lower the contact pressures. Some of the most common soft surfaces used are teflon (Chojnicka-Paszun and De Jongh, 2014), polydimethylsiloxane (PDMS) (Bongaerts et al., 2007) and rubber, and there are studies which used pig's tissue as a tongue substrate (Chen et al., 2014), (Joyner Melito et al., 2014), (Dresselhuys et al., 2008a,b). The ideal material for lubrication studies would be the one that imitates the mechanical and chemical properties of the human palate and tongue (Ranc et al., 2006). PDMS is currently used as model tongue surface material for many food tribology studies (Sarkar et al., 2019a,b), although there are significant differences in the mechanical and chemical properties compared to the human tissue. The Young's modulus of PDMS ranges between 1–3 MPa (Payan and Perrier, 1997) which is higher than the human tongue (46–150 kPa) (Sarkar et al., 2019a,b). Despite this difference it is usual to use a PDMS-glass combination as a soft-hard model of the tongue-palate (Devezeaux de Lavergne et al., 2016), (Liu et al., 2016), (Rovers et al., 2016). One significant difference between the human tongue and the PDMS samples used is the surface texture. In some studies, the tongue texture has been replicated using surface-moulded PDMS (Ranc et al., 2006) but this approach has not been adopted for the current work.

A new configuration is proposed, consisting of two flat surfaces to mimic the tongue-palate contact. The upper surface, a PDMS disk is loaded and reciprocates against the lower stationary surface, a glass microscope slide. This motion simulates the movement of the tongue against the palate (Liu et al., 2016). In addition, due to the combination of a large contact area and a relatively short stroke length, the food under test is essentially isolated and new material is not continuously entrained. When eating semi-solid food is introduced into the mouth as individual samples which are mechanically degraded before swallowing. There are already studies that use flat on flat configurations (Masen and Cann, 2018) to successfully distinguish between confectionery products with different cocoa solids. In this paper, the experimental setup presented by Masen and Cann (Masen and Cann, 2018) is further developed by using a new device that is able to operate at the appropriate kinematics, a lower reciprocating frequency and by including the effect of artificial saliva in the system whilst recording the friction forces and high temporal resolution. The test was used to assess friction for a range of chocolate compositions, since these are expected to show differences in oral perception.

2. Materials and methods

2.1. Tribology test device

The BTM (PCS Instruments, London) was used to measure the friction properties of lubricated and unlubricated contacts. In the setup, shown in Fig. 1a, a model tongue comprising a flat cylindrical PDMS specimen (5 mm height, 6 mm diameter) is loaded against a stationary glass slide representing the palate. A load of 1 N was used, giving an approximate contact pressure of 30 kPa, which is comparable to reported pressures in the tongue/palate contact. The 6 mm diameter PDMS cylindrical specimen was punched from a 5 mm thick silicone sheet (Silex UK) using a biopsy punch, and the surface texture was measured using a White Light Interferometer. The PDMS disc is glued on the upper holder and reciprocates against the glass slide, imitating the movement of the tongue. The glass slide is attached to a temperature-controlled metal base, where the temperature is monitored through a thermocouple. The glass slide is preheated to 35 °C for 2 min before the start of the test. After the 2 min of preheating, a small amount (~ 0.2 g) of shaved chocolate is placed on the glass slide and allowed to melt for 30 s. Then, the PDMS flat disc is brought into a reciprocating sliding contact with the glass slide, with a specified load, stroke length and frequency. The BTM was specifically designed to mimic the physiological in-mouth conditions (Table 1) with an applied load of 1 N, a reciprocating frequency of 1 Hz and a stroke length of 5 mm, resulting in a mid-stroke sliding speed of 120 mm s⁻¹. The test conditions are summarized in Table 1. Friction data was recorded over 5 cycles at a temporal resolution of 100 Hz, continuously over the reciprocation cycle and was directly available in LabVIEW. Mean values were calculated averaging the 15 middle points of each stroke and data was plotted. The error bars in the graphs represent the entire range (minimum and maximum) of all the results. Ten different chocolate specimens of each type were used to collect samples from five different areas of each specimen. The significant difference between the samples was validated using a one way Analysis of Variance (ANOVA) at 95% confidence interval with a p-value < 0.05.

2.2. Cleaning procedure of the surfaces

As surface contaminants and thin films affect friction and wetting properties, all surfaces were cleaned before the friction measurements. After each test the PDMS cylinder was wiped and cleaned with isopropanol and it was replaced every five tests. The glass slide was cleaned with isopropanol after every test. Care was taken when handling the surfaces in order to reduce the risk of further contamination. Immediately after drying of the specimens the experimental procedure was started.

2.3. Artificial saliva

Human saliva is a complex fluid that contains several proteins and the design of an artificial saliva substrate is not straightforward. In the recent paper of Sarkar et al., 2019a,b) the complexity of human saliva is presented and the difficulty of creating an exact human saliva substrate is also highlighted. In this study, in order to mainly focus on the material under test, a relatively simple artificial saliva recipe is used, consisting of 100 ml deionized water, 0.27 g mucin and half a tablet of phosphate buffered saline (Ionta & et al., 2014). The chemicals are mixed in a magnetic stirrer at room temperature, stored at 5 °C and discarded after seven days (Sumarokova & et al., 2018). The glass slide palate substrate was submerged in 10 ml of artificial saliva for 30 min, allowing the absorption of the proteins onto the glass slide. The glass slide was then carefully retracted leave a thin saliva film on the surface. It was then positioned on the heated base of the BTM and the chocolate sample was added. Table 2 summarizes the materials used in the experiment.

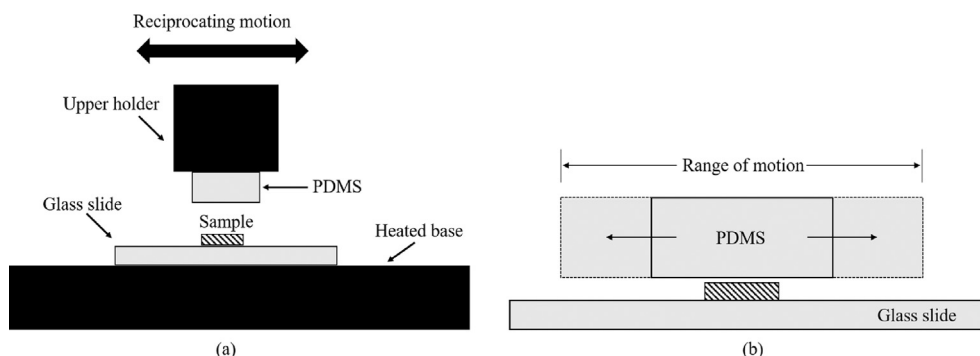


Fig. 1. (a) Schematic representation of BTM test configuration, (b) Close up of the interaction area between the PDMS and the sample, indicating the range of motion.

Table 2
Summary of materials used in the experiment.

Material	Description
PDMS	- Silex Silicone Youngs modulus 2 MPa (Sarkar et al., 2019a,b) 5 mm thickness, 6 mm diameter - Surface texture: array of dimples 10 μm deep, 0.4 mm diameter, 0.8 mm between dimples
Glass	S8400 Sigma microscope slide
Artificial saliva	100 ml deionized water 0.27 g mucin from porcine stomach Type II (M2378 Sigma) 0.5 PBS tablet (P4417 Sigma)

2.4. Chocolate specimens

Various commercially available chocolate specimens were selected to represent different levels of cocoa solids and aeration level. The commercial chocolate samples with different cocoa solids are all made by the same manufacturer with cocoa solid amounts ranging from 30 wt% to 80 wt%. Further details of the commercial chocolate can be found in Masen and Cann (Masen and Cann, 2018).

Nestlé provided both non-aerated samples and micro-aerated samples with 10 vol%, 12 vol% and 15 vol% aeration, with a bubble size of $\sim 50 \mu$ m. The chocolate was prepared as shavings, using a razor blade and 25 g was placed on the heated glass slide (either clean, or with an absorbed saliva film) and allowed to melt for 120 s.

Table 3 summarizes the composition of the chocolate samples. Scanning electron microscopy (SEM) images from a non-aerated and an aerated sample are depicted in Fig. 2. A Hitachi S-3400 SEM was used at variable pressure mode and at a relatively low accelerating voltage of 10 kV to prevent the samples from melting.

3. Results

3.1. Procedure for acquisition of experimental data

Friction tests were performed on the BTM using the configuration described above. Fig. 3a depicts a typical measurement of the friction force

Table 3
Composition of chocolate test samples.

Designation	Cocoa solid (wt%)	Fat content (wt%)	Sugar (wt%)	Aeration (vol%)
85% Dark Chocolate	85	32	14	0
70% Dark Chocolate	70	25	29	0
37% Milk Chocolate	37	20	46	0
28% Milk Chocolate	28	27	44	0
28% Milk Chocolate	28	27	44	10
28% Milk Chocolate	28	27	44	12
28% Milk Chocolate	28	27	44	15

during reciprocation. The positive and negative values correspond to left and right reciprocating motion of the upper holder, respectively. The data are processed, as shown in Fig. 3b-d. The first two strokes are eliminated from the data since the force becomes constant after the second stroke. The coefficient of friction is shown in Fig. 3b as the absolute values of the friction force divided by the applied load of 1 N. Data measured at the reciprocation points is ignored and only the part of the motion that corresponds to sliding is considered (Fig. 3c). Each sample was tested once and discarded after the end of the test. The bar chart of Fig. 3d can be calculated and used for comparison between the various conditions.

3.2. Repeatability of the friction results

Overall, the results are very repeatable, as shown in Fig. 4, where three different tests are superimposed for dry conditions, i.e. PDMS against the glass slide, and for lubricated conditions PDMS - glass with artificial saliva. The coefficient of friction for the artificial saliva was measured to be $\mu = 0.23$, which lies in good agreement with results reported in literature (Xu & et al., 2020).

3.3. Friction results for commercial chocolate samples with different cocoa solid content

Fig. 5a shows a comparison for the friction coefficient between the commercially available chocolate samples tested without artificial saliva present. Differences were observed depending on the cocoa solids content. The high content samples (~ 85 wt%) showed higher coefficient of friction during the first 5 strokes compared to the 70 wt% and 37 wt% cocoa solids samples. The tests were repeated with the artificial saliva and the results followed the same behaviour (Fig. 5b), but the values of the friction coefficient are, as expected, lower. Statistical analysis through ANOVA indicated a clear difference between the samples in both cases.

3.4. Aerated chocolate friction results

Fig. 6a compares averaged coefficient of friction without artificial saliva for chocolate samples with levels of micro-aeration of 0, 10, 12, and 15 vol%. The friction trace changes over the stroke length. It is at a minimum at the reciprocation point and increases to a maximum in the middle of the stroke. The maximum friction coefficient decreases with aeration level from $\mu = 0.4$ – 0.44 for the 0 vol% to $\mu = 0.2$ – 0.23 for the 15 vol% aeration.

Fig. 6b shows the results obtained for the various aerated chocolates with artificial saliva present. Again, differences were recorded depending on aeration level although overall the friction was much lower. The maximum friction coefficient decreases with aeration level from $\mu = 0.16$ – 0.175 for the 0 vol% to $\mu = 0.07$ – 0.08 for the 15 vol% aeration. Clear differences are observed in the friction coefficient values in both figures, which was also justified by the ANOVA analysis.

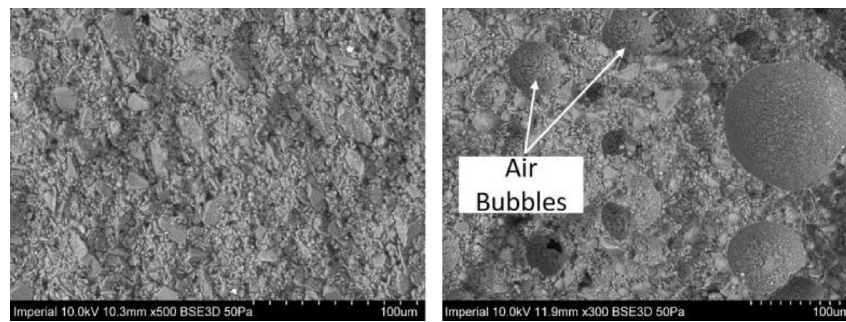


Fig. 2. SEM images from (a) non-aerated and (b) micro-aerated sample.

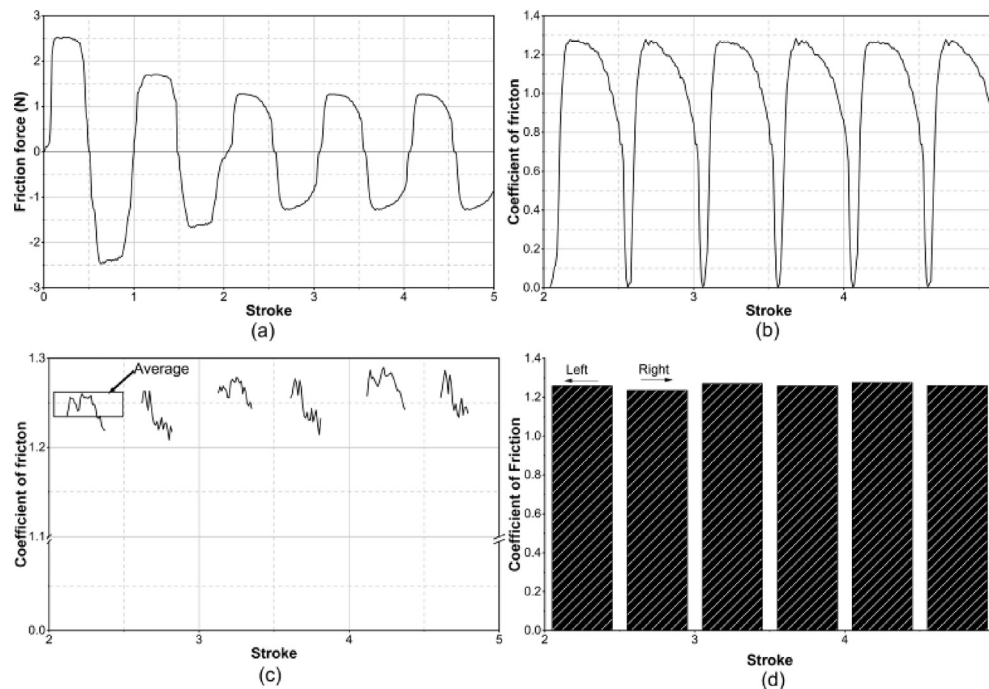


Fig. 3. Measurement of friction force during reciprocating motion. (a) Typical friction force measurement during reciprocation, (b) coefficient of friction corresponding the absolute values of friction force with elimination of first two strokes, (c) data corresponding to the sliding, (d) bar chart by averaging the 15 middle points of each stroke.

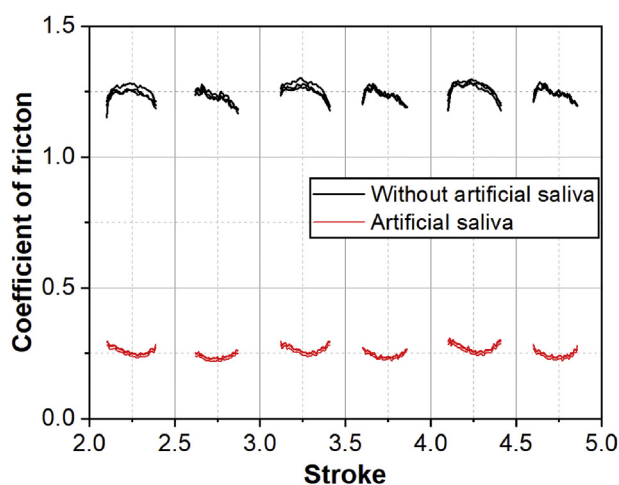


Fig. 4. Measurement of coefficient of friction as a function of reciprocating motion. Repeatability for dry conditions (without artificial saliva) and artificial saliva.

A major difference between the friction traces without (Fig. 6a) and with (Fig. 6b) artificial saliva is the shape of the curves. The results with artificial saliva are characterized by a convex curve, whereas non-lubricated conditions result in curves that are concave. For reference, Fig. 8 also shows coefficient of friction measurements obtained for lubricated sliding of the PDMS against glass, when no chocolate was present. This friction is significantly higher than when chocolate is present in the contact.

These results show that the proposed experimental setup can measure and identify differences in the coefficient of friction for different levels of micro-aeration. The test design aims to capture a single sample of chocolate and subject it to repeated shearing. The assumption is that no new material enters the contact during the reciprocating motion and although the chocolate is degraded, a thin layer remains intact on the glass surface without significant loss of the film. At the end of the test, the rubbed film on the glass slide was observed under a low power microscope (Fig. 7a and b). Fig. 7a shows the overall view of the chocolate sample with the rubbed portion in the middle. Fig. 7b shows a DIC optical image from the rubbed region of non-aerated chocolate after the test. The image clearly shows that a macroscopic film remains in the contact and its structure is shear degraded. Immediately after the test, free fat

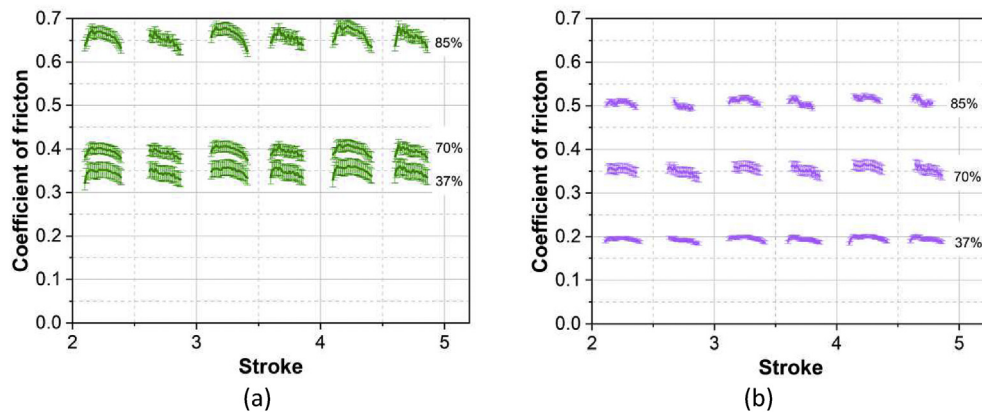


Fig. 5. Measurement of coefficient of friction as a function of reciprocating motion. Comparison for samples with different cocoa solids content (a) without an artificial saliva film present and (b) with an artificial saliva film present.

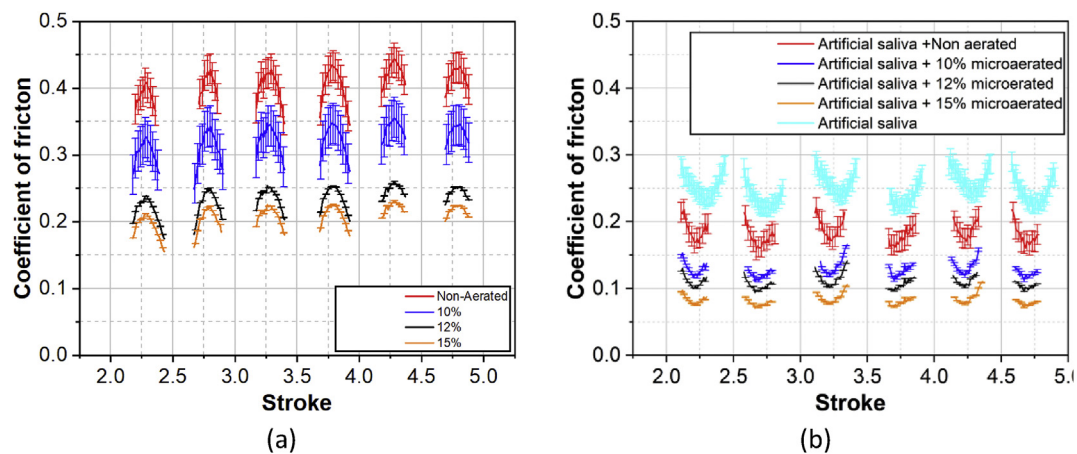


Fig. 6. Coefficient of Friction traces as a function of stroke position for chocolate samples with different aeration levels (a) without an artificial saliva film present and (b) with an artificial saliva film present.

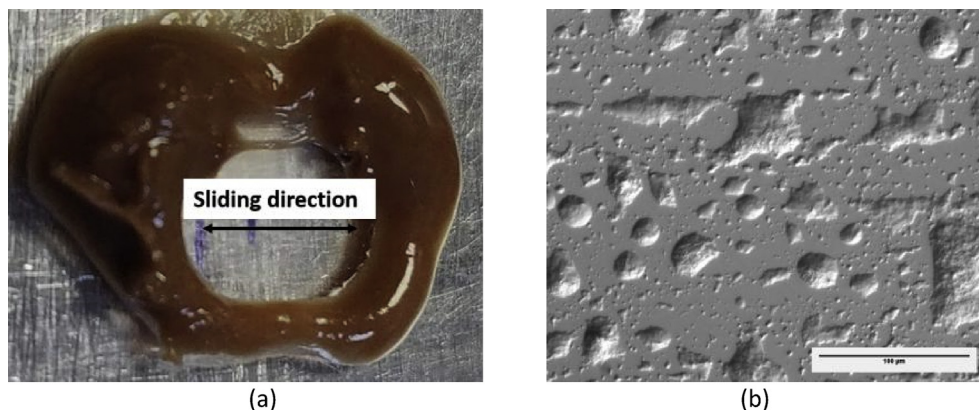


Fig. 7. (a) Sliding direction after the test, (b) microscope image of non-aerated chocolate in the sliding direction film after the test.

droplets are present (these solidify on cooling) and discrete sugar/cocoa particles.

4. Discussion

A tribology test was developed to measure friction of different types of molten chocolate as it mechanically degraded. The test uses a reciprocating flat-on-flat configuration, which simulated the conditions and motion in the tongue-palate contact. In this arrangement the converging

inlet, which is usually present in classical lubrication tests, is absent, thus the chocolate is not pre-sheared before friction is measured. Friction was measured over 10 strokes (5 reciprocating cycles at 1 Hz) to simulate in-mouth degradation over 5 s of mastication. The effect of the composition in terms of cocoa solid content and aeration was studied in a series of tests with and without artificial saliva present.

The presence of artificial saliva decreased the friction coefficient from $\mu = 1.25$ (dry) to $\mu = 0.25$ (artificial saliva) which is similar to values reported in the literature (Andrysewicz et al., 2014). The average

mid-stroke friction coefficient is compared for the commercial samples in Fig. 8. The results are plotted against cocoa, sugar and fat content for tests with and without artificial saliva present.

In all cases the friction coefficient decreased in the presence of the artificial saliva. It is difficult to draw concrete conclusions about the effect of composition on friction as the cocoa, fat and sugar content all change in the samples. However, some general trends may be identified. Friction coefficient *decreased* with *decreasing* cocoa, *decreasing* fat and *increasing* sugar content. Clearly it is difficult to isolate the effect of a single component on friction.

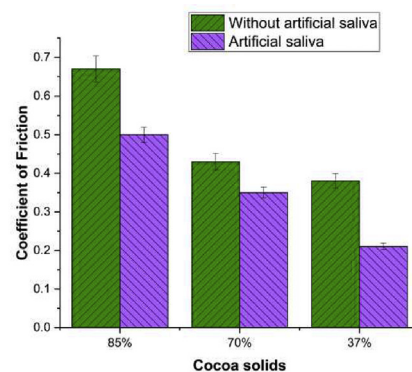
Fig. 9 compares mid-stroke average friction for the aerated samples with and without artificial saliva present. Again, friction reduces in the presence of the artificial saliva. The results for these chocolates are less ambiguous as the fat, sugar and cocoa solid content were all constant for all samples in this group. Friction coefficient *decreases* with *increasing* aeration levels.

For both sets of chocolate samples, non-aerated and aerated, the friction coefficient reduced in the case of artificial saliva being present in the contact. For the commercial chocolate samples, this decrease was in the range of 19–45%, depending on the cocoa-solid content. The reduction was much larger for the aerated samples, where the largest decrease (65%) occurred for the 15 vol% aerated samples.

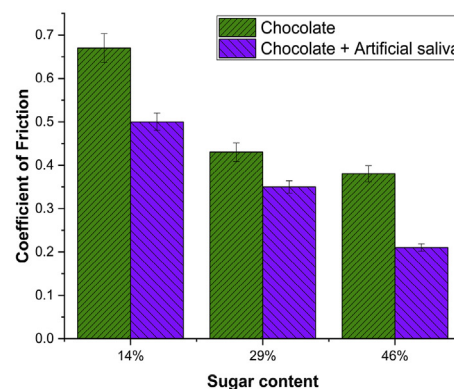
Examination of the rubbed film post-test provides insights into structure degradation and component loss. The DIC image of the rubbed film in Fig. 7b clearly shows the structure of chocolate has degraded and coalescence of fat. The amount of sample left in the track indicates the surfaces are separated by a lubricating film and that direct interaction of the glass and the silicone did not contribute to the measured friction. Chocolate is a complex material and the friction response will be influenced by the original composition, phase changes due to melting and mechanical degradation, and loss of some components during rubbing. The addition of saliva and the possible dissolution of some components, for example sugar, will add to this complexity. To understand these processes, it will be necessary to test simpler, model systems with controlled compositions. Post-test analysis of the chemistry and distribution of the remaining film (Masen and Cann, 2018; Rodrigues et al., 2017) is also necessary.

Several chocolate studies (Lee et al., 2004), (Rodrigues et al., 2017), (Carvalho-Da-Silva et al., 2013), (Mantihal et al., 2019) have used a classical Stribeck-type analysis to explain their tribology results. Friction coefficient is plotted as a function of entrainment speed to identify different lubrication regimes. An idealised example is shown in Fig. 10 where the boundary, mixed and hydrodynamic regimes are clearly delineated. The interpretation of Stribeck curves generated for chocolate have not been so clear cut. In some studies friction has been reported to increase with speed, typically over the range $0.01\text{--}100\text{ mm s}^{-1}$ for $\mu\sim 0.1\text{--}0.6$. (Carvalho-Da-Silva et al., 2013), (Mantihal et al., 2019). However, Rodrigues et al. (2017) reported fairly constant friction coefficient of $\mu\sim 0.08\text{--}0.1$ over for a speed range of $1\text{--}100\text{ mm s}^{-1}$.

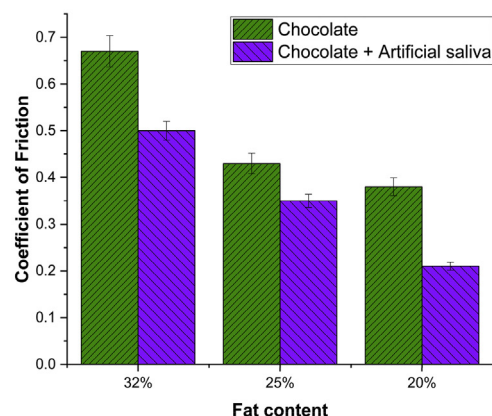
Although we have doubts about this type of analysis, we can use this method to examine the chocolate friction response in the current work. In the BTM test the speed profile of the contact is sinusoidal, and the friction coefficient was measured over the entire stroke length, meaning the speed varied from 0 at the beginning and end of the stroke to 12 mm s^{-1} at the centre. The shape of the friction curve over the stroke differed between the various chocolate samples (commercial and aerated) and with the addition of artificial saliva. The friction coefficient for the commercial chocolate was fairly constant over the stroke, both with and without artificial saliva. However, there was significantly different behaviour for the aerated samples: without artificial saliva the friction coefficient *increased* with speed and the highest friction was recorded in the middle of the stroke. With added artificial saliva the friction coefficient *decreased* with speed and the lowest friction coefficient was in the middle of the stroke. Classical lubrication theory predicts that the thickness of the lubricating film will increase with speed due to increased entrainment of fluid and that the friction response will be described by a



(a)



(b)



(c)

Fig. 8. Average mid-stroke coefficient of friction plotted for different chocolate composition for non-aerated samples with and without artificial saliva present. (a) cocoa solid content (b) sugar content (c) fat content.

Stribeck-type curve. An example of a Stribeck curve, where the coefficient of friction is plotted as a function of the hydrodynamic film building parameter defined as viscosity $\times$ speed/load, is shown in Fig. 10. For low speed and/or low viscosity the hydrodynamic component of the film is reduced, and friction is determined by the thin film chemistry of the contact. Increasing viscosity or speed increases the film thickness and

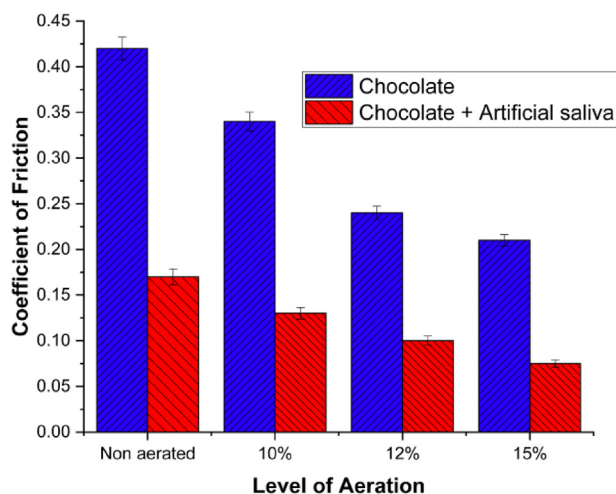


Fig. 9. Comparison of coefficient of friction measurements for different aeration levels with and without an artificial saliva film present.

shifts the friction response to the right-hand side of the diagram, where fluid-shear effects start to dominate.

The results for the chocolate with 10 vol% aeration are superimposed onto the Stribeck curve for two positions in the stroke; (1) near the end of the stroke ($\sim 1 \text{ mm s}^{-1}$) and (2) in the middle ($\sim 12 \text{ mm s}^{-1}$) with and without artificial saliva. Although the applied speed ranges during the saliva and non-saliva experiments are identical, it is reasonable to assume that the addition of a thin artificial saliva layer reduces the chocolate viscosity and thus the friction response shifts towards the left-hand side of the curve. The same friction-speed response was measured for all the aerated samples; however, this was not seen for the commercial chocolate samples chocolate, where the friction coefficient was fairly constant over the speed range. Similar speed-independent friction response has been reported for commercial chocolate samples (Rodrigues et al., 2017). The friction coefficient was higher for the tests without artificial saliva, as expected, since the viscosity of the chocolate is expected to be higher.

Although we have used a Stribeck-type analysis, it must be remembered these curves were developed from friction tests on oils containing low concentrations of soluble additives, which behave quite different than chocolate. The validity of applying these models to complex multiphase fluids with the inherent assumptions of film formation mechanisms and speed dependency is uncertain. This discussion uses a simple lubrication analysis to explain the effect of adding artificial saliva; however, it is likely the mechanism is more complex. For example, by adding an

adsorbed/fluid mucin layer to the contact might also change the position of the shear plane. It is possible that shearing occurs predominantly at the chocolate-mucin interface, which would have a much lower shear stress and thus friction.

The paper has reported a new approach to studying friction of mechanically degraded molten food samples. The advantage of the experimental setup is that it replicates the real timescale that food is rubbed between tongue and palate and that it measures transient friction changes rather than averages of longer timescales. The other improvement is the combination of a large contact area and a short reciprocation stroke, which is a better simulation of the tongue-palate contact and movement. This configuration also means that a single sample of food is captured in the contact and subjected to repeated rubbing rather than the continued entrainment of fresh material as it occurs in most other tribological tests.

The results have shown chocolate composition and structure influences friction. The individual roles of cocoa solids, fat, sugar, emulsifiers etc must be studied in a more methodical and controlled manner through model systems. The contribution of aeration is interesting and promising. Aeration will affect many properties some of which will impact texture attributes for example bite, melting, rheology, mechanical breakdown, chocolate structure/cohesion. At present we do not have enough information to propose a model for these complex processes and the relationship to friction and sensory perception. It requires more detailed research to address it properly and we are currently carrying out rheology, structural breakdown and modelling to address this. However, the results clearly show that aeration can be used to control chocolate friction and thus is a promising method of altering the texture attributes in a controlled manner.

5. Conclusions

This paper describes a novel experimental setup to measure the friction coefficient in reciprocating contact operating at low contact pressure and low speeds. The setup simulates the movement of the tongue and the palate using a combination of a large contact area and a relatively short stroke length. This means food under test is essentially isolated and new material is not continually entrained which is a closer simulation of oral processing. The test was used to distinguish between chocolate samples of different aeration levels and cocoa-solid content. In commercial samples friction coefficient decreased with decreasing cocoa (85–37 wt%), decreasing fat (32–20 wt %) and increasing sugar (14–46 wt%). The friction coefficient decreased with increasing aeration levels over the range of 0–15 vol % aeration. For all chocolate samples the friction coefficient decreased with the presence of an artificial saliva film. In addition, different friction-speed response curves were observed depending on the

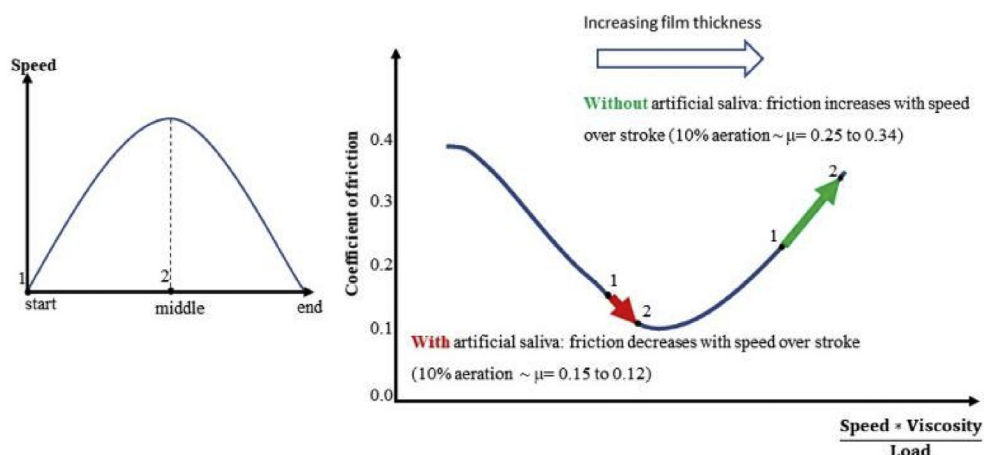


Fig. 10. Idealised Stribeck curve, showing the friction to speed response for 10 vol% aerated chocolate.

nature of the chocolate and the presence of artificial saliva. The commercially available chocolate shows a friction coefficient that is constant over the stroke. For the aerated chocolates the friction coefficient changes with speed. An increase with speed was observed when artificial saliva was not present. A decrease with speed was observed when the artificial saliva was present. These research challenges might be addressed by combining the results from the tribology experiments with rheology measurements with and without artificial saliva present.

Credit author statement

Georgios Samaras, Philippa Cann and Marc Masen conceived the idea, performed the experiments, postprocessed the data and wrote the manuscript.

Dimitrios Bikos, Josélio Vieira, Christoph Hartmann, Maria Charalambides and Yannis Hardalupas: reviewed and commented on the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors would like to acknowledge Imperial College President's PhD scholarship for the financial support. This work was also supported by the Centre for Doctoral Training on Theory and Simulation of Materials at Imperial College London.

References

- Afoakwa, E.O., Paterson, A., Fowler, M., 2007. Factors influencing rheological and textural qualities in chocolate - a review. *Trends Food Sci. Technol.* 18 (6), 290–298. <https://doi.org/10.1016/j.tifs.2007.02.002>.
- Akhtar, M., Stenzel, J., Murray, B.S., Dickinson, E., 2005. Factors affecting the perception of creaminess of oil-in-water emulsions. *Food Hydrocolloids* 19 (3), 521–526. <https://doi.org/10.1016/j.foodhyd.2004.10.017>.
- Alsanei, W.A., Chen, J., 2014. Studies of the oral capabilities in relation to bolus manipulations and the ease of initiating bolus flow. *J. Texture Stud.* 45 (1), 1–12. <https://doi.org/10.1111/jtxs.12041>.
- Andrysewicz, E., Mystkowska, J., Dąbrowski, J.R., Olchowik, R., 2014. Influence of self-made saliva substitutes on tribological characteristics of human enamel. *Acta Bioeng. Biomech. Orig. Pap.* 16, 2. <https://doi.org/10.5277/abb140208>.
- Bongaerts, J.H.H., Fourtouni, K., Stokes, J.R., 2007. Soft-tribology: lubrication in a compliant PDMS-PDMS contact. *Tribol. Int.* 40 (10–12), 1531–1542. <https://doi.org/10.1016/j.triboint.2007.01.007>.
- Breslin, P.A.S., 2013. An evolutionary perspective on food and human taste. *Curr. Biol.* 23 <https://doi.org/10.1016/j.cub.2013.04.010>.
- Carvalho-Da-Silva, A.M., Van Damme, I., Taylor, W., Hort, J., Wolf, B., 2013. Oral processing of two milk chocolate samples. *Food Funct.* 4 (3), 461–469. <https://doi.org/10.1039/c2fo30173c>.
- Chen, J., Engelen, L., 2012. *Food Oral Processing: Fundamentals of Eating and Sensory Perception*.
- Chen, J., Stokes, J.R., 2012. Rheology and tribology: two distinctive regimes of food texture sensation. *Trends Food Sci. Technol.* 25 (1), 4–12. <https://doi.org/10.1016/j.tifs.2011.11.006>.
- Chen, J., Liu, Z., Prakash, S., 2014. Lubrication studies of fluid food using a simple experimental set up. *Food Hydrocolloids* 42, 100–105. <https://doi.org/10.1016/j.foodhyd.2014.01.003>.
- Chojnicka-Paszun, A., De Jongh, H.H.J., 2014. Friction properties of oral surface analogs and their interaction with polysaccharide/MCC particle dispersions. *Food Res. Int.* 62, 1020–1028. <https://doi.org/10.1016/j.foodres.2014.05.028>.
- Cutler, A.N., Morris, E.R., Taylor, L.J., 1983. Oral perception of viscosity in fluid foods and model systems. *J. Texture Stud.* 14 (4), 377–395. <https://doi.org/10.1111/j.1745-4603.1983.tb00357.x>.
- de Wijk, R.A., van Gemert, L.J., Terpstra, M.E.J., Wilkinson, C.L., 2003. Texture of semi-solids; sensory and instrumental measurements on vanilla custard desserts. *Food Qual. Prefer.* 14 (4), 305–317. [https://doi.org/10.1016/S0950-3293\(02\)00107-6](https://doi.org/10.1016/S0950-3293(02)00107-6).
- De Wijk, R.A., Janssen, A.M., Prinz, J.F., 2011. Oral movements and the perception of semi-solid foods. *Physiol. Behav.* 104 (3), 423–428. <https://doi.org/10.1016/j.physbeh.2011.04.037>.
- Devezeaux de Lavergne, M., Stribosch, V.M.G., Van den Broek, A.W.M., Van de Velde, F., Stieger, M., 2016. Uncoupling the impact of fracture properties and composition on sensory perception of emulsion-filled gels. *J. Texture Stud.* 47 (2), 92–111. <https://doi.org/10.1111/jtxs.12164>.
- Dresselhuys, D.M., de Hoog, E.H.A., Cohen Stuart, M.A., Vingerhoeds, M.H., van Aken, G.A., 2008a. The occurrence of in-mouth coalescence of emulsion droplets in relation to perception of fat. *Food Hydrocolloids* 22 (6), 1170–1183. <https://doi.org/10.1016/j.foodhyd.2007.06.013>.
- Dresselhuys, D.M., Stuart, M.A.C., van Aken, G.A., Schipper, R.G., de Hoog, E.H.A., 2008b. “Fat retention at the tongue and the role of saliva: adhesion and spreading of ‘protein-poor’ versus ‘protein-rich’ emulsions. *J. Colloid Interface Sci.* 321 (1), 21–29. <https://doi.org/10.1016/j.jcis.2008.01.051>.
- Hahn Berg, I.C., Rutland, M.W., Arnebrant, T., 2003. Lubricating properties of the initial salivary pellicle - an AFM study. *Biofouling* 19 (6), 365–369. <https://doi.org/10.1080/08927010310001618571>.
- He, Q., Bramante, F., Davies, A., Elleman, C., Fourtouni, K., Wolf, B., 2018. Material properties of ex vivo milk chocolate boluses examined in relation to texture perception. *Food Funct.* 9 (6), 3532–3546. <https://doi.org/10.1039/c8fo00548f>.
- Hiimae, K.M., Palmer, J.B., 2003. Tongue movements in feeding and speech. *Crit. Rev. Oral Biol. Med.* 14 (6), 413–429.
- Humphrey, S.P., Williamson, R.T., 2001. A review of saliva: normal composition, flow, and function. *J. Prosthet. Dent* 85 (2), 162–169. <https://doi.org/10.1067/mp.2001.113778>.
- Ionta, F.Q., et al., 2014. In vitro assessment of artificial saliva formulations on initial enamel erosion remineralization. *J. Dent.* 42 (2), 175–179. <https://doi.org/10.1016/j.jdent.2013.11.009>.
- Janssen, A.M., Terpstra, M.E.J., De Wijk, R.A., Prinz, J.F., 2007. Relations between rheological properties, saliva-induced structure breakdown and sensory texture attributes of custards. *J. Texture Stud.* 38 (1), 42–69. <https://doi.org/10.1111/j.1745-4603.2007.00085.x>.
- Joyner Melito, H.S., Pernell, C.W., Daubert, C.R., 2014. Impact of formulation and saliva on acid milk gel friction behavior. *J. Food Sci.* 79 (5), E867–E880. <https://doi.org/10.1111/1750-3841.12439>.
- Lee, S., Heuberger, M., Rousset, P., Spencer, N.D., 2002. Chocolate at a sliding interface. *J. Food Sci.* 67 (7), 2712–2717. <https://doi.org/10.1111/j.1365-2621.2002.tb08803.x>.
- Lee, S., Heuberger, M., Rousset, P., Spencer, N.D., 2004. A tribological model for chocolate in the mouth: general implications for slurry-lubricated hard/soft sliding counterfaces. *Tribol. Lett.* 16 (3), 239–249. <https://doi.org/10.1023/B:TRIL.0000009735.06341.32>.
- Liu, K., Stieger, M., van der Linden, E., van de Velde, F., 2016. Effect of microparticulated whey protein on sensory properties of liquid and semi-solid model foods. *Food Hydrocolloids* 60, 186–198. <https://doi.org/10.1016/j.foodhyd.2016.03.036>.
- Liu, D., Deng, Y., Sha, L., Abul Hashem, M., Gai, S., 2017. Impact of oral processing on texture attributes and taste perception. *J. Food Sci. Technol.* 54 (8), 2585–2593. <https://doi.org/10.1007/s13197-017-2661-1>.
- Malone, M.E., Appelqvist, I.A.M., Norton, I.T., 2003. Oral behaviour of food hydrocolloids and emulsions. Part 2. Taste and aroma release. *Food Hydrocolloids* 17 (6), 775–784. [https://doi.org/10.1016/S0268-005X\(03\)00098-5](https://doi.org/10.1016/S0268-005X(03)00098-5).
- Mantihal, S., Prakash, S., Godoi, F.C., Bhandari, B., 2019. Effect of additives on thermal, rheological and tribological properties of 3D printed dark chocolate. *Food Res. Int.* 119, 161–169. <https://doi.org/10.1016/j.foodres.2019.01.056>.
- Masen, M., Cann, P.M.E., 2018. Friction measurements with molten chocolate. *Tribol. Lett.* 66 (1), 1–13. <https://doi.org/10.1007/s11249-017-0958-x>.
- Myant, C., Underwood, R., Fan, J., Cann, P.M., 2012. Lubrication of metal-on-metal hip joints: the effect of protein content and load on film formation and wear. *J. Mech. Behav. Biomed. Mater.* 6, 30–40. <https://doi.org/10.1016/j.jmbbm.2011.09.008>.
- Napadow, V.J., Chen, Q., Wedeen, V.J., Gilbert, R.J., 1999. *Intramural Mechanics of the Human Tongue in Association with Physiological Deformations*.
- Nishinari, K., Ishihara, S., Hori, K., Fang, Y., 2020. Tongue-palate squeezing of soft gels in food oral processing. In: *Trends in Food Science and Technology*, vol. 99. Elsevier Ltd, pp. 117–132. <https://doi.org/10.1016/j.tifs.2020.02.023>.
- Payan, Y., Perrier, P., 1997. Synthesis of V-V sequences with a 2D biomechanical tongue model controlled by the equilibrium point hypothesis 1. *Speech Commun.* 22 (2–3), 185–205. [https://doi.org/10.1016/S0167-6393\(97\)00019-8](https://doi.org/10.1016/S0167-6393(97)00019-8).
- Pradal, C., Stokes, J.R., 2016. Oral tribology: bridging the gap between physical measurements and sensory experience. In: *Current Opinion in Food Science*, vol. 9. Elsevier Ltd, pp. 34–41. <https://doi.org/10.1016/j.cofs.2016.04.008>.
- Prakash, S., Dan, D., Tan, Y., Chen, J., 2013. Applications of tribology in studying food oral processing and texture perception. *FRIN* 54, 1627–1635. <https://doi.org/10.1016/j.foodres.2013.10.010>.
- Ranc, H., Elkhyat, A., Servais, C., Mac-Mary, S., Launay, B., Humbert, P., 2006. Friction coefficient and wettability of oral mucosal tissue: changes induced by a salivary layer. *Colloids Surfaces A Physicochem. Eng. Asp.* 276 (1–3), 155–161. <https://doi.org/10.1016/j.colsurfa.2005.10.033>.
- Rayment, S.A., Liu, B., Offner, G.D., Oppenheim, F.G., Troxler, R.F., 2000. Immunouniquantification of human salivary mucins MG1 and MG2 in stimulated whole saliva: factors influencing mucin levels. *J. Dent. Res.* 79 (10), 1765–1772. <https://doi.org/10.1177/00220345000790100601>.
- Rodrigues, S.A., Selway, N., Morgenstern, M.P., Motoi, L., Stokes, J.R., James, B.J., 2017. Lubrication of chocolate during oral processing. *Food Funct.* 8, 533. <https://doi.org/10.1039/c6fo00950f>.
- Rovers, T.A.M., Sala, G., Van der Linden, E., Meinders, M.B.J., 2016. Potential of microbubbles as fat replacer: effect on rheological, tribological and sensorial properties of model food systems. *J. Texture Stud.* 47 (3), 220–230. <https://doi.org/10.1111/jtxs.12175>.
- Sarkar, A., Krop, E.M., 2019. Marrying oral tribology to sensory perception: a systematic review. In: *Current Opinion in Food Science*, vol. 27. Elsevier Ltd, pp. 64–73. <https://doi.org/10.1016/j.cofs.2019.05.007>.

- Sarkar, A., Andablo-Reyes, E., Bryant, M., Dowson, D., Neville, A., 2019a. Lubrication of soft oral surfaces. *Curr. Opin. Colloid Interface Sci.* 39, 61–75. <https://doi.org/10.1016/j.cocis.2019.01.008>.
- Sarkar, A., Xu, F., Lee, S., 2019b. “Human saliva and model saliva at bulk to adsorbed phases – similarities and differences. In: *Advances in Colloid and Interface Science*, vol. 273. Elsevier B.V. <https://doi.org/10.1016/j.cis.2019.102034>
- Scholten, E., 2017. Composite foods: from structure to sensory perception. *Food and Function* 8 (2), 481–497. <https://doi.org/10.1039/c6fo01099g>.
- Shewan, H.M., Pradal, C., Stokes, J.R., 2019. Tribology and its growing use towards the study of food oral processing and sensory perception. *J. Texture Stud.* 12452 <https://doi.org/10.1111/jtxs.12452>.
- Shi, L., Caldwell, K.D., 2000. Mucin adsorption to hydrophobic surfaces. *J. Colloid Interface Sci.* 224 (2), 372–381. <https://doi.org/10.1006/jcis.2000.6724>.
- Stribițaia, E., Krop, E.M., Lewin, R., Holmes, M., Sarkar, A., 2020. Tribology and rheology of bead-layered hydrogels: influence of bead size on sensory perception. *Food Hydrocolloids* 104. <https://doi.org/10.1016/j.foodhyd.2020.105692>.
- Sumarokova, M., et al., 2018. Influencing the adhesion properties and wettability of mucin protein films by variation of the environmental pH. *Sci. Rep.* 8 (1) <https://doi.org/10.1038/s41598-018-28047-z>.
- Upadhyay, R., Chen, J., 2019. “Smoothness as a tactile percept: correlating ‘oral’ tribology with sensory measurements. *Food Hydrocolloids* 87, 38–47. <https://doi.org/10.1016/j.foodhyd.2018.07.036>.
- van Aken, G.A., Vingerhoeds, M.H., de Hoog, E.H.A., 2007. Colloidal Behaviour of Food Emulsions under Oral Conditions, pp. 356–366. <https://doi.org/10.1039/9781847552389-00356>.
- Xu, F., et al., 2020. “A self-assembled binary protein model explains high-performance salivary lubrication from macro to nanoscale. *Adv. Mater. Interfaces* 7 (1), 1901549. <https://doi.org/10.1002/admi.201901549>.